International Journal of Advanced Information in Engineering Technology (IJAIET)
Vol.5, No.4, April 2018 ISSN: 2454-6933

Quality Assessment of Sediments in Bharathapuzha with Special Reference to Phosphate Fractionation and Metallic Contamination

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***Abstract*—Data from sediments provide information on the impact of distant human activity on the wider ecosystem. Sediment is responsible for transport of essential nutrients as well as pollutants. Therefore, the assessment of sediment is more conservative than water quality assessment for determining the degree of contamination and toxicity. Different forms of phosphorus and heavy metal prominence of the surface sediments in Bharathapuzha will be studied and reported in this paper.**

***Index Terms*—- Assessment, Contamination, Sediments, Phosphorus fractions, Metals**

I. INTRODUCTION

Typically, as part of the management of contaminated sites, it is required that the risk of harm from any potential contaminants be assessed before the sites undergo any major disturbance through redevelopment or remediation orders placed on them. The input and accumulation of phosphorus from external sources are the main cause of eutrophication of surface waters and, as such, the control of phosphorus pathways is regarded as the best strategy for the management of water bodies. Heavy metal pollution in aquatic ecosystems is a worldwide environmental problem that has received increasing attention over the last few decades because of its adverse effects. The contamination of aquatic systems by heavy metals, especially in sediments, has become one of the most challenging pollution issues owing to the toxicity, abundance, persistence, and subsequent bio-accumulation of these materials

A.Bharathapuzha

Bharathapuzha, also known as the River Nila, is a river in India in the state of Kerala. With a length of 209 km, it is the second-longest river in Kerala, after the Periyar River. The word "Nila" indicates the culture more than just a river. Nila has groomed the culture and life of south Malabar part of Kerala. The river went through a series of challenges which saw its degradation that has reached a point of no return. Due to the sand mining in the last 30 years, the thick sand bed has been completely vanished and has then been replaced with grasses and bushes which has become an environmental catastrophe. Environmentalists have predicted dire consequences and the untimely death of the river within the near future.
(https://en.wikipedia.org/wiki/Bharathappuzha).

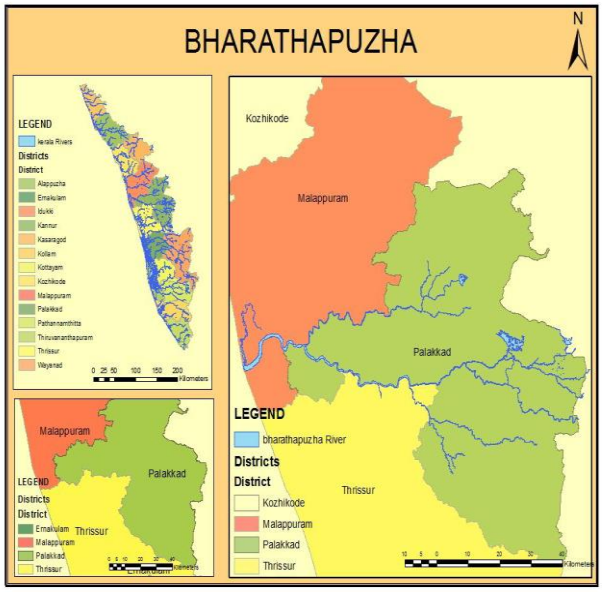


Fig.1. Bharathapuzha

II.METHODOLOGY

The surface sediment samples were taken at a depth of nearly 10 cm and placed in polythene bags, preserved in ice and transported to laboratory. The sediment samples were sieved, dried, powdered and transported to laboratory. Sediment pH was measured electrometrically with glass electrode pH meter in water. Total alkalinity was measured using acid base titration. The wet oxidation method of Walkley and Black was used to determine the organic carbon content in the sediment samples. The sediment particle size was determined using sieve analysis. Fractionations of phosphorus in the sediment samples were done using the Williams method. Iron content is determined using titration with permanganate solution followed by measurement using spectrophotometer. Manganese and zinc were determined using EDTA titrimetric method. Copper was determined using iodometric method. Distribution of lead and cadmium was determined using atomic absorption spectrophotometer.

TABLE I
SAMPLING STATIONS OF BHARATHAPUZHA WITH LATITUDE AND LONGITUDE

SI No:	Sampling stations	Latitude (in decimals)	Longitude (in decimals)
1.	Ponnani	10.787096	75.918693
2.	Chamravattom	10.820229	75.957320
3.	Kuttipuram	10.840417	76.023291
4.	Thrithala	10.808152	76.118898
5.	Pattambi	10.799198	76.184574
6.	Shoranur	10.761641	76.249130
7.	Ottapalam	10.764761	76.366868
8.	Peringottukurissi	10.755117	76.482192

A. Assessment according to United States Environmental Protection Agency (USEPA)

The chemical contaminations in the sediments were evaluated by comparison with the sediment quality guideline proposed by USEPA. These criteria are shown in TABLE II.

