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Quantitative Analysis of Water Quality and Heavy Metals in Water, Sediments, and Tissues of Grey Mullet (*Mugil cephalus*) from the Negombo Estuary

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Abstract

The objectives of this study were to assess the levels of water quality and heavy metals of the water, sediments and fish tissues in the Negombo estuaries. For this study, samples were collected from sixteen sampling locations during the one year study period from January to December 2014. The results revealed that the average concentrations of ammonia, nitrate, pH, electrical conductivity, total suspended solids of 0.215±0.4mg/l, 1.08±0.98mg/l, 7.8±0.28, 29.6±1.56ms/cm, 29.2±0.35mg/l, levels were below the maximum permissible limits. Biochemical Oxygen Demand 24.8±1.3mg/l and Chemical Oxygen Demand 469.0±36.5mg/l levels were much higher than the permissible threshold limits of industrial wastewater quality standards for CEA,(2001) Sri Lanka. The results indicated that the concentrations (ppm) of heavy metals in water were Pb,0.01±0.0003; Cd,0.015±0.003; Hg,0.013±0.001; Zn,0.695±0.06; Cu,0.03±0.02; Cr,0.055±0.004 and Fe,0.485±0.04 respectively. The sediments concentrations (mg/kg) were Pb,7.95±0.95; Cd,1.06±0.23; Hg,0.001±0.52; Cu,0.042±3.2; Cr,14.30±1.5, Zn,154.25±3.2 and Fe,78.6±101.2 respectively. The concentrations (mg/kg) of metals in the fish tissues were Pb,0.57±0.48; Cd,1.045±0.28; Hg,0.05±0.046; Zn,119.2±111.3; Cu,4.60±1.64; Cr,5.22±2.49 and Fe,32.8±59.3 respectively. The highest concentrations of pollution status were recorded in the Northern region of the estuary.

Key Words: Fish Tissue, Heavy Metals, Pollution Status, Sediment, Water Quality

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Introduction

Negombo estuary is becoming heavily polluted due to rapid industrialization and urbanization in the area (Silva 1996). Anthropogenic activities continuously increase the amount of heavy metals added to the environment, especially in aquatic ecosystem, at an alarming rate which has become an important worldwide problem (Malik *et al.*2010). Increase in population, urbanization, industrialization and agricultural practices have further aggravated the situation (Gupta *et al*. 2009). As heavy metals cannot be degraded, they are deposited, assimilated or incorporated in water, sediments and aquatic animals (Al-Khafaji 2010).Water pollution is thus a cosmopolitan problem that needs urgent attention and prevention (Ali *et al*. 1996). It is resulted from many sources, e.g. accidental spillage of chemical wastes, discharge of industrial or sewerage effluents, agricultural drainage, domestic wastewater and gasoline from fishery boots (Handy 1994). Water pollution is one of the principal environmental and public health problems in Negombo estuary in Sri Lanka (Silva 1996). Monitoring determines trends in the quality of the aquatic environment and how the environment is affected by the release of contaminants, by other anthropogenic activities. Sediments are one of the possible media for monitoring of aquatic systems.

Apart from water, sediments are also responsible of nutrients and pollutant transportation in aquatic environment (McCready *et al.* 2006). Fish accumulate toxic chemicals directly from the water and through their diet, contaminant residues may ultimately reach concentrations of hundreds or thousands of times above those measured in the water, sediment and fish. For this reason, monitoring fish tissue contamination serves an important function as an early warning indicator of sediment contamination or related water quality problems (Osman *et al.* 2007). Monitoring of fish tissue contamination also enables us to detect concentrations of toxic chemicals in fish that may be harmful to consumers, and take appropriate action to protect public health and the environment. A combination of bioaccumulation and measurements of water and sediment quality can provide a good indication of conditions and potential risks to the water body. The present study was aimed to study the heavy metal monitoring in water, sediments, and tissues of the mullet species (*Mugil Cephalus*) along the whole course of the Negombo estuary. Heavy metals such as copper and zinc are essential for life whereas some metal including mercury, lead and cadmium are biologically non essential which can be toxic to biota at very low levels. High concentration of some essential trace metals may become toxic at concentration which exceeds the required limits (Wright and Welbourn 2002). The objectives of this study were to assess the levels of water quality and heavy metals of the water, sediments and edible fish tissues.

