SEUSL

Sri Lankan Journal of Technology
Research

SLJoT
2021 2(01) 30-38
ISSN 2773-6970

Assessment of current water pollution status and accumulation of trace metals in *Mugil cephalus* from Negombo estuary

B.R.C Mendis[1*], M.M.M. Najim[2,3] and H.M.P. Kithsiri[3]

[1]National Aquatic Resources Research and Development Agency, Colombo 15, Sri Lanka
[2]Department of Zoology and Environmental Management, University of Kelaniya, Sri Lanka
[3]Department of Biosystems Technology, Faculty of Technology, South Eastern University of Sri Lanka. Oluvil, Sri Lanka
[*]Corresponding Author: chani004@yahoo.com

Received: 01-12-2020 * Accepted: 12-04-2021 * Published Online: 30-06-2021

***Abstract*—**Industrial and domestic solid waste dumping are the major pollution sources polluting the Negombo estuary. Polluted water and sediments in the estuary may cause heavy metals accumulation in fish. Therefore, this study was aimed to assess physico - chemical parameters in water, heavy metals (Zn, Pb, Hg, Cd, Cu and Cr) in water and fish tissues, to compare the levels of metals in fish tissues with the available maximum recommended standards. The study was carried out from January to December 2016 and monthly sampling was done from the selected sampling sites. Physico - chemical parameters and metal levels were detected through standard methods. Water quality results revealed that temperature, salinity, pH, EC, TSS, BOD and nutrient levels were below the maximum permissible limits and COD levels were higher than the discharge of industrial wastewater quality standards of the CEA, Sri Lanka. According to the results, metal levels in water of the estuary were below the tolerance limits for the wastewater standards of the CEA except for Hg. In the Northern region of the estuary, higher Pb, Cu and Cr levels were reported from the fish species collected. As the "X-Press Pearl" shipwreck has posed a serious threat to the Negombo estuary, this study can be considered as a baseline for the future studies on water and sediment pollution and bioaccumulation of heavy metals in edible fish species from the estuary.

***Keywords*—**Heavy metals, Negombo estuary, Physico - chemical parameters, Standard methods

I. Introduction

Fish is considered as one of the vital sources of food for millions of people worldwide. Sri Lanka faces environmental problems mostly due to increased human population, rapid urbanization and industrialization. Today, Negombo estuary faces many environmental problems due to the changes in the catchment and by an increased external demand on the estuary and wetland for other purposes related to expansion of urban and industrial areas. Land based pollution sources such as treated and untreated industrial effluents, agricultural runoff, domestic and municipal effluents are the main sources of water pollution that enter into the Negombo estuary (Indrajith *et al*., 2008). Sewage from low-income settlements and effluents from metal finishing and processing industries are also discharged into the estuary. Since the estuary is located in an urban area, it receives industrial wastes, human wastes and automobile wastes leading to possible risks of contamination aquatic life with heavy metals. Heavy metals can be toxic at very low levels and ultimately affect the human health due to consumption of contaminated fish. It is reported that some pollutants have affected some food fishes such as *A. commersoni* and *Etroplus suratensis* collected from the Negombo estuary (Indrajith *et al*., 2008). This study aimed to assess physico-chemical parameters in water, heavy metals (Zn, Pb, Hg, Cd, Cu and Cr) in water and fish tissues, and to compare the levels of metals in fish tissues with the available maximum recommended standards.

II. Material and Methods

A. Study Area

Negombo estuary with 3,164 is a shallow basin located between latitude 7' - 7°12' N and longitude 79° 79°53' E in the West coast of Sri Lanka (Hettiarachchi and Samarawickrama, 2003). Water and fish samples were collected from seven selected locations (Figure 1) representing to the entire estuary;

B. Sampling site

In 2016 seven sampling sites, representing various pollution inputs, were selected for this study. The geographical locations of each selected sampling sites within the estuary is shown in Table I. Sampling site 1 (Figure 2), site 2 (Figure 3), site 3 (Figure 4) and site 4 (Figure 5) were located in the Northern region (Sea mouth, Pitipana, Duwa

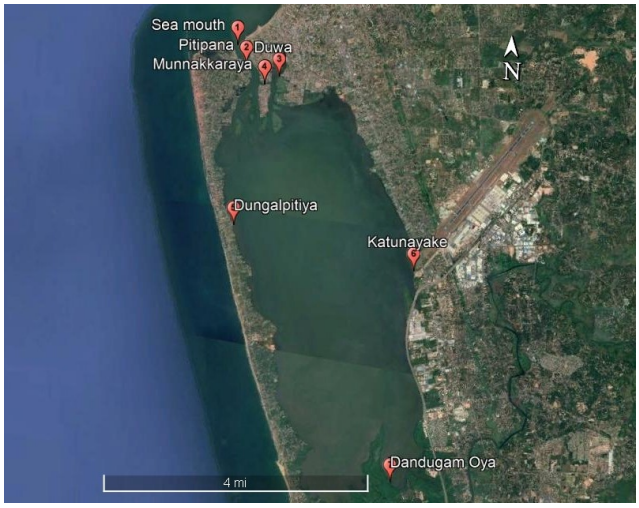

Figure 1: Sampling locations in Negombo Estuary

and Munnakkaraya) of the estuary. This is the area of the estuary that receives more channel inputs. Northern region of the estuary is polluted due to various anthropogenic activities such as solid waste dumping, wastewater discharge from industries, wastes and wastewater from slaughter houses, shrimp farm effluenyts and wastes from boat yards. This is an area where the estuary is directly receiving the effluent from the hospital as well as urban runoff.