TABLE II
EPA GUIDELINES FOR SEDIMENTS (MG/KG DRY WEIGHTS)
(Javed Iqbal and Munir H Shah,2014)

Metal	Not polluted	Moderately polluted	Heavily polluted
Fe	<17000	17000 -25000	>25000
Mn	<300	300 -500	>500
Zn	<90	90-200	>200
Cu	<25	25-50	>50
Pb	<40	40-60	>60
Cd	-	-	>6

B. *Assessment according to Geo-accumulation index (I_{geo})*

A common criterion to evaluate the heavy metal pollution in sediments is the geo-accumulation index (I_{geo}), which is originally defined by Muller (1979) to determine metal contamination in sediments, by comparing current concentrations with pre-industrial levels and can be calculated by the following equation (Muller 1979)

$$I_{geo}=\log_2 [C_n /1.5\ B_n]$$

where, C_n is the concentration of element 'n' and B_n is the geochemical background value. The factor 1.5 is incorporated in the relationship to account for possible variation in background data due to lithogenic effect (P. K. Saha and M.D. Hossain,2011). Muller has defined seven classes of geoaccumulation index ranging from Class 0($I_{geo}\leq$ 0, unpolluted) to Class 6(I_{geo} >6, extremely polluted).

TABLE III
MULLER'S CLASSIFICATION FOR GEO-ACCUMULATION INDEX
(P. K. Saha and M.D. Hossain,2011)

I_{geo} Value	Class	Sediment quality
≤ 0	0	Unpolluted
0-1	1	From unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	From moderately to strongly polluted
3-4	4	Strongly polluted
4-5	5	From strongly to extremely polluted
>6	6	Extremely polluted

C. *Assessment of pollution by calculating contamination factor, degree of contamination and pollution load index.*

Contamination factor (C^i_f) and the degree of contamination (C_d) are used to describe the contamination of given toxic substance and is given by

$$C^i_f = C^i{}_{0\text{-}1} / C^i_n$$

and $C_d = \Sigma^n_{i=1} C^i_f$

where $C^i{}_{0\text{-}1}$ is the mean content of the substance; C^i_n is the reference shale value for the substance. The contamination factor C_f and the degree of contamination will be used to determine the contamination status of the sediment in the present study. The degree of contamination (C_d) is defined as the sum of all contamination factors. Sediment pollution load index (PLI) is calculated using the equation, PLI = (product of n number of C_f values)$^{1/n}$, where, C_f is the contamination factor, n is the number of metals and world average concentration of elements reported for shale is taken as their background values. The PLI values for each of the stations will be calculated. The PLI value of >1 is polluted whereas < 1 indicates no pollution. (Moonampadiyan Shiji et.al,2015).

TABLE IV
CONTAMINATION AND THEIR DESCRIPTION
(Moonampadiyan Shiji et.al,2015).

C^i_f	C_d	Description
$C^i_f < 1$	$C_d < 7$	Low degree of contamination
$1 < C^i_f < 3$	$7 < C_d < 14$	Moderate degree of contamination
$3 < C^i_f < 6$	$14 < C_d < 28$	Considerable degree of contamination
$C^i_f > 6$	$C_d > 28$	Very high degree of contamination

D. *Data analysis*

Xlstat software will be used for the statistical interpretation. Pearson correlation coefficients will be calculated in order to study inter-elemental relationship with their sediment properties. Principal component analysis

(PCA) will be used for evaluation and characterization of analytical data. The PCA will be performed using varimax normalized rotation on the dataset. Analytical results were elaborated by using the Geographical Information System(GIS) application, ArcGIS 10.3.1 software. It was used to show geochemical indices and spatially explain the contaminated areas in the form of interpolated maps.

III. RESULTS AND DISCUSSIONS

A. *Plotting of sampling points*

Sampling stations of Bharathapuzha were plotted using ArcGIS 10.3.1 software with the help of latitude and longitude.

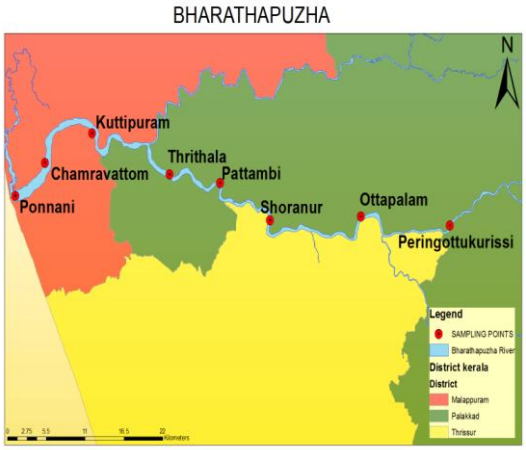


Fig. 2. Sediment sampling stations of Bharathapuzha

B. *Preliminary test results*

pH values of Bharathapuzha sediments ranged from 5.89 to 8.55 and total alkalinity values ranged from 113.2 to 412.5. Highest amount of organic carbon was found in sediments collected from Shoranur. The sediment sample was found to be silt based in general. Sand also had a noteworthy percentage, but very less amount of clay was observed in all stations.