Methodology

Study Site

Negombo estuary is a shallow basin estuary of approximately 3,164 ha in extent, located between latitude 7'- 7°12' N and longitude 79°-79°53' E in the West coast of Sri Lanka. It is connected to the sea by a single narrow opening, the Negombo channel segment at its northern end, which is kept open year round.

To analyze water quality, sampling locations were selected to represent the entire estuary based on the channels and pollutant inputs from industrial, tourism, domestic, agricultural and municipal sources. Hence, sixteen sampling locations were selected at which the water channels enter into the estuary (Figure 1).

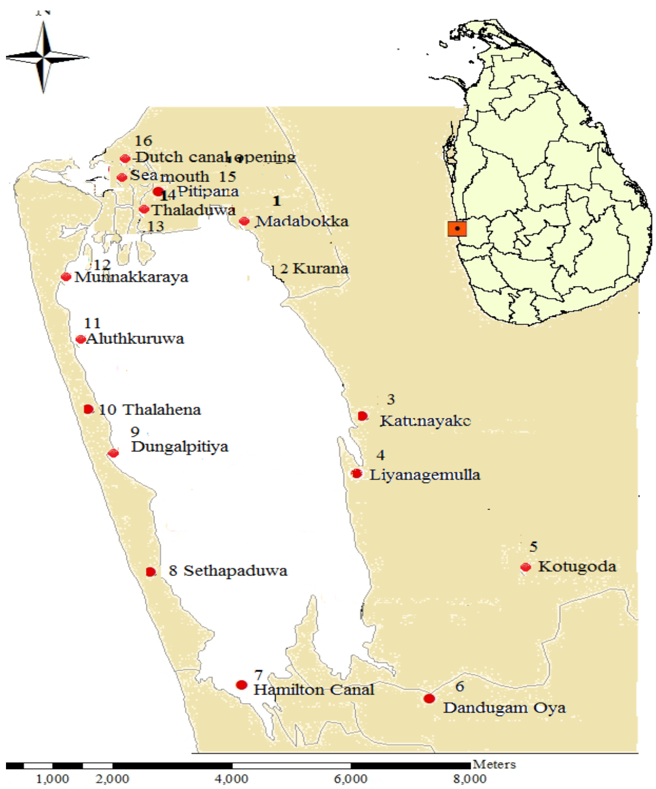

Figure 1: Sampling Locations at Negombo Estuary

These sampling locations included discharge points of effluent inlets from most of the industries at Ja-ela and Katunayeke free trade zones, boat anchoring locations and wastewater discharge points from houses (Table1).Water analysis was mainly focused on physico chemical parameters of water including pH, Electrical Conductivity, Total Suspended Solids, Chemical Oxygen Demand, Biochemical Oxygen Demand, Ammoniacal Nitrogen, Nitrate Nitrogen, and Heavy metals levels as Lead (Pb), Cadmium (Cd), Mercury (Hg),Copper (Cu), Zinc (Zn), Chromium (Cr) and Iron (Fe) in water, sediment and selected edible fish tissues.

Analysis of Water

Water sample were collected monthly from sixteen sampling locations, for a one year period from January 2014 to December 2014. Using polyethylene bottles in ice and transported to the laboratory. Some of the physico chemical parameters such as pH, Electrical Conductivity, Chemical Oxygen Demand, Total Suspended Solids, Biochemical Oxygen Demand, Ammoniacal Nitrogen, and Nitrate Nitrogen, were measured according to the Standard Methods for Examination of Water and Wastewater (20^{st} Edition APHA Standard Methods). Heavy metals in water, sediments and fish tissues Pb, Cu, Cr, Zn, Hg, Fe and Cd were measured by Atomic Absorption Spectrophotometer (Thermo Elemental–Solaar).