Table I: Geographical coordinates of sampling sites in the Negombo estuary

Estuary Region	Sample sites	GPS Location (N)	GPS Location (E)
1 - (North)	Sea Mouth	N 07° 12′ 27.67″	E 079°49′ 38.26″
2 - (North)	Pitipana	N 07° 12′ 12.12″	E 079° 49′ 45.73″
3 - (North)	Duwa	N 07°12′ 03.92″	E 079° 50′ 12.11″
4 - (North)	Munnakkaraya	N 07° 11′ 57.59″	E 079°50′ 00.87″
5 - (West)	Dungalpitiya	N 07° 10′ 05.52″	E 079°49′ 40.67″
6 - (East)	Katunayake	N 07° 09′ 34.87″	E 079°52′ 48.20″
7 - (South)	DandugamOya	N 07° 06′ 48.00″	E 079°51′ 52.69″

Wastes and wastewater from hotels, shrimp farms and dry fish processing factories are discharged to sampling site 5 (Dungalpitiya) in the West region (Figure 6), effluent from mainly Katunayake Export Processing Zone, hotels, housing scheme and airport drainage are discharged to sampling site 6 (Katunayake) in the East region (Figure 7). Factories such as textile, ceramic and electrical equipment are also releasing effluents into the estuary. Sampling site 7, (Dandugam Oya) in the South region (Figure 8) mainly receives water from the Hamilton canal and Dandugam Oya and they carry various effluents from Ekala Industrial Zone.

C. Collection of samples

Water and fish samples were collected using fiberglass motor boats (FRP) in the demarcated sampling sites using

Figure 2: Sampling site 1(Sea Mouth)

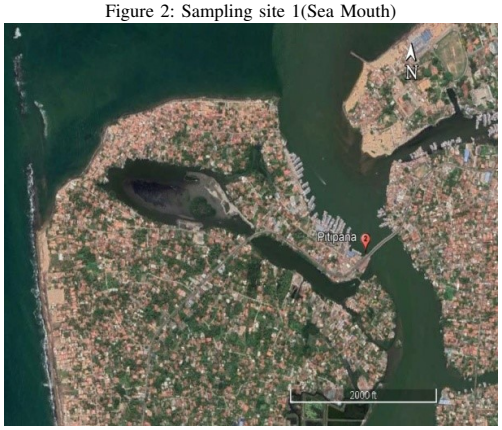

Figure 3: Sampling site 2 (Pitipana)

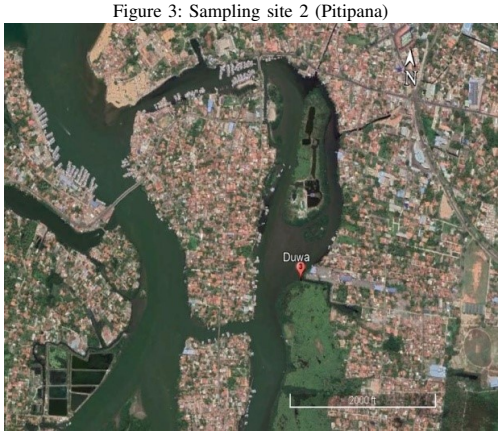

Figure 4: Sampling site 3 (Duwa)

Figure 5: Sampling site 4 (Munnakkaraya)

Figure 6: Sampling site 5 (Dungalpitiya)

Figure 7: Sampling site 6 (Katunayake)

Figure 8: Sampling site 7(Dandugam Oya)

GPS to identify the locations. Water samples were collected to polyethylene bottles that were preconditioned with 05% nitric acid. The samples were filtered through a 0.45 μm micro pore membrane filter paper and the filtrate was kept at 0°C until the analysis is performed. Fish samples were collected by using cast nets from the same sampling locations and transported on the same day to the laboratory in an ice container. Water and fish samples were analyzed using standard procedures given in American Public Health Association (APHA, 2012) and (AOAC, 2002). Atomic Absorption Spectrophotometer (Thermo Elemental Solaar - S_4) was used to measure heavy metals in water and fish tissues.

D. In - situ analysis of water

The status of pollution of the Negombo estuary and some of its tributaries were investigated in 2016 with respect to temperature, pH, salinity, electrical conductivity and Dissolved Oxygen. The following physico - chemical variables of water were determined in situ at the seven sampling sites using the instruments indicated in parenthesis. Instruments and methods used are listed in Table II.

Table II: Instruments used to measure water quality parameters

Parameter	**Methodology**	**Instruments/Method**
Water Temperature	Thermometric	Glass - Mercury thermometer in °C, 2550 B (APHA, 2012)
Salinity	Refractive index	Digital Salinometer, 2520 (APHA,2012)
Electrical Conductivity	Electrical Conductometric	HANNA HI - 8633N Multi - range Conductivity Meter
pH	Photentiometric	Eutech Cyberscan 600 pH/mV Meter
Dissolved Oxygen	Electrometric	YSI Pro DO Optical Dissolved Oxygen Meter range: 0 to 50 mgL^{-1}

Sources: (APHA, 2012)

E. Ex-situ analysis of water

Sample collection and analysis was done considering the sampling sites. Water samples were transported in ice containers to the laboratory and stored in refrigerators at less than 4.0°C. Physico-chemical parameters Total Suspended Solids (TSS), Ammoniacal-N, Ortho-phosphate, Nitrate-N, nitrite-N, Biochemical Oxygen Demand (BOD), and Chemical Oxygen Demand (COD) of water were measured according to standard methods (Table III). Heavy metals Zinc (Zn), Lead (Pb), Mercury (Hg), Cadmium (Cd), Chromium (Cr) and Copper (Cu) were also analyzed through standard procedures (Table IV). All the water quality tests were performed according to Standard Methods for the Examination of Water and Waste Water (22^{nd} Edition) (APHA, 2012).

F. Fish tissues for metal analysis

Twenty-five grams of muscle was acquired by pooling fish tissue samples that were removed using a plastic knife. Weight of each sample was recorded and the sample was kept in a drying oven at 100°C for more than 24 hours. The dried samples were powdered using a porcelain motor and pestle to obtain fine particles. Powdered fish tissues were digested (AOAC, 2002). Then the solution was transferred into plastic tubes for analysis using flame/cold graphite Atomic Absorption Spectrophotometry (Solaar - Thermal Elemental - S_4) (Table V).