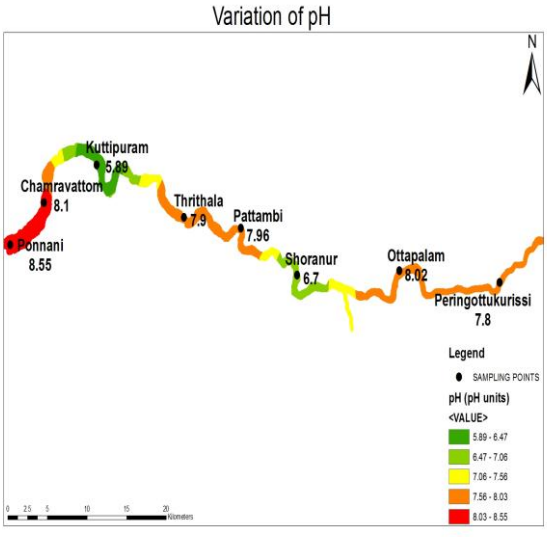


Fig.3. Variation in pH values in sediments of Bharathapuzha

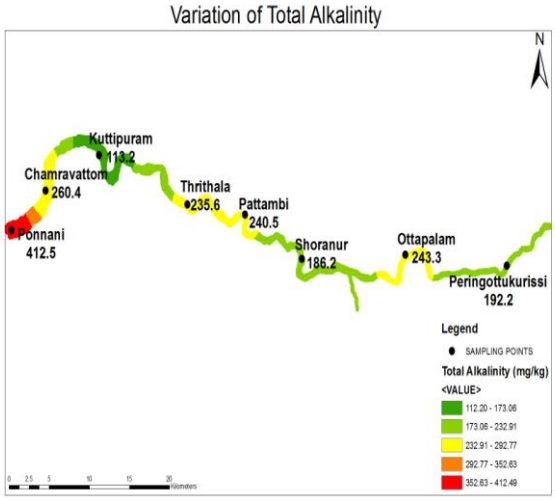


Fig.4. Variation in total alkalinity values in sediments of Bharathapuzha

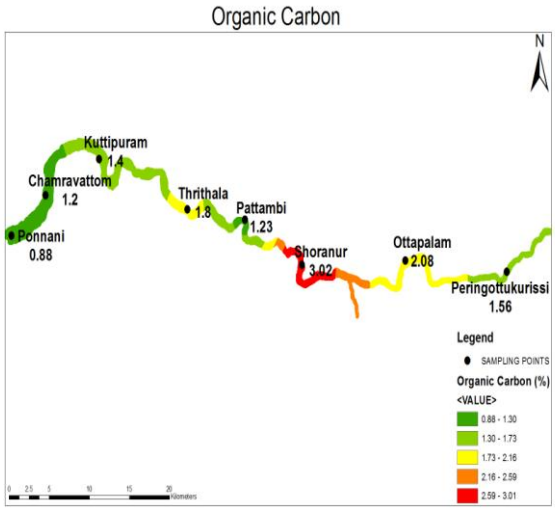


Fig.5. Spatial distribution of organic carbon (in %) in sediment samples of Bharathapuzha

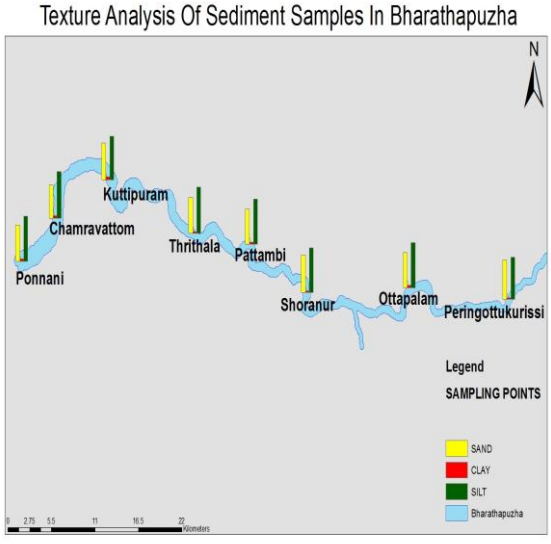


Fig.6. Texture analysis of sediment samples in Bharathapuzha

TABLE V
PRELIMINARY TEST RESULTS OF SEDIMENTS IN BHARATHAPUZHA

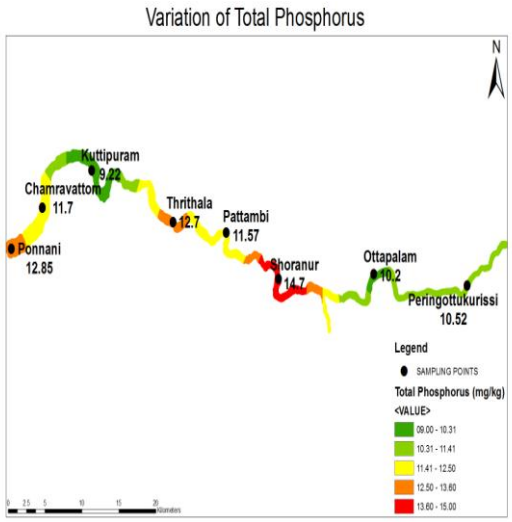
SI No.	Parameters	Unit	Variation of characteristics							
			1	2	3	4	5	6	7	8
1.	**pH**	pH units	8.55	8.1	5.89	7.9	7.96	6.7	8.02	7.8
2.	**Total alkalinity**	mg/kg	412.5	260.4	113.2	235.6	240.5	186.2	243.3	192.2
3.	**Organic carbon**	%	0.88	1.2	1.4	1.8	1.23	3.02	2.08	1.56
4.	**Particle size**	%								
	Sand		43.1	40.2	43.9	42.6	42.7	44.8	42.3	47.4
	Silt		53.9	56.2	51.9	55.2	54.4	53.3	54.1	50.2
	Clay		2.6	3.4	3.7	1.6	2.5	1.5	2.8	2.1