Analysis of Sediments

Sediment samples from the selected sites were also collected at monthly intervals during the one year period from January 2014 to December 2014. These were collected using PVC pipe and kept frozen until analyzed. Sediment samples were allowed to defrost and air dried in a circulating oven at 30°C and sieved mechanically using a 02 mm sieve. For the digestion of samples, one gram (01g) sieved sediment was digested with repeated addition of nitric acid and 30% hydrogen peroxide. For Atomic Absorption Spectrometry analysis, the resultant digest was reduced in volume and then diluted to a final volume of 100 ml. The elements of concern (Fe, Zn, Cu, Pb, Hg, Cr and Cd) in the samples were determined by Atomic Absorption Spectrophotometer (Thermo Elemental–Solaar).

Analysis of fish tissues

Fresh fish samples (*Mugil cephalus*) were collected by using cast nets from the same sampling locations during experiment period from January 2014 to December 2014. These tissues were washed with tap water followed by distilled water, oven dried to constant weight at 105 ^{0}C. 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 by means of a microwave digestion after addition of nitric acid and hydrogen peroxide. The results were calculated in milligram per kilogram wet weight (mg/kg wet wt). Pb, Cd, Hg, Cu, Zn, Cr and Fe were tested using AOAC (2002). Hg in the digested fish sample was analyzed using cold vapor Atomic Absorption Spectrophotometer (Thermo Elemental–Solaar).

Sampling sites were categorized as North, South, East, and West estuary (Table 1).

Table 1: Description of Study Sites at Negombo Estuary

Region of the estuary Categorize	**Sampling locations Discharge inlets**	**Input sources (if any)**
Northern Estuary **(A)**	13,14,15,16	Municipal solid waste, industrial effluents, animal livestock urbanization, hotels, fishing harbor and boat repair stations and domestic sewage outlets.
Southern Estuary **(B)**	6,7,8	Two fresh water canals, Ekala industrial zone, seaplane landing site, various effluents in Hamilton canal outlet.
Western Estuary **(C)**	9,10,11,12	Hotels, shrimp farm and fish processing industries.
Eastern Estuary **(D)**	1,2,3,4	Katunayake industrial processing zone, hotels and housing scheme.
Reference Point **(R)**	5	Ekala industrial zone (near 01 km distance)

Results

Water Analysis

Results are presented as mean ± standard error values of physical chemical parameters in Negombo estuary during a study period are given in Table 2.

Table 2: Mean ± SE of annual variations of physico chemical parameters (n=16)

Region of the estuary	pH	EC (ms/cm)	BOD (mg/l)	COD (mg/l)	TSS (mg/l)	Nitrate N (mg/l)	Ammonia (mg/l)
North Region	7.82±0.28	29.6±1.56	24.8±1.3	469.0±29.0	24.9±0.2	0.71±0.63	0.14±0.01
South Region	6.70±0.01	13.9±0.45	22.8±2.7	161.5±12.7	11.2±0.54	1.08±0.98	0.20±0.02
West Region	7.60±0.27	24.5±2.45	23.5±2.6	361.1±24.5	29.2±0.35	0.66±0.56	0.09±0.01
East Region	7.45±0.32	20.0±1.89	21.9±2.5	133.0±11.8	13.5±0.25	0.90±0.89	0.135±0.4
Reference Point	7.45±0.34	22.1±2.67	20.7±2.2	229.7±23.5	18.0±0.56	0.95±0.78	0.215±0.4

