Universal Journal of Environmental Research and Technology
 eISSN 2249 0256
Available Online at: www.environmentaljournal.org
Volume 2, Issue 