C. Phosphorus fractionation of sediment samples

TABLE VI
PHOSPHORUS FRACTIONATION OF SEDIMENTS IN BHARATHAPUZHA

SI.No	Sampling stations	Phosphorus fractions(mg/kg)				
		TP	Fe-Al P	Ca-P	OP	IP
1	**Ponnani**	12.85	1.67	0.03	8.76	0.62
2	**Chamravattom**	11.7	1.5	ND	8.5	0.5
3	**Kuttipuram**	9.22	1.02	ND	5.23	ND
4	**Thrithala**	12.7	2.18	0.2	8.1	0.56
5	**Pattambi**	11.57	1.2	0.04	7.52	0.49
6	**Shoranur**	14.7	2.03	0.06	9.28	0.69
7	**Ottapalam**	10.2	1.71	ND	7.17	0.47
8	**Peringottukurissi**	10.52	1.8	ND	8.16	0.39
	Average	11.682	1.638	0.082	7.84	0.531

Phosphorus (P) fractionation in sediments from aquatic ecosystems plays a critical role in determining P mobility and exchanges with the overlying water. The contents of different P fractions varied greatly. Total P concentrations of sediments varied from 9.22 to 12.85 mg/kg. Fe-Al P is used to estimate both short- and long-term available P in the sediments and is verified to be an indicator of algal-available P. Its average value in sediments of Bharathapuzha is 1.638mg/kg. HCl-P mainly represents calcium-bound P which appears to be non-motile and is not easily bio-available in the sediments. HCl-P was the least present P fraction in the sediments with an average value of 0.082 mg/kg.Most abundant P fraction was organic phosphorus with an average value of 7.84 mg/kg. Significant amount of inorganic phosphorus was not present and its value ranged from ND to 0.69.



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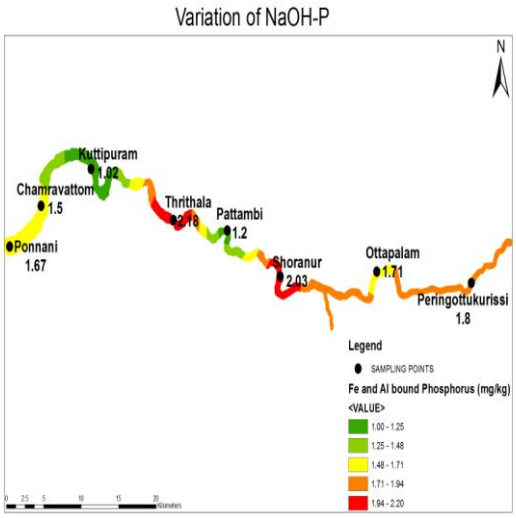

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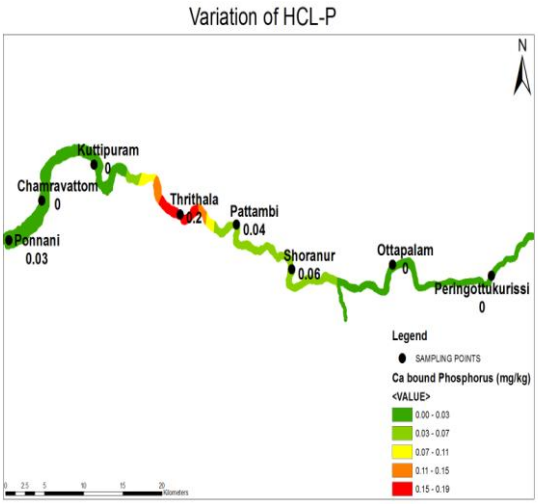

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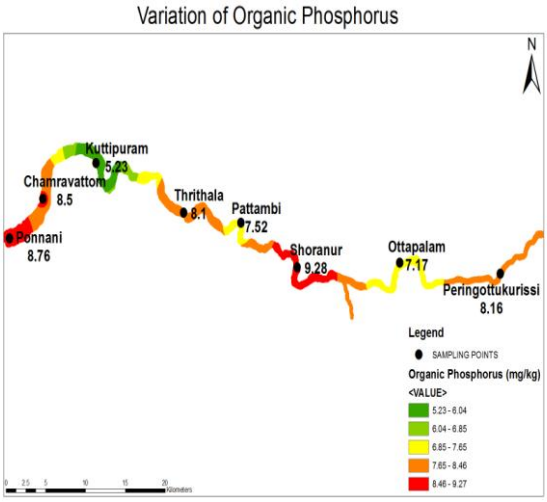