Table III: Laboratory analysis and test methods

Parameter	Methodology	Method
Ammonical - nitrogen	Spectrophotometric	4500 -NH_3 F, Phenate method(APHA,2012)
Nitrate- nitrogen	Spectrophotometric	4500 -NO_3^- E, Cadmium reduction method (APHA,2012)
Nitrite - nitrogen	Spectrophotometric	4500 B, Colorimetric method NED/Sulphanilamide (APHA, 2012)
Orthophosphate	Spectrophotometric	4500 - PE Ascorbic acid method (APHA2012)
Bio-chemical Oxygen Demand (BOD)	Titrimetric	5210 B, 5 - days BOD Test (APHA, 2012)
Chemical Oxygen Demand (COD)	Titrimetric	5220 B, Open reflux method (APHA, 2012)
Total Suspended Solid (TSS)	Gravimetric	2540 C, Total Suspended Solids dried at 103 - 105 ˚C, (APHA, 2012)

Sources: (APHA, 2012)

Table IV: Methods of water heavy metal analysis

Parameter	Principle	Method
Copper (Cu)	Atomic Absorption Spectrophotometry (AAS)	3113B Cu, (APHA, 2012)
Lead (Pb)	(AAS)	3113B Pb, (APHA, 2012)
Zinc (Zn)	(AAS)	3113B Zn, (APHA, 2012)
Chromium (Cr)	(AAS)	3113B Cr, (APHA, 2012)
Cadmium (Cd)	(AAS)	3113B Cd, (APHA, 2012)
Mercury (Hg)	Cold vapor (AAS)	3112B Hg, (APHA, 2012)

Sources: (APHA, 2012)

G. Statistical Analysis

Data analysis was done using SPSS 21.0 statistical software package. Differences in level of each metal in water and fish samples were tested by One way Analysis of Variance (ANOVA). One way ANOVA was performed to determine whether statistically significant differences prevail in the mean values of metal concentrations and physico - chemical parameters between each sampling sites. Differences in mean values were accepted as being statistically significant if $P < 0.05$ compared by Tukey's test. Significant differences between major clusters in the resulting dendrogram were analyzed using one-way ANOSIM.

III. Results

Physico-chemical parameters of the bottom - overlying surface water at seven sampling sites shown in Table VI with the statistical analysis of variance (One - way ANOVA) of each parameter. Most of the physico-chemical parameters indicated a significant variation between the sampling sites ($p < 0.05$, at 95% level of significance), except for Electrical Conductivity (EC), pH, Ammoniacal - nitrogen, Nitrite - nitrogen and Orthophosphate ($p > 0.05$) in accordance with the results of One - way ANOVA.

Table V: Methods of fish analysis

Parameter	Principle	Method
Copper (Cu)	Atomic Absorption Spectrophotometer (AAS)	AOAC 999.11:2000 Cu, (AOAC, 2012)
Zinc (Zn)	(AAS)	AOAC 999.11:2000 Zn, (AOAC, 2012)
Chromium (Cr)	(AAS)	AOAC 999.11:2000 Cr, (AOAC, 2012)
Cadmium (Cd)	(AAS)	CML/MM/Fish 002/v 1.3 Cd, (AOAC, 2012)
Mercury (Hg)	Cold vapour (AAS)	Hg, (AOAC, 2012)

Sources: (AOAC, 2012)

Throughout the study period, the lowest monthly mean water temperature (29.50 ± 0.40 ºC) was recorded from site 7, while the highest value (30.7 ± 0.19 ºC) was observed at site 3 (Duwa) (Table VI). One - way analysis of variance (ANOVA) showed a significant difference in surface water temperature between sites ($p < 0.05$). Tukey's test for separation of means revealed that mean water temperature varied significantly from mean water temperature observed at sites 1 to 5 and site 6 (Table VI). According to these parameters, highest value of Dissolved Oxygen (DO) was recorded at site 3 (10.8 ± 2.80mgL^{-1}) while site 7 (Dandugam Oya) denoted the minimum concentration of 3.90 ± 1.10 mgL^{-1}. The pH values of the seven sampling sites showed a wide fluctuation, ranging from 7.20 ± 0.22 to 8.30 ± 0.36 (Table VI). The pH values of the estuarine water at all the sites remained slightly alkaline, whereby site 7 (Dandugam Oya) reported the lowest mean value of 7.20 ± 0.22. The pH values observed at all the sampling sites ranged (7.20 ± 0.22 to 8.30 ± 0.36) within the standard limits (5.5 - 9.0) as recommended permissible threshold limits for the discharge of industrial wastewater standards by the CEA (2007). The highest average EC value of 26.7 ± 6.3 $mScm^{-1}$ at the site 4 (Munnakkaraya) was recorded while the lowest mean value (17.86 ± 4.3 $mScm^{-1}$) was recorded at the site 6 (Table VI). The concentrations of total suspended solids ranged from 14.2 ± 0.30 to 79.3 ± 5.30 mgL^{-1} (Table VI). TSS levels were slightly above the maximum recommended value for the discharge of industrial wastewater standards by the CEA (2007). Spatially, the highest salinity values were at site 4 (Munnakkaraya) and site 3 (Duwa) followed by site 2 (Pitipana) and site 1 (Sea mouth) with average salinities of 24.72 ± 3.21, 23.6 ± 2.4, 23.1 ± 2.80 and 15.4 ± 1.20, respectively. The results indicated that the levels of salinity are comparatively low in site 7 (Dandugam Oya), in the Southern regions of the estuary (Table VI). This may be attributed to the dilution effect caused by the high freshwater discharge in the downstream of Dandugam Oya and Ja - Ela canal entering from the Southern region of the estuary. During the study period, Biochemical Oxygen Demand varied in between 19.45 ± 6.2 to 29.8 ± 3.7 mgL^{-1} (Table VI). Maximum value of BOD was recorded

Table VI: The mean (± SD) values of physico-chemical parameters at the sampling sites