Source: Survey 2014

The pH of surface water fluctuated between 6.70±0.01 to 7.82±0.28 respectively. The results of this study indicated a high pH value recorded in North region (Table 2). It was within the standard limits of the proposed tolerance limits for the discharge of industrial wastewater quality standards for Central Environmental Authority in Sri Lanka. (CEA, 2001). Electrical Conductivity of surface water varied from 13.9±0.45to 29.6±1.56 ms/cm among the regions. The EC values also highest at north region and the lowest was reported southern region (Table 2). Biochemical Oxygen Demand depends on temperature, extent of biochemical activities, concentration of organic matter and such other related factors. During the study period, BOD was observed to be in the range from 20.7±2.2 to 24.8±1.3 mg/l among sites. Maximum value of BOD was recorded at north region and the minimum was observed at reference point. The Chemical Oxygen Demand is used as a measure of oxygen equivalent of the organic matter content of a sample that is susceptible to oxidation by strong chemical oxidants. The COD levels shown that Table 2, was highest at north region. This may be due to the discharge of industrial effluents into the estuary by some untreated effluents release by factories in these areas, in addition to the discharge of municipal wastewater and other wastes into the estuary.Nitrate and Ammonia are the most common forms of nitrogen in aquatic systems. The high value nitrate was recorded in southern region (Table 2).

Heavy metal levels in water

The levels of seven heavy metals in water (Pb, Cd, Hg, Cu, Zn, Cr and Fe) at sixteen sampling locations of the estuary are presented in Table 3.

Table 3: Mean concentrations ± SE of annual variations of metals in water (n=16)

Regions	Pb (ppm)	Cd(ppm)	Zn (ppm)	Cu (ppm)	Cr (ppm)	Fe(ppm)	Hg(ppm)
North	0.026±0.002	0.015±0.003	0.360±0.05	0.03±0.04	0.039±0.05	0.462±0.03	0.013±0.001
South	0.023±0.001	0.001±0.002	0.695±0.06	0.03±0.02	0.003±0.003	0.485±0.04	0.001±0.001
West	0.005±0.003	ND	0.165±0.007	0.035±0.04	0.055±0.004	0.335±0.02	0.004±0.003
East	0.017±0.09	0.001±0.002	0.021±0.003	0.025±0.02	0.007±0.002	0.480±0.04	ND
Reference Point	0.01±0.0003	0.0005±0.008	0.290±0.02	0.022±0.04	0.023±0.006	0.445±0.05	ND

ND = Not Detectable

The concentration (ppm) of the metals in water showed a lead (Pb) values were fluctuated within a narrow range 0.01±0.0003 to 0.005±0.003 ppm respectively. The high Pb values were recorded in north region (Table 3). The Cd exhibited a wide range of variation between 0.0 to 0.015±0.003 ppm. The high Cd value was recorded at north region (Table 3). Zinc concentrations fluctuated between 0.021± 0.02 to 0.695± 0.06 ppm and Cu concentration ranged from 0.025 ± 0.02to 0.03±0.02 ppm (Table 3). The highest concentration of Cr was recorded at west region lowest was recorded at south region (Table 3). Mercury concentration seems to be very rare in the estuary water as it was recorded only in some regions along the estuary. Generally the increase in heavy metal concentrations in the estuary water might be attributed to the direct inputs from different sources (industrial wastes). The result indicate that the levels of non essential (toxic) heavy metals as Pb, Cd and Hg in water were comparatively high in north region of the estuary (Table 3). The limit of detection of water in Lead, Copper, Cadmium, Mercury, Zinc, Iron and Chromium contents in water were below the standard limits defined for each element are 0.5 mg/l, 0.5 mg/l, 0.2 mg/l, 10 mg/l, 40 mg/l, 20 mg/l and 10 mg/l respectively (EU,2002).