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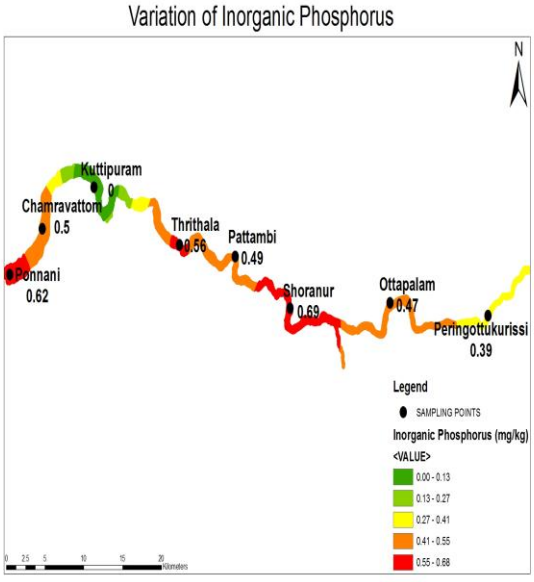


(e)

Fig.7. Spatial distribution of (a) TP (b) NaOH -P (c)HCl-P (d) OP (e) IP in sediment samples of Bharathapuzha

D. *Correlation of different phosphorus fractions versus organic carbon, iron, total alkalinity and pH*

The results of the correlation analysis indicate that organic carbon exhibited weak positive correlation with phosphorus fractions. Iron showed positive correlation with organic carbon (r =0.844). pH showed significant positive correlation with IP (r=0.611) and negative correlation with OC and Fe. TA showed strong positive correlation with pH (r=0.824). OP and IP exhibited high positive correlation(r=0.925). Other highly correlated pairs include TP with OP (r=0.843) and IP (r=0.855)(see TABLE VII).

E. *Principal component analysis of phosphorus fractions*

TABLE VIII
COMPONENT LOADINGS OF PHOSPHORUS FRACTIONS

Variables	Component	
	PC1	PC2
Fe-Al P	0.856	0.243
Ca-P	0.600	0.776
OP	0.909	-0.373
IP	0.934	-0.254
TP	0.917	-0.106
% of Variance	72.624	17.518

PCA rendered two PCs explaining 90.142 % of the total variance of the data set. PC1 explaining 77.624% of total variance has strong positive loadings (>0.70) on Fe-Al P, OP, IP and TP. PC2 accounting for 17.518 % of total variability was strongly related to Ca-P.

F. *Distribution of metals in sediments of Bharathapuzha*

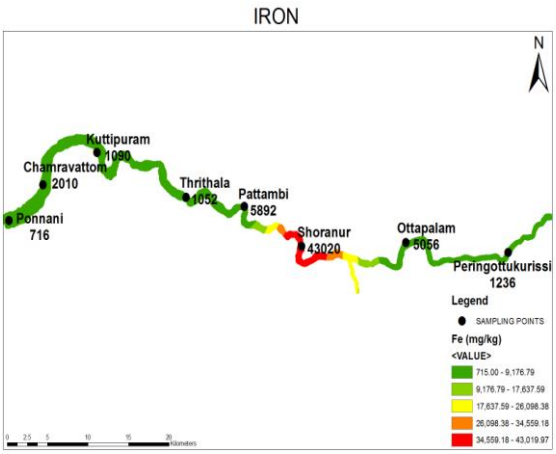
Concentration of Fe was less than the average shale value (46700) in all sampling stations. Concentration of Mn varied from 7.16 to 98.18 mg/kg whereas for Zn it varied from ND to 72.29 mg/kg. Concentration of Cu observed was less except at sediments collected from Shoranur, where a concentration of 28.8 mg/kg was obtained. Concentration of Pb observed was higher than the average shale value (20 mg/kg) for the sediments collected from Kuttipuram and Shoranur. Cd concentrations varied from ND to 8.5 mg/kg with an average value of 2.295 mg/kg. On the average basis, the metals follow a decreasing order: Fe >Mn>Pb>Zn>Cu> Cd.

TABLE VII
CORRELATION COEFFICIENT MATRIX SHOWING CORRELATION OF DIFFERENT PHOSPHORUS FRACTIONS, OC, FE, pH AND TA

Variables	Fe-Al P	Ca-P	OP	IP	TP	OC	Fe	pH	TA
Fe-Al P	1	-	-	-	-	-	-	-	-
Ca-P	0.617	1	-	-	-	-	-	-	-
OP	0.702	0.258	1	-	-	-	-	-	-
IP	0.704	0.382	0.925	1	-	-	-	-	-
TP	0.640	0.501	0.843	0.855	1	-	-	-	-
OC	0.552	0.231	0.253	0.308	0.432	1	-	-	-
Fe	0.364	0.083	0.447	0.449	0.678	0.844	1	-	-
pH	0.307	0.106	0.541	0.611	0.191	-0.418	-0.396	1	-
TA	0.213	0.063	0.544	0.617	0.382	-0.448	-0.237	0.824	1