Parameters	Site - 1	Site - 2	Site - 3	Site - 4	Site - 5	Site - 6	Site - 7	Standard Limits CEA(2007)
Water temperature (°C)	30.50± 0.34^{b} (30.16-30.84)	30.37±0.18^{b} (30.19-30.55)	30.70±0.19^{b} (30.51-30.51)	30.56±0.30^{b} (30.26-30.26)	30.57±0.45^{b} (30.12-31.02)	29.95 ± 0.6^{a} (29.89-30.55)	29.50±0.40^{a} (29.10-29.90)	**40 ^{0}C**
Salinity (ppt)	15.4 ± 1.20^{c} (14.2 - 16.6)	23.1 ± 2.80^{d} (20.3 - 25.9)	23.6 ± 2.4^{d} (21.2 - 26.0)	24.72 ± 3.2^{d} (21.5 - 27.9)	12.9 ± 0.90^{bc} (11.1 - 13.8)	13.9 ± 1.50^{bc} (12.4 - 15.4)	1.4 ± 0.30^{a} (11.1 - 11.7)	**0.1 - 28.0**
Electrical Conductivity (mScm^{-1})	27.5 ± 4.20^{a} (18.3 - 26.7)	23.7 ± 5.30^{a} (18.4 - 29.0)	25.9 ± 5.3^{a} (20.6 - 31.2)	26.7 ± 6.30^{a} (20.4 - 33.0)	25.1 ± 6.50^{a} (18.6 - 31.6)	17.86 ± 4.3^{a} (13.5 - 22.1)	17.9 ± 4.40^{a} (13.5 - 22.3)	-
pH	7.67 ± 0.27^{a} (7.4 - 7.87)	7.70 ± 0.39^{a} (7.31 - 8.09)	7.60 ± 0.58^{a} (7.02 - 8.18)	8.30 ± 0.36^{a} (7.94 - 8.66)	7.60 ± 0.39^{a} (7.21 - 7.99)	7.60 ± 0.22^{a} (7.38 - 7.82)	7.20 ± 0.22^{a} (6.98 - 7.42)	**5.5 - 9.0**
Dissolved Oxygen (mgL^{-1})	7.61 ± 1.80^{cd} (5.81 - 9.41)	8.30 ± 1.50^{d} (6.8 - 9.8)	10.8 ± 2.80^{e} (8.0 - 13.6)	7.1 ± 1.10^{cd} (6.0 - 8.2)	5.60 ± 1.40^{bc} (4.2 - 7.0)	4.50 ± 1.60^{ab} (2.9 - 6.1)	3.90 ± 1.10^{a} (2.8 - 5.0)	-
Ammoniacal - nitrogen (mgL^{-1})	0.42 ± 0.20^{a} (0.22 - 0.62)	0.33 ± 0.11^{a} (0.44 - 0.22)	0.43 ± 0.20^{a} (0.23 - 0.63)	0.37 ± 0.11^{a} (0.26 - 0.48)	0.33 ± 0.10^{a} (0.43 - 0.23)	0.31 ± 0.1^{a} (0.41 - 0.21)	0.35 ± 0.20^{a} (0.15 - 0.55)	**50.0**
Nitrate - nitrogen (mgL^{-1})	1.03 ± 0.68^{c} (0.35 - 1.71)	0.75 ± 0.09^{bc} (0.66 - 0.84)	1.02 ± 0.10^{c} (0.92 - 1.12)	0.47 ± 0.16^{ab} (0.31 - 0.63)	0.47 ± 0.01^{ab} (0.46 - 0.48)	0.53 ± 0.02^{ab} (0.51 - 0.55)	0.79 ± 0.06^{bc} (0.3 - 0.85)	**5.0**
Nitrite - nitrogen (mgL^{-1})	0.38 ± 0.01^{d} (0.37 - 0.39)	0.44 ± 0.05^{ab} (0.39 - 0.49)	0.23 ± 0.01^{c} (0.22 - 0.24)	0.37 ± 0.02^{d} (0.35 - 0.39)	0.17 ± 0.02^{b} (0.15 - 0.19)	0.48 ± 0.04^{ab} (0.44 - 0.52)	0.16 ± 0.05^{b} (0.11 - 0.21)	
Orthophosphate (mgL^{-1})	0.40 ± 0.11^{d} (0.29 - 0.51)	0.33 ± 0.01^{ab} (0.32 - 0.34)	0.52 ± 0.03^{da} (0.49 - 0.55)	0.38 ± 0.11^{c} (0.27 - 0.49)	0.31 ± 0.05^{ab} (0.26 - 0.36)	0.33 ± 0.05^{ab} (0.28 - 0.38)	0.35 ± 0.02^{c} (0.33 - 0.37)	**5.0**
Total Suspended Solid (mgL^{-1})	10.3 ± 0.03^{a} (10.2 - 10.3)	18.2 ± 0.09^{a} (18.1 - 18.2)	15.6 ± 0.17^{a} (15.4 - 15.7)	17.3 ± 0.05^{a} (17.2 - 17.35)	76.8 ± 4.10^{b} (72.7 - 80.9)	79.3 ± 5.30^{b} (74.0 - 84.6)	14.2 ± 0.30^{a} (13.9 -14.5)	**150.0**
Chemical Oxygen Demand (mgL^{-1})	423 ± 110.0^{c} (313.0-533.0)	424 ± 128.3^{c} (295.7-552.3)	510.0 ± 315^{c} (195.0-825.0)	426.4 ± 132^{c} (294.4-558.0)	396.5 ± 167^{c} (229.5-563.5)	245.5 ± 113^{b} (132.5-358.0)	96.4 ± 70.0^{a} (24.4-166.4)	**250.0**
Biochemical Oxygen Demand (mgL^{-1})	19.45 ± 6.2^{b} (13.25 - 25.6)	27.4 ± 6.10^{da} (21.3 - 33.5)	23.3 ± 4.50^{abc} (18.8 - 27.8)	24.9 ± 6.40^{cd} (18.5 - 31.3)	23.15 ± 5.5^{abc} (17.6 - 28.6)	23.6 ± 5.50^{abc} (18.1 - 29.1)	29.8 ± 3.70^{e} (26.1 - 33.5)	**30.0**