Sediments Analysis

Analysis of sediments existing at the bottom of the brackish water provides evidence for long term pollution. Low level discharge of a contaminant may meet the water quality criteria, but long term partitioning to the sediments could result in the accumulation of high loads of pollutants. Therefore the determination of heavy metals in sediments is fundamental to realize the toxic pollutants in the estuary sediment. Table 4 illustrates the levels of selected metals in sediments at sixteen sampling location of Negombo estuary.

Table 4: Mean concentrations ± SE of annual variation of metals in Sediment (n=16)

Regions	Pb (mg/kg)	Cd (mg/kg)	Hg (mg/kg)	Zn (mg/kg)	Cu (mg/kg)	Cr (mg/kg)	Fe (mg/kg)
North	3.88±0.35	1.06±0.23	0.001±0.25	142.6±0.54	0.033±2.3	11.40±1.1	523.5±56.3
South	5.90±0.65	0.45±0.05	0.001±0.52	152.5±0.52	0.025±0.2	9.55±2.5	778.6±101.2
West	3.10±0.23	0.40±0.52	ND	154.25±3.2	0.042±3.2	13.15±2.5	538.5±2.32
East	7.95±0.95	0.40±0.65	ND	50.75±2.3	0.025±0.25	14.30±1.5	437.5±4.050
Reference Point	4.05±0.32	0.45±0.56	ND	1.50±0.2	0.025±3.2	9.85±3.2	388.0±36.0

ND = Not Detectable

The range of the concentrations (mg/kg) of all metals in sediments were Pb, 3.10±0.23 to 7.95±0.95; Cd,0.40±0.52 to 1.06±0.23; Hg,0 to 0.001±0.52; Cu,0.025±0.2 to 0.042±3.2; Cr,9.55±2.5 to 14.30±1.5; Cr,9.55±2.5 to 14.30±1.5and Fe,388.0±36.0 to 778.6±101.2 respectively. The highest level of Pb in sediments was found in east region (Table 4). The highest level of Cd and Hg were in the sediment was found at the north region (Table 4). Whereas levels of Cr was high in the east region located in the west region. Cu and Fe levels were the highest in the sediments from west and south region respectively. Mercury was recorded in not detected levels in west and east region (Table 4). North and

South region concentrations seem to be constant level. The wide ranges of metal concentrations recorded form locations may be attributed to variations and heavily flow of urban effluents draining into estuary.

Heavy metal levels in fish

Total metal levels in tissue of the fish (*Mugil cephalus)* caught at sixteen sampling location. Knowledge of heavy metal concentrations in fish is important with respect to human consumption of fish. In the estuary, fish are often at the top of the food chain and have the tendency to concentrate heavy metals from water.

Table 5: Mean concentrations ± SE of annual variations of metals in Fish tissue (n=16)

Regions	Pb (mg/kg)	Cd (mg/kg)	Hg (mg/kg)	Zn (mg/kg)	Cu (mg/kg)	Cr (mg/kg)	Fe (mg/kg)
North	0.57±0.48	1.045±0.28	0.03±0.018	51.43±10.11	4.32±1.15	4.27±1.14	31.08±60.5
South	0.30±0.19	0.45±0.28	0.0005±0.0004	119.2±111.3	4.04±1.13	4.33±1.45	32.8±59.3
West	0.25±0.04	0.55±0.48	0.001±0.0005	31.4±60.6	3.24±0.56	5.22±2.49	26.49±31.5
East	0.0±0.01	0.35±0.16	0.05±0.046	37.2±38.9	2.35±0.98	4.73±1.34	32.29±50.3
Reference Point	0.50±0.59	0.36±0.35	0.01±0.005	52.18±110.4	4.60±1.64	3.90±1.29	32.84±35.4