TABLE IX
DISTRIBUTION OF METALS IN SEDIMENTS OF BHARATHAPUZHA

SI.No	Sampling stations	Heavy metals(mg/kg)					
		Fe	Mn	Zn	Cu	Pb	Cd
1	**Ponnani**	716	32.12	0.9	5.9	10.52	ND
2	**Chamravattom**	2010	22.32	0.3	6.6	9.3	ND
3	**Kuttipuram**	1090	18.96	ND	4.1	22.01	0.85
4	**Thrithala**	1052	28.6	ND	5.2	12.6	0.42
5	**Pattambi**	5892	25.55	9.5	6.5	16.5	1.2
6	**Shoranur**	43020	98.18	72.29	28.8	26.2	8.5
7	**Ottapalam**	5056	18.24	3.6	14.7	19.6	1.9
8	**Peringottukurissi**	1236	7.16	1.3	3.6	9.4	0.9
	Average	7509	31.391	14.648	9.425	15.766	2.295
	Average shale value	46700	900	95	45	20	0.3



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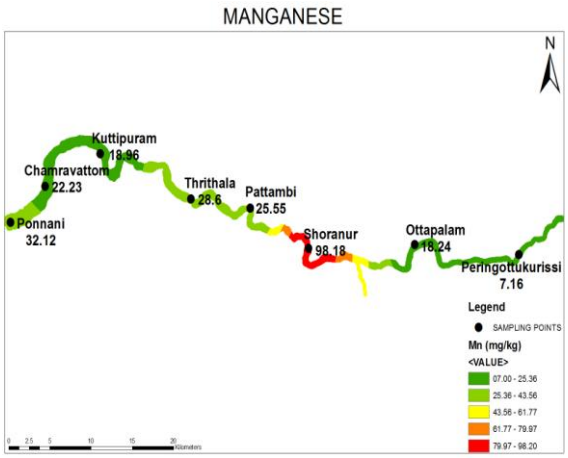

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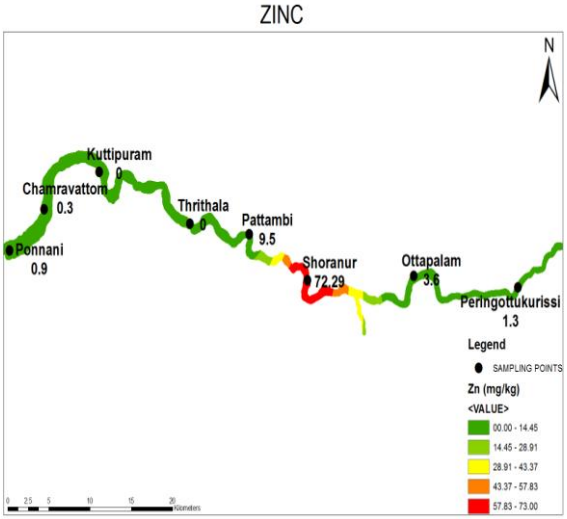

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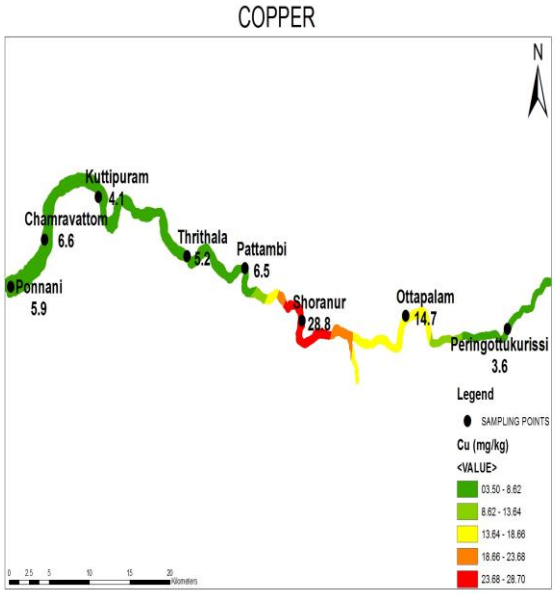

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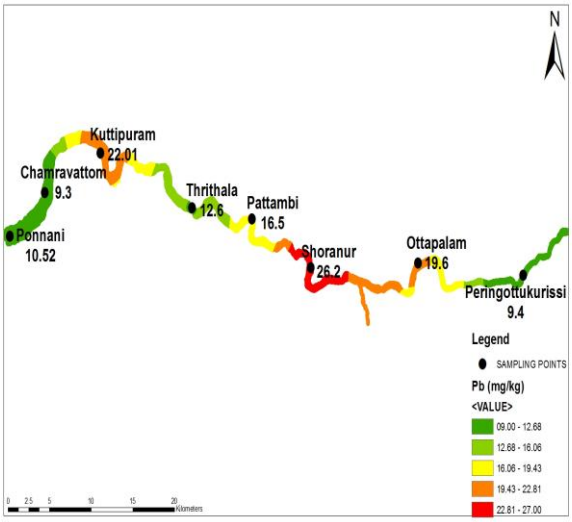