at site 07 (Dandugam Oya). The results of BOD levels at the sampling site 7 (Dandugam Oya) slightly exceeded the CEA recommended limits of 30 mgL^{-1}. In case of Chemical Oxygen Demand, site 3 (Duwa) showed the highest value of 510.0 ± 315.0 mgL^{-1}, symbolizing the presence of a high pollution load within the estuarine water (Table VI). High levels of COD recorded in this area may be partly due to the discharge of effluents from boat repair yards. BOD values at site 7 (Dandugam Oya) and COD at site 3 were higher compared with the permissible limits for the discharge of industrial wastewater standards of CEA (2007). According to the results obtained, it is clear that the highest value of Nitrate - nitrogen (1.03 ± 0.68 mgL^{-1}) was noted at site 1 (Table VI) which is highly polluted when compared with the other sampling locations examined in the estuary.

Highest concentration of Nitrite - nitrogen (0.48 ± 0.04mgL^{-1}) was recorded at site 06 (Table VI). The ammoniacal-nitrogen concentration varied from 0.35 ± 0.20 mgL^{-1}(site 7) to 0.43± 0.20 mgL^{-1} during (Site 3) the study period. Highest concentration of Orthophosphate (0.52 ± 0.03 mgL^{-1}) was recorded at site 3 (Table VI). Orthophosphate concentration also showed similar variations across the sam-

pling sites within the Negombo estuary. According to water quality standards for the nutrient concentrations, the sampling sites did not satisfy with the wastewater standards of CEA (2007). The dendrogram of the cluster analysis (Figure 9, based on Euclidean Distance) suggested the presence of three major clusters. Sites 1 to 4 formulated the first cluster, while sites 5 and 6 formed the second cluster at a Euclidean distance of 6.56. On the other hand, sites 7 is in the third cluster, which is separated at a Euclidean distance of 13.24 from the other two clusters (Figure 9). The results of the Analysis of Similarities (ANOSIM) also confirmed the statistical significance of the above three clusters ($p < 0.05$, Global R 0.751) in terms of overall water quality among the studied sampling sites. Physico-chemical parameters in the first cluster in the Northern region shows a level of 25% similarity. In the second cluster, the sites 7 (Dandugam Oya) shows a level of similarity of 50%.

A. Heavy metal concentrations in water

The mean concentration values are presented in Table VII. From the results, it can be depicted that the concentration (μgL^{-1}) of metals in water showed ranges as: Zn from 320.0 ± 75.3 ± 15.3 to 460 ± 132.0; Pb from 4.5 ± 0.42 to 70.4 ± 13.0; Hg from 1.0 ± 0.01 to4.0 ± 0.2; Cd from 2.8 ± 0.20 to 13.2 ± 0.04; Cu from 16.0 ± 0.23 to 22.6 ± 0.8 and Cr from 6.5 ± 0.20 to 14.3 ± 0.24, respectively.

Variation of physico-chemical factors at the sampling sites and the reference site (n = 24). Results are presented as mean ± standard error of the mean. Means not followed by the same superscript along a row are significantly different from each other (ANOVA, Tukey's test, $p < 0.05$).

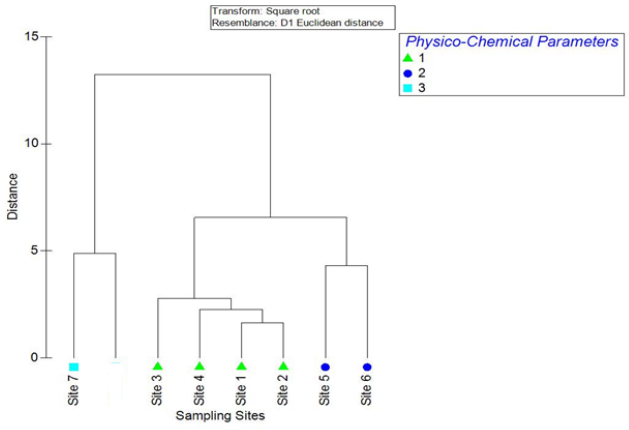

Figure 9: Dendrogram of the cluster analysis for physico-chemical parameters of water at sampling sites

The concentration of Zn in surface water recorded in this study did not exceed the recommended limit of 2000 μgL^{-1}, which is the tolerance limit for the wastewater quality standards for CEA (2007). The concentrations of Pb in water were comparatively high in site 1 (70.4 ±13.0 μgL^{-1}) and the lowest Pb levels were recorded in site 5 (2.1 ± 0.23 μgL^{-1}) and site 8 (2.1 ± 0.8 μgL^{-1}) (Table VII) but the levels of Pb was not exceeding the recommended tolerance limits of 100.0 μgL^{-1} for the discharging industrial wastewater into inland surface water bodies as stated in the quality standards for CEA (2007). The levels of Hg in water were high in sites 3 (4.0 ± 0.20 μgL^{-1}) 4 (4.0 ± 0.2 μgL^{-1}) and 6 (5.7 ± 0.5μgL^{-1}) (Table VII). The Hg levels observed in sampling sites were exceeding the standard threshold levels in recommended limits (0.5 μgL^{-1}) of wastewater quality standards in Sri Lanka CEA (2007). The mean concentrations of Cd (μgL^{-1}) in surface water from all the sampling sites are given in Table VII. The lowest mean Cd levels were recorded in site 8 (2.3 ± 0.4 μgL^{-1}), while the highest mean Cd level was observed at site 2 (13.2±0.04 μgL^{-1}) (Table VII). The present study indicates that Cd concentration observed in all the sampling sites were not exceeding the recommended tolerance limits of 200.0 μgL^{-1} for the discharging industrial wastewater into inland surface water bodies as stated in the quality standards for Sri Lanka by CEA (2007). The mean concentrations of Cu (μgL^{-1}) in surface water range from 2.4 ± 0.25 to 22.6 ± 0.8 (Table VI). The mean Cu levels recorded from the study sites did not exceed the CEA standard limit (3000.0 μgL^{-1}) for industrial and domestic wastewater discharged into inland surface water quality standards of CEA (2007). The mean Cr levels (Table V) ranged from 2.6 ± 0.2 to 14.3 ± 0.24 μgL^{-1}. The mean Cr levels recorded in this study did not exceed the recommended limit of 500.0 μgL^{-1} of tolerance limits for industrial and domestic wastewater discharged into inland surface water quality standards in Sri Lanka.