Source: Survey 2014

The concentrations of metals in the edible muscle tissues were Pb,0.0±0.01 to 0.57±0.48;Cd,0.35± 0.16 to 1.045±0.28; Hg,0.0005±0.0004 to 0.05±0.046; Zn, 31.4±60.6 to 119.2±111.3; Cu,2.35± 0.98 to 4.60 ±1.64; Cr,3.90±1.29 to 5.22±2.49 and Fe,26.4±31.5 to 32.8±59.3 mg/kg respectively. The highest level of Pb, Cd and Cu were in the fish tissue were found at the sampling at north region. Whereas levels of Cr was high in the west region. Zn and Fe levels were the highest in south region (Table 5).The detected mercury, copper, zinc, cadmium, lead and iron contents in fish tissue were below the standard limits defined for each element <0.05 mg/kg, <10 mg/kg, <40 mg/kg, 0.5 mg/kg, <0.5 mg/kg, <0.5 mg/kg and <40 mg/kg respectively (FAO, 1983).

Discussion

Water quality of Negombo estuary is influenced by two main inland water bodies (Dandugam Oya and Hamilton Canal) and the sea. The fresh water canal and the tidal floods may transport toxic pollutants originate in different parts of the catchments, land use or the estuary itself. Estuary water is not utilized for human consumption because of its salty nature. 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). Metals may be enter the resident fish in different ways and absorb metals could accumulate in various organs of the fish body. Metal uptake and their toxicity in aquatic fauna is influenced by many factors such as Chemical Oxygen Demand, Biochemical Oxygen Demand and Nutrients levels in water. Copper can combine with other contaminants such as ammonia, mercury and zinc to produce an additive toxic effect on fish. Urbanization and sewage disposal are major source of zinc pollution.

In the present study revealed that, levels of heavy metal variations exist in different regions of the estuary, with respect to the dissolved Pb, Cu, Cr, Zn, Hg, Fe and Cd. The levels of Pb, Cd and Hg in water collected from some sampling sites located in the North region were higher compared to the other regions. North region of the estuary is being polluted due to the various anthropogenic activities such as solid waste from industries, slaughter houses, boat yards and animal farms. High levels of Pb in water samples collected from north in comparison to other sampling regions may be partly due to the discharge of fuel from motor boats to the estuary water. North region of the estuary is connected to the sea mouth and mixing to sea water, through the levels of Pb, Cd and Hg in water in all sampling locations in this region were higher than other regions. The levels of sediment Pb north region were comparatively low. The status of the sediment of this area is affected by tidal waves, sedimentation and human activities. Hence the level of metals in the sediments of north region to change throughout the year and this may have partly contributed to low Pb accumulation in sediment of this area in comparison to the other sites. The south region receives water mainly from the Jaela canal and Dandugam oya and they carry various effluents from Ekala industrial zone (CEA 1994). In comparison to the north region of the estuary the levels of metals in water (Pb, Hg, and Cd) were high but levels of metals in sediments (Pb, Hg, and Cd) were higher in east region receives waste from hotels, shrimp farm and fish processing industries. Generally the levels of Pb, Cd and Hg in water of this area were low. West region receives waste from hotels, shrimp farm and fish processing industries. Generally the levels of Pb, Cd, Zn and Fe in water of this area were low but the levels of sediment bound metal were comparatively high. East region receives effluent from Katunayake industrial processing zone, hotel and housing scheme. Hence the levels of metals in water Pb, Cd and Fe were high in this region.

Conclusions

Results indicated that the levels of BOD, COD and heavy metals revealed that high pollution takes place in industrial and domestic sewage sampling locations. The South region receives water mainly from Dandugam Oya which carries various effluents from Ekala industrial zone. East region receives discharge of effluents from mainly Katunayake industrial processing zone. North region of the estuary, the levels of BOD, COD and heavy metals in water, sediment and fish tissues were higher due to the discharge of industrial effluents and domestic solid waste discharge into the estuary. The detected average concentration of mercury in water was above the standard limits. The highest concentrations of BOD, COD and heavy metals were recorded in the Northern area followed by Southern, Eastern and Western regions indicating the pollution status of the estuarine water.

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