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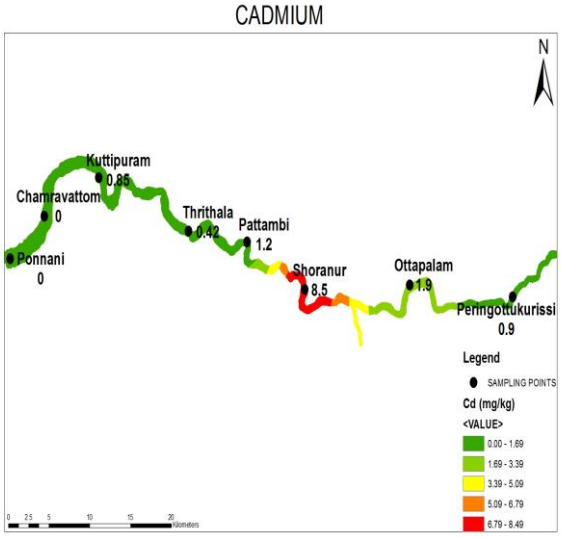

(f)

Fig.8. Spatial distribution of (a) Fe (b) Mn (c) Zn (d) Cu (e) Pb (f) Cd in sediment samples of Bharathapuzha

G. *Sediment quality guidelines*

TABLE X
CONTAMINATION STATUS OF METALS IN SEDIMENTS OF BHARATHAPUZHA

Metals	Average values of contamination of metals(mg/kg)	Contamination status
Fe	7509	Non-polluted (<17000)
Mn	31.391	Non-polluted (<300)
Zn	14.648	Non-polluted (<90)
Cu	9.425	Non-polluted (<25)
Pb	15.766	Non- polluted (<40)
Cd	2.295	Non-polluted (<6)

As per TABLE II, contamination status of metals in Bharathapuzha are shown in TABLE Sediments in Bharathapuzha were non-polluted for Fe, Mn, Zn, Cu, Pb and Cd.

H. Correlation study of metals and organic carbon

The correlation coefficient matrix of heavy metals and organic carbon is given in TABLE XI. The correlation coefficient matrix showed strong positive correlation between OC and metals. Fe showed significant positive correlation with Mn (r=0.953), Zn(r=0.998), Cu(r=0.942), and Cd(r=0.988). Mn exhibited strong positive correlations with Zn(r=0.957), Cu(r=0.884) and Cd(r=0.910), suggesting they probably originated from some common sources. Other highly correlated pairs include Zn with Cu and Cd, and Cu with Cd.

TABLE XI
CORRELATION COEFFICIENT MATRIX SHOWING INTER-ELEMENT AND ELEMENT -ORGANIC CARBON RELATIONSHIPS IN SEDIMENTS

Variables	Fe	Mn	Zn	Cu	Pb	Cd	OC
Fe	1	-	-	-	-	-	-
Mn	0.953	1	-	-	-	-	-
Zn	0.998	0.957	1	-	-	-	-
Cu	0.942	0.884	0.923	1	-	-	-
Pb	0.700	0.627	0.684	0.720	1	-	-
Cd	0.988	0.910	0.982	0.948	0.760	1	-
OC	0.844	0.737	0.821	0.881	0.710	0.896	1

I. *Principal component analysis of metals*

PCA extracted two factors and the principal component loadings of the heavy metals in the sediments are given in TABLE XII. Sum of first two factors accounted for 96.963 % of the variance of the sediment data. Factor 1 was dominated by all the metals evaluated namely, Fe, Mn, Zn, Cu, Pb and Cd and accounted for 89.267 % of the total variance. PC2(7.696% variance) is manifested by the prominent loadings of Pb only.

TABLE XII
COMPONENT LOADINGS OF HEAVY METALS IN SEDIMENTS

Variables	Component	
	PC1	**PC2**
Fe	0.990	-0.117
Mn	0.947	-0.208
Zn	0.984	-0.140
Cu	0.960	-0.024
Pb	0.781	0.620
Cd	0.990	-0.010
% of Variance	89.267	7.696

J. *Geo-accumulation index*

Muller's geo-accumulation index was used to quantify metal pollution in sediments. According to Muller scale, the calculated results of I_{geo} values indicate that for Cd, sediment quality is considered as moderately polluted ($1 \leq I_{geo} \leq 2$) (Class 2) while for Fe,Mn,Zn,Cu and Pb ,sediment quality was recorded unpolluted ($I_{geo} \leq 0$)(Class 0) . On the basis of the mean values of I_{geo} sediments are enriched for metals in the following order: Cd>Pb>Cu>Fe >Zn>Mn.

K. *Contamination factor, degree of contamination and pollution load index*

Samples collected from Ponnani, Chamravattom, Kuttipuram, Thrithala, Pattambi and Peringottukurissu showed low degree of contamination (C_d <7). whereas samples collected from Ottapalam showed moderate degree of contamination($7<C_d<14$). Very high degree of contamination was observed in Shoranur($C_d>28$).
The PLI values showed high pollution loads in sediments collected from Shoranur (PLI >1). Waste from the Shoranur railway station dumped at the railway yard is allegedly polluting the Bharathapuzha. Roads were cut through the riverbed in Shoranur recently to smuggle the 40-ft high heap of sand, costing crores of rupees, mined from the river during the construction of the Shoranur-Cheruthuruthy check dam. Leaching of metals into water bodies from urban, agricultural, industrial runoffs etc add to the problem. Strong steps must be taken against industrial units which are exploiting river and polluting it.

