Heavy metal levels in water, sediments and edible fish tissues (*Mugil cephalus*) of Negombo Estuary, Sri Lanka

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Abstract

Industrial pollution and domestic solid waste dumping are considered as the major pollution (or pollutant) sources of the Negombo Estuary. Polluted water and sediments in the estuary closely related to the heavy metal concentration in fish. This could lead to a serious health hazard and/or affect the health of the fish consumers. Therefore, a study was done with the objectives to assess the current levels of heavy metals (Pb, Cd, Hg, Cu, Cr and Zn) in the water, sediments and fish tissues. For this study water, sediment and fish samples were collected from six sampling locations along the North (n=2), South (n=2), West (n=1), and East (n=1) regions during one year study period from January to December 2014 and heavy metals levels were analysed employing standard methods. Heavy metals concentration in water, sediment and fish tissues were comparatively high at the Northern side of the estuary followed by Southern, Eastern and Western side indicating that the pollutant accumulation within the estuary vary spatially and influenced by discharges of industrial untreated effluents, domestic and municipal solid waste and sewage into the estuary.

Keywords: Heavy metal, *Mugil cephalus*, Water, Sediment, Fish tissue

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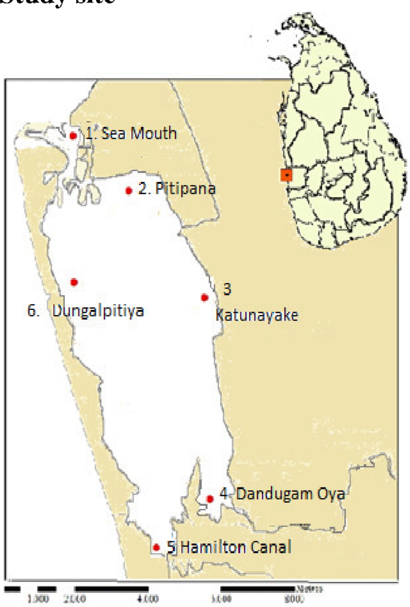
Introduction

The pollution of the aquatic environment with toxic and nontoxic heavy metals has become a worldwide problem during recent years, because most of them have toxic effects on organisms (MacFarlane and Burchett, 2000). Heavy metal concentrations in aquatic ecosystems are usually monitored by measuring their concentrations in water, sediments and biota (Camusso *et al.,* 1995). Sediments are important sinks for various pollutants including heavy metals and also play a significant role in remobilizing them between different media (water, sediments and biota) in aquatic systems under favorable conditions. Namminga and Wilhm (1976) stated that heavy metals generally exist in low concentrations in water and attain considerable concentrations in sediments and biota. Once accumulated in water, metals may remain in solution as free ions or as soluble complexes of organic and inorganic anions. Insoluble complexes with organic particulate or inorganic anions, such as carbonates, precipitate to the sediments. Aquatic sediments can act as both a sink and a source for contaminants, whereby long term input of contaminants can lead to a situation where the concentration of contaminants shall exceed in sediment than water (Burger *et al.,* 2002). Heavy metals including both essential and nonessential elements have a particular significance in ecotoxicology, since they are highly persistent and all have the potential to be toxic to living organisms (Storelli *et al.,* 2005). Fish can be considered as one of the most significant indicators in brackish water systems for the estimation of metal pollution level (Rashed, 2001).The commercial and edible species have been widely investigated in order to check for those hazardous to human health (Begum *et al.,* 2005). Heavy metals such as copper, iron and chromium are essential metals since they play an important role in biological systems, whereas cadmium, mercury and lead are nonessential metals, as they are toxic, even in trace amounts (Fernandes *et al.,* 2008). For the normal metabolism of the fish, the essential metals must be taken up from water, food or sediment (Canil and Atli, 2003). Low level discharge of a contaminant may meet the water quality criteria, but long term partitioning could result in the accumulation of high loads of pollutants (Harikumar and Jisha, 2010).