B. Heavy metal concentrations in Mugil cephalus tissues

The concentrations ($mgkg^{-1}$) of heavy metals in edible muscle tissues of *Mugil cephalus* were: Zn, 0.6 ± 0.14 to 5.0 ± 1.61; Pb, 0.03 ± 0.02 to 0.7 ± 0.01; Hg, 0.02 ± 0.01 to 0.38 ± 0.11; Cd, 0.03 ± 0.01 to 0.04 ± 0.01; Cu, 0.16 ± 0.05 to 0.39 ± 0.07 and Cr, 0.02 ± 0.01 to 0.36 ± 0.03 (Table VIII).The average highest accumulation of Zn in site 3 and 4; Pb in site 3 and 4; Hg in sites 1, 2, 3 and 4; Cd in site 4; Cu in site 1, 2, 3 and 4; Cd in site 4 and site 6 and Cr in site 3 were identified in this study (Table VIII).

IV. Discussion

The mean temperature of the study sites ranged as the same throughout the sampling sites. Low dissolved oxygen values found in site 7 could be attributed to wastewater received from Ekala industrial zone, discharging organic wastewater, which increased organic matter and decomposed dissolve organic matter. The highest DO recorded at site 3 may be due to closeness to sea mouth and mixing of surface waters by wind and water due to increased tide at which oxygen from the air can be dissolved or mixed into water, daily variations in the tides, and may be due to the relatively free from organic load (Mitra, *et al*., 2010). The DO values were significantly different ($p > 0.05$). According to the range of DO given by USEPA (Weiner 2008), the value of DO at site 2 and site 3 were in the range of good quality of water (above 8.0) but the DO values at Pitipana and Duwa

Table VII: The concentrations of heavy metals in water (μgL-1) (Mean ± SE) at sampling sites

Metals	Site - 1	Site - 2	Site - 3	Site - 4	Site - 5	Site - 6	Site - 7	Standard Limits CEA(2007)
Zn	433.0±112.0^{b}	400 ± 102.2^{b}	440.0± 118.2^{b}	460.0 ± 132.0^{b}	360.0 ± 98.3^{b}	320.0 ± 75.3^{b}	350.0 ± 83.2^{b}	**2000**
	(321.0-545.0)	(297.8-502.2)	(321.8 -558.2)	(328.0 -592.0)	(261.7- 458.3)	(244.7 -395.3)	(266.8 -433.2)	
Pb	70.4 ± 13.0^{b}	40.4 ± 6.30$^{a\,b}$	30.8 ± 5.40^{ab}	20.2 ± 3.20^{a}	2.1 ± 0.23^{a}	6.5 ± 0.85	4.5 ± 0.42^{ab}	**100**
	(57.4 - 83.4)	(34.1 - 46.7)	(10.6 - 36.2)	(17.0 - 23.4)	(1.87 - 2.33)	(5.65 - 7.35)	(4.08 - 4.92)	
Hg	3.0 ± 0.01^{ab}	2.1 ± 0.01^{ab}	4.0 ± 0.20^{ab}	4.0 ± 0.20^{ab}	2.0 ± 0.01^{ab}	5.7 ± 0.50^{b}	1.0 ± 0.01^{ab}	**0.5**
	(2.9 - 3.01)	(2.09 - 2.11)	(3.8 - 4.2)	(3.8 - 4.2)	(1.99 - 2.01)	(5.2 - 6.2)	(0.99 - 1.01)	
Cd	11.5 ± 0.02^{ab}	13.2 ± 0.04^{ab}	10.7 ± 0.02^{ab}	9.6 ± 0.01^{ab}	4.5 ± 0.20^{ab}	4.6 ± 0.20^{ab}	2.8 ± 0.20^{ab}	**200**
	(11.4 - 11.52)	(13.16 -13.24)	(10.68 -10.72)	(9.59 - 9.61)	(4.3 - 4.7)	(4.4-4.8)	(2.6 - 3.0)	
Cu	13.6 ± 0.50^{abc}	15.3 ± 0.6^{abc}	22.6 ± 0.80^{b}	18.2 ± 0.50^{bc}	13.3 ± 0.50	6.6 ± 0.05^{ab}	16.0 ± 0.23^{abc}	**3000**
	(13.1 - 14.2)	(14.7 - 15.9)	(21.8 - 23.4)	(17.7 - 18.7)	(12.8 - 13.8)	(6.55 - 6.65)	(15.7 - 16.23)	
Cr	13.4 ± 0.23^{a}	10.6 ± 0.25^{a}	13.2 ± 0.22^{a}	14.3 ± 0.24^{a}	12.6 ± 0.01^{a}	10.5 ± 0.52^{a}	6.5 ± 0.20^{a}	**500**
	13.17 -13.63)	10.35 - 10.85)	(12.98 -13.42)	(14.06 -14.54)	(12.5 - 12.61)	(9.98 - 11.02)	(6.3 - 6.7)	

Results presented are as mean ± standard error. Means indicated for each parameter by different superscripts are significantly different from each other according to the ANOVA and Tukey's test, $p < 0.05$.