IV.CONCLUSIONS

Dying river Bharathapuzha, also known as "River Nila", is fast becoming a source of health hazards for the people of several panchayaths and municipalities on its banks. Years of steady dumping of effluents, pollutants and waste into the river from human habitats on both banks have contaminated Bharathapuzha, which is already in the process of a cruel death due to incessant and unscrupulous sand-mining and encroachments. All the collected samples of Bharathapuzha were subjected to phosphorus fractionation and heavy metal analysis. Multivariate statistical techniques were done to evaluate and characterize the analytical data. Creation of spatial distribution maps of phosphorus fractions and heavy metals using ArcGIS 10.3.1 software helped to identify the pollution sources and vulnerable sites. Most abundant P fraction was organic phosphorus with an average value of 7.84 mg/kg. Metal concentrations were generally low in sediments of Bharathapuzha when compared to Chalakudy and Periyar river, except in Shoranur region (sampling station-4) (PLI -1.104). On an average basis, the metals follow a decreasing order: Fe >Mn>Pb>Zn>Cu> Cd. Based on I_{geo} values Bharathapuzha river sediments were unpolluted for Fe,Mn,Zn,Cu and Pb (Class 0) and moderately polluted for Cd with I_{geo} value of 1.591(Class 2).A poisoned river means a dying population. Kerala has failed to assess the socioeconomic and environmental impact of the pollution of its rivers. Various sources of heavy metals should be closely monitored and discharge of industrial effluent and domestic sewage discharge should be reduced.

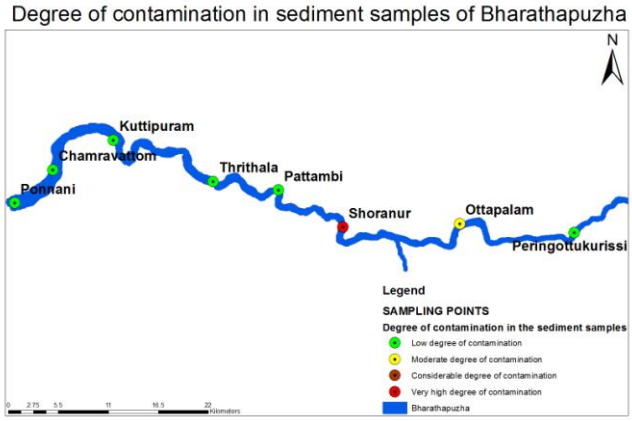

Fig.9. Degree of contamination in sediment sampling stations of Bharathapuzha

TABLE XIII
GEO-ACCUMULATION INDEX VALUES FOR THE SEDIMENT SAMPLES OF BHARATHAPUZHA

SI.No	Sampling stations	Fe	Mn	Zn	Cu	Pb	Cd
1	**Ponnani**	-6.612	-5.393	-7.306	-3.516	-1.511	0
2	**Chamravattom**	-5.123	-5.918	-8.891	-3.354	-1.689	0
3	**Kuttipuram**	-6.005	-6.153	0	-4.041	-0.446	0.917
4	**Thrithala**	-6.057	-5.560	0	-3.698	-1.251	-0.099
5	**Pattambi**	-3.571	-5.723	-3.906	-3.376	-0.862	1.415
6	**Shoranur**	-0.703	-3.781	-0.979	-1.228	-0.195	4.239
7	**Ottapalam**	-3.792	-6.209	-5.306	-2.199	-0.614	2.078
8	**Peringottukurissi**	-5.824	-7.558	-6.776	-4.228	-1.674	1
	Average	-4.710	-5.786	-5.527	-3.205	-1.030	1.591

TABLE XIV
CONTAMINATION FACTOR, DEGREE OF CONTAMINATION AND POLLUTION LOAD INDEX OF SEDIMENTS FROM BHARATHAPUZHA

SI No.	Sampling stations	Contamination factor(C^i_f)						Degree of contamination(C_d)	PLI
		Fe	Mn	Zn	Cu	Pb	Cd		
1	**Ponnani**	0.015	0.035	0.009	0.131	0.526	0	0.716	0.050
2	**Chamravattom**	0.043	0.024	0.003	0.146	0.465	0	0.681	0.046
3	**Kuttipuram**	0.023	0.021	0	0.091	1.100	2.833	4.068	0.168
4	**Thrithala**	0.022	0.031	0	0.115	0.63	1.4	2.198	0.147
5	**Pattambi**	0.126	0.028	0.1	0.144	0.825	4	5.223	0.234
6	**Shoranur**	0.921	0.109	0.760	0.64	1.31	28.333	32.073	1.104
7	**Ottapalam**	0.108	0.020	0.037	0.326	0.98	6.333	7.804	0.233
8	**Peringottukurissi**	0.026	0.007	0.013	0.08	0.47	3	3.596	0.080

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