Water pollution is one of the principal environmental and public health problems in Negombo Estuary (Silva, 1996) and it receives pollutants from various point and nonpoint sources. The major heavy metal source of the estuary is the discharges of the central sewage treatment plant of Katunayake Export Processing Zone (Asanthi *et al.,* 2007). The treated, partially treated or untreated effluents of metal use industries that locate in Katunayake Export Processing Zone discharge their wastewater into the central sewage treatment plant. The other pollutant sources of the estuary include oil released by the large number of boats anchored in the estuarine area, domestic discharges including septic tanks, soak away pits or drainage ditches and fish waste from fishing activities. Munnakkara area also releases large amount of untreated sewage into the estuary. Asanthi *et al.,* (2007) stated that the mean concentrations of most metal ions in water, sediments and aquatic plants were relatively high in Negombo Estuary. The estuary is susceptible to chemical pollution due to ongoing development activities of Negombo and Katunayake areas (CEA, 1994). However monitoring the heavy metal pollution in Negombo Estuary would be important to human health because of the fishery of the estuary (Indrajith *et al*., 2008). The objectives of this study were to assess the current levels of heavy metals (Pb, Cd, Hg, Cu, Cr and Fe) in the water, sediments and fish tissues in Negombo Estuary.

Materials and Methods

Study site



Six sampling locations between latitude 7° 7’-7°12’ N and longitude 79°00’-79°53’ E were selected considering the inputs of industrial pollutants and domestic solid waste dumping in the estuary (Fig. 1). Based on the pollution inputs, it may be considered that different localities within the estuary carry different rates of pollutants. Hence the sampling sites were categorized as North, South, East, and West Estuary (Table 1).

Fig. 1. Sampling locations in Negombo Estuary

Table 1. Descriptions of study sides at Negombo Estuary

Side of the estuary	Sampling locations	Potential Pollutant sources
Northern (Side I)	1 and 2	Municipal solid waste, industrial effluents, fish waste disposal, hotel wastewater, Fishing harbour waste, boatyards and domestic sewage.
Southern (Side II)	3	Ekala industrial zone, seaplane landing site, discharges factory effluent from Dadugam Oya and Hamilton Canal.
Western (Side III)	4	Hotels, shrimp farms and fish processing industries.
Eastern (Side IV)	5 and 6	Katunayake Industrial Processing Zone, housing scheme.

Sample collection and pretreatment

Water samples were collected monthly from six sampling locations, for a period of one year in 2014. The sampling bottles were preconditioned with 5% nitric acid and later rinsed thoroughly with distilled deionized water. At each sampling site, the polyethylene sampling bottles were rinsed at least three times before sampling was done. Pre-cleaned polyethylene sampling bottles were immersed about 10 cm below the water surface. At each sampling site, about 250 ml of water sample was collected. Samples were acidified with 10% Nitric acid and placed in an ice bath during the transportation. The samples were filtered through a 0.45 μm membrane filter paper and stored at 0°C until analysis.

Sediment samples were collected using PVC tube and placed in an ice bath. Fresh fish samples (*Mugil cephalus*) were collected by using cast nets from the same sampling locations and transported to the laboratory in an ice container on the same day. The samples were analysed by the method given in American Public Health Association (APHA, 1998) and Association of Official Analytical Chemists (AOAC, 2002). Atomic Absorption Spectrophotometer (Thermo Elemental Solaar - S_4) was used to measure the heavy metals; Pb, Cu, Cr, Hg, Zn and Cd in water, sediments and fish tissues.

Analysis of sediment samples

Sediment samples were allowed to defrost and air dried in a circulating oven at 30°C and sieved mechanically using a 2 mm sieve. Sediments analysis was carried out according to the procedure described by APHA (1998). For the digestion of samples, one gram (01g) of sieved sediment was digested with repeated addition of nitric acid and 30% hydrogen peroxide. The resultant digested volume was reduced and then diluted to a final volume of 100 ml. Atomic Absorption Spectrophotometer (Thermo Elemental Solaar - S_4) determined the elements of concern (Zn, Cu, Pb, Hg, Cr and Cd) in water and extracted sediment samples.