Table VIII: The levels of selected metals in fish tissues (mgkg-1) (Mean ± SE) at eight sampling sites

Metals	Site - 1	Site - 2	Site - 3	Site - 4	Site - 5	Site - 6	Site - 7	EU (2006) Standard
Zn	3.6 ± 0.87^{bc}	4.2 ± 0.85^{cd}	5.0 ± 1.61^{d}	4.9 ±1.35^{d}	2.8 ± 0.41^{b}	3.1 ± 0.14^{b}	0.6 ± 0.14^{a}	-
	(2.73 - 4.47)	(3.35-5.05)	(3.39 - 6.61)	(3.55 - 6.22)	(2.39 - 3.21)	(2.96 - 3.24)	(0.46 - 0.74)	
Pb	0.5 ± 0.02^{b}	0.6 ± 0.02^{bc}	0.7 ± 0.01^{d}	0.7 ± 0.02^{d}	0.04 ± 0.02^{a}	0.04 ± 0.02^{a}	0.03 ± 0.02^{a}	**0.3**
	(0.48 - 0.52)	(0.58 -0.62)	(0.69 - 0.71)	(0.68 - 0.72)	(0.02 - 0.06)	(0.02 - 0.06)	(0.01 - 0.05)	
Hg	0.36 ± 0.07^{c}	0.34 ± 0.12^{c}	0.38 ± 0.11^{c}	0.36 ± 0.04^{c}	0.14 ± 0.04^{b}	0.19 ± 0.06^{b}	0.02 ± 0.01^{a}	**1.0**
	(0.29 - 0.43)	(0.22 -0.46)	(0.27 - 0.49)	(0.32 - 0.40)	(0.1 - 0.18)	(0.13 - 0.25)	(0.01 - 0.03)	
Cd	0.03 ± 0.01^{c}	0.03 ± 0.01^{c}	0.03 ± 0.01^{c}	0.04 ± 0.01^{c}	0.03 ± 0.01^{b}	0.03 ± 0.01^{b}	0.03 ± 0.01^{b}	**0.05**
	(0.02 - 0.04)	(0.02 -0.04)	(0.02 - 0.04)	(0.03 - 0.05)	(0.02 - 0.04)	(0.02 - 0.04)	(0.02 - 0.04)	
Cu	0.39 ± 0.07^{c}	0.37 ± 0.07^{c}	0.37 ± 0.09^{c}	0.36 ± 0.07^{c}	0.36 ± 0.07^{c}	0.39 ± 0.05^{c}	0.16 ± 0.05^{b}	**0.01**
	(0.32 - 0.46)	(0.3 - 0.44)	(0.28 - 0.46)	(0.29 - 0.43)	(0.29 - 0.43)	(0.34 - 0.44)	(0.11 - 0.21)	
Cr	0.26 ± 0.02^{b}	0.34 ± 0.02^{c}	0.36 ± 0.02^{c}	0.36 ± 0.03^{c}	0.24 ± 0.06^{b}	0.02 ± 0.01^{a}	0.02 ± 0.01^{a}	**0.03**
	(0.24 - 0.28)	(0.32- 0.36)	(0.34 - 0.38)	(0.33 - 0.39)	(0.18 - 0.3)	(0.01 - 0.03)	(0.01 - 0.03)	

Results presented are as mean ± standard error. Means indicated for each parameter by different superscripts are significantly different from each other according to the ANOVA and Tukey's test, $p < 0.05$.

was within the range (6.5 - 8.0 mgL^{-1}). In this study, the Negombo estuary water was found to be slightly alkaline where pH varied from 7.2 to 8.3. The sampling sites 1 and 3 situated at the Northern region recorded relatively high levels of Nitrate nitrogen concentration, which could be due to release of effluents from the fish processing factories in this area. Biochemical Oxygen Demand (BOD) in Dandugam Oya is mainly due to industrial effluents and high amounts of organic matter from different pollution sources. The high BOD levels are attributed mostly to low flushing rates in the estuary (CCD, 2005). Higher COD values were recorded in sites 1, 2, 3, 4 and 5. This may be due to the presence of higher concentrations of organic matter containing discharges from untreated industrial effluents into the estuary by some of the factories in the area, discharge of municipal wastewater (untreated and detergent carrying wastewater) and other domestic wastes dumped directly into the estuary. The pollution in the Northern region could be due to pollution from various anthropogenic activities such as solid waste dumping, wastes and wastewater discharges from industries, slaughterhouses, shrimp farms, and hatcheries and wastes disposed from boat yards. Higher concentration of Pb in water samples collected from the Northern region compared to the other sampling sites may be partly due to discharge of burned and unburned fuel from motor boats to the estuarine water. Mercury received to the estuary could be due to direct discharge of industrial effluents and deposits from municipal waste incineration. Comparison to the Northern region of the estuary, the levels of metals in the South region were lower but Zn and Cr levels in sediments were higher. Site

5 in the Western region receives wastes from hotels, shrimp farms and dry fish processing industries. Concentrations of metals in water of this area were low but concentrations of sediment bound metals (Zn and Cr) were comparatively higher. Site 6 in the Eastern region receives effluents from the Katunayake industrial processing zone, hotels and a housing schemes hence the concentration of Hg in this area were relatively higher. The main sources of pollutants received in Bolgoda Lake were urban and industrial wastes from multiple sources (Senarathne Pathiratne, 2007). Pb is mainly from industrial processes and consumer products such as batteries rolled and extruded products, cable sheathing, paints, alloys and pigments (Indrajith, *et al*., 2008). Copper gets into aquatic ecosystems from diverse sources such as Cu compounds used in wood preservatives; electroplating and azo dye manufacture (Akan, *et al*., 2010).

Pb, Cd and Hg, biologically non essential metals, are accumulated in human tissues. These metals pose health risks to human. The concentrations of Pb, Cd and Hg in edible muscles of *Mugil cephalus* collected from Negombo estuary ranged from 0.03 ± 0.02 to 0.7 ± 0.01; 0.03 ± 0.01 to 0.04 ± 0.01 and 0.02 ± 0.01 to 0.38 ± 0.11 $mgkg^{-1}$ wet weight, respectively. Wet weight maximum allowable limits of Pb, Cd and Hg in fish for human consumption specified by European Union are 0.3, 0.05 and 1.0 $mgkg^{-1}$, respectively (EU, 2006). Pb, Cu and Cr metal levels detected in edible tissues of *Mugil cephalus* exceed the allowable limits in fish for human consumption specified by the European Union. Therefore, a serious threat is posed on the consumers of these fish from the estuary, which are contaminated with these metals. The differences observed in heavy metal concentrations in fish and shellfish studied was due to the feeding behaviour of these organisms (Madkour, 2012). The present study showed higher amounts of Pb, Cu and Cr in *Mugil cephalus* which is a benthic species feed on sediment materials. At sampling sites 1, 2, 3 and 4, Pb levels exceeded allowable limits in fish for human consumption specified by the European Union.