Analysis of fish tissues

Mugil cephalus samples were collected in monthly intervals and sample preparation and analysis were conducted according to the procedure described in AOAC, 2002. The mean length and weight of the fish were 103.6 ± 12.4 mm and 226.6 ± 32.15g. These tissues were washed with tap water followed by distilled water, oven dried to constant weight at 105^0C. The dried fish was crushed and powdered in an agate mortar and kept in polyethylene bottles for analysis. One gram (01g) portions of fish tissues were digested in nitric acid and hydrogen peroxide. The results were calculated in milligram per kilogram wet weight ($mgkg^{-1}$). Pb, Cd, Hg, Cu, Cr and Zn were tested using AOAC, 2002.

Statistical analysis

Differences in the levels of each metal in water, sediment and fish tissue collected from the estuary were tested by one way analysis of variance (ANOVA).

Results

Metal levels in water

Comparing the average concentrations of heavy metals (Pb, Cd, Hg, Cu, Cr and Zn) in the different study sites, of the estuary are presented in Table 2.

Table 2. Monthly mean heavy metals concentration (in μgL^{-1}) ± SE of water samples 2014

Regions	Pb	Cd	Cu	Cr	Zn	Hg
North	25.1 ± 10.0	2.5 ± 0.1	3.1 ± 0.4	5.0 ± 0.2	460 ± 163.0	1.3 ± 0.01
South	20.1 ± 10.5	1.2 ± 0.1	3.0 ± 0.2	3.0 ± 2.0	248 ± 104.6	ND*
West	5.0 ± 0.12	ND*	2.0 ± 0.4	5.0 ± 0.5	133 ± 20.5	0.004 ± 0.001
East	17.0 ± 0.9	1.2 ± 0.2	2.5 ± 0.2	3.0 ± 2.0	248 ± 14.0	ND*

ND* = Not Detectable

The concentration (in μgL^{-1}) of the metals in water had shown wide spatial variation (see Table 2). Results indicated that the levels of Pb, Cd and Hg were comparatively low at the Western side of the estuary. The levels of these metals in the Southern and Eastern side were similar and lower than the levels in Northern side.

Table 3. Relationship ('p' values) between heavy metal levels in water and fish tissue

Metal	'p' Value
Pb	< 0.05
Cd	< 0.05
Cu	> 0.05
Cr	< 0.05
Hg	< 0.05
Zn	> 0.05

The higher heavy metal concentrations in the estuarine water might be attributed to the direct inputs from different sources (industrial wastes). The result indicates that the levels of nonessential (toxic) heavy metals such as Pb, Cd and Hg in water were comparatively high at the Northern part of the estuary (Table 2). Heavy metal (Pb, Cd, Cr and Hg) concentration of water and fish tissue shows a significantly high ($p < 0.05$) correlation (Table 3).

Heavy metals in sediments

The levels of selected metals in sediments at six sampling locations of Negombo Estuary are shown in Table 4. Metal concentration in sediment ($mgkg^{-1}$) showed irregular spatial distribution, indicating varying pollutant input into the estuary. The high levels of Cd in sediments were found at sampling site 1 located in the Northern side whereas levels of Pb were high in the Eastern side.

Table 4. Monthly mean heavy metals concentration (in $mgkg^{-1}$) ± SE of sediment samples-2014

Regions	Pb	Cd	Hg	Cu	Cr	Zn
North	3.88 ± 0.35	1.06 ± 0.23	0.001 ± 0.25	0.033 ± 2.3	11.4 ± 1.1	523.5 ± 56.3
South	5.90 ± 0.65	0.45 ± 0.05	0.001 ± 0.52	0.025 ± 0.021	9.5 ± 2.5	778.6 ± 101.2
West	3.10 ± 0.23	0.40 ± 0.32	ND*	0.042 ± 0.032	13.1 ± 2.5	538.5 ± 2.32
East	7.95 ± 0.95	0.40 ± 0.25	ND*	0.025 ± 0.25	14.3 ± 1.5	437.5 ± 4.05

ND* = Not Detectable

Table 5. Relationships between heavy metal levels in sediment and fish tissue ('p' values)

Metal	'p'Value
Pb	< 0.05
Cd	> 0.05
Cu	< 0.05
Cr	> 0.05
Hg	< 0.05
Zn	> 0.05

The high level of Pb in sediments was found at Eastern side. However, the high level of Cd and Hg in the sediment was found at the Northern side (Table 4). The high Cu and Zn levels in sediments were found at the Western and Southern side respectively. Mercury was not detected at the Western and Eastern sides of the estuary (Table 4). The relationship between heavy metal Pb, Cu and Hg concentration in sediment and fish tissue was significantly high ($p < 0.05$) (Table 5).

Heavy metal levels in fish tissues

Total heavy metal levels in muscle tissues of the fish (*Mugil cephalus)* caught at six sampling locations are shown in the Table 6.

Table 6. Monthly mean heavy metals concentration (in $mgkg^{-1}$) ± SE of fish tissue- 2014

Regions	Pb	Cd	Hg	Cu	Cr	Zn
North	0.57 ± 0.48	1.04 ± 0.28	0.03 ± 0.018	4.32 ± 1.15	4.27 ± 1.14	31.08 ± 10.5
South	0.30 ± 0.19	0.45 ± 0.28	0.0005 ± 0.0004	4.04 ± 1.13	4.33 ± 1.45	32.8 ± 19.3
West	0.25 ± 0.04	0.55 ± 0.48	0.001 ± 0.0005	3.24 ± 0.56	5.22 ± 2.49	26.49 ± 11.5
East	ND*	0.35 ± 0.16	0.05 ± 0.046	2.35 ± 0.98	4.73 ± 1.34	32.29 ± 5.3

ND* = Not Detectable

The highest levels of Pb, Cd and Cu in fish tissue samples were found at Northern side. In addition, the high Cr and Cd were observed in the fish samples collected from Western side.

Discussion

Water quality of Negombo Estuary is influenced by several industrial discharges into the estuary, including river Dandugam Oya and Hamilton Canal. The inflowing fresh water and the tidal circulation, induced by the sea level difference between sea and estuary may transport and/or redistribute toxic pollutants. The present study reveals that, the levels of heavy metal vary spatially, especially in respect to dissolved Pb, Cu, Cr, Hg, Zn and Cd. The levels of dissolved Pb, Cd and Hg in the water collected from some sampling sites located in the Northern side were higher than the other sides of the estuary. Northern side of the estuary is being polluted due to the various anthropogenic activities such as domestic solid wastes from industries, houses, boatyards and animal farms. Soil erosion within catchment coupled with municipal waste discharges and industrial effluents discharged could be the source of Pb levels observed in this study. The Pb levels observed in north region was not exceeding the standard threshold levels in recommended limits for the discharge of industrial waste water into inland surface water quality standards in Sri Lanka (Government Gazette, 2008). However, the level of sediment Pb at Northern side was comparatively low. It is possibly due to the strong tidal currents at the estuarine mouth, which may not allow the heavy metal to get deposited into the sediments but transported away from the site along the water column. Metals levels in water (Pb, Hg and Cd) were higher in Northern side than that of Southern side. Higher level in the Northern side may be at least partially attributed to enhanced anthropogenic activities, which may result in discharge of industrial untreated effluents, domestic and municipal solid waste and sewage, burned and unburned fuel from motor boats into the estuary. The maximum allowable levels of Pb, Cd and Hg in the fish for human consumption specified by the European Union (EU), (2002).

Conclusions

In conclusion, the levels of metals in edible muscles of fish collected from the Negombo Estuary exceed the food safety limits specified by the international standards. The results showed that, heavy consumption of *Mugil cephalus* from the Negombo Estuary may pose a health risks to the consumers due to accumulation of high levels of metals concentrations.

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