Negombo estuary is influenced by many anthropogenic activities and the "X-Press Pearl" disaster, which has occurred at a close proximity to the estuary, may pose a serious threat on the water, sediment quality and thereby influencing aquatic organisms. Annibaldi *et al*. (2011) reported that "Nicole" shipwreck (Ancona coast, Central Adriatic Sea) posed a serious Pb, Cd and Cu pollution affecting the sediments due to the presence of hulk. Annibaldi *et al*. (2011) also reported that dissolved Cd and Pb contents of the contaminated area exceeded the Italian legal limits. Dimitrakakis *et al*. (2014) reported that due to "Sea Diamond" shipwreck, approximately 75–80 g of mercury, 630–1,050 g of cadmium and 1.14–1.26 tons of lead were added to the marine environment due to electrical and electronic equipment present in the ship. They further reported a significant amount of heavy metals such as copper, nickel, ferrous and chromium also added to the marine environment due to the hulk of the ship. Lead, zinc and cadmium were concentrations higher than the permissible limits set by the Unites States Environmental Protection Agency for seawater was reported in the shipwreck environment (Dimitrakakis *et al*., 2014). "X-Press Pearl" shipwreck that were carrying huge amounts of chemicals hence has posed a serious environmental pollution to the Negombo estuary and this study presented in this paper can be considered as a baseline or the future studies on water, sediment pollution and bioaccumulation of heavy metals in edible fish.

V. Conclusion

In conclusion, higher concentrations of Pb, Cu and Cr in fish species collected from the selected sampling sites in the Northern region of the estuary may pose a health risk to the consumers due to accumulation of high levels of these heavy metals. As Negombo estuary continuously receives urban and industrial wastes from multiple sources, it is very important to monitor heavy metals in the food fish species in North region of Negombo estuary regularly for the safety of fish consumers. As the "X-Press Pearl" shipwreck has posed a serious threat to the Negombo estuary, this study can be considered as a baseline for the future studies on water and sediment pollution and bioaccumulation of heavy metals in edible fish species from the estuary.

VI. Acknowledgements

The Author wish to express their sincere gratitude to National Aquatic Resources Research and Development Agency (NARA) for providing financial assistance to conduct the present study.

References

Akan, J., Abdulrahman, F., Mamza, P. and Aishatu, N., Akan, J. C, Abdulrahman, F. I, Mamza, P. T, Aishatu, N., (2010). Effect of environmental pollution on the quality of river Ngada, Maiduguri Metropolis, Borno State, Nigeria. *Global Science books. Terrrestial and Aquatic Environ. Toxicology, 6*(1), pp. 40-46.

American Public Health Association (APHA), (2012). Standard methods for the Examination of Water and Wastewater 22nd edition, Published by American Water Works Association Washington, DC - 20005.

Annibaldi, A., Illuminati, S., Truzzi, C., Scarponi, G. (2011). SWASV speciation of Cd, Pb and Cu for the determination of seawater contamination in the area of the Nicole shipwreck (Ancona coast, Central Adriatic Sea). *Marine pollution bulletin, 62*(12), 2813-2821.

Association of Official Analytical Chemists (AOAC), (2012). Official Methods of Analysis. 19th ed. Atomic Absorption Method for Fish. Washington, DC. 20005.

Central Environmental Authority (CEA), (2007). Ambient water quality standards for inland waters Sri Lanka. *Central Environmental Authority, Sri Lanka*.

Costal Conservation Department, (2005). Special Area Management Plan for Negombo Lagoon Coast First Edition, Colombo. p. 70.

Dimitrakakis, E., Hahladakis, J., Gidarakos, E. (2014). The "Sea Diamond" shipwreck: environmental impact assessment in the water column and sediments of the wreck area. *International Journal of Environmental Science and Technology, 11*(5), 1421-1432.

European Union (EU), (2006). The Commission of the European Communities, Commission regulation, (EC) No 1881/2006 amending regulation (EC) No. 466/2001. *Official Journal of the European Communities*, Volume 1, pp. 37/5 - 1-37/6.

Hettiarachchi, S. S. L. and Samarawickrama, S. P., (2003). Towards Improved Environmental Management of the Negombo Lagoon. COPEDEC VI. *Journal of the institution of engineers*, XXXIV(1), pp. 43-55.

Indrajith, H. A. P., Pathirane, K. A. S. and Pathirane, A., (2008). Heavy metal levels in two food fish species from Negombo estuary, Sri Lanka; Relationships with the body size. *Sri Lanka Journal of Aquatic Science*, Volume 13, pp. 63-81.

Madkour, N. K., (2012). The beneficial role of celery oil in lowering of di (2 - ethylhexyl) phthalate-induced testicular damage. *Toxicological Industrial Health*, Volume 53, pp. 112-118.

Mitra, A., Banerjee, K., Ghosh, R. and Ray, S. K., (2010). Bioaccumulation pattern of heavy metals in the shrimp's ofthe lower stretch of the River Ganga. *Mesopotamian Journal of Marine Science, 25*(2), pp. 1-14.

Senarathne, P. and Pathiratne, K., (2007). Accumulation of heavy metals in a food fish, Mystus gulio inhabiting Bolgoda Lake, Sri Lanka.. *Sri Lanka Journal of Aquatic Sciences*, Volume 12, pp. 61-75.

Weiner, E. R., (2008). Application of Environmental Aquatic Chemistry. Taylor and Francis, LLC. USA. 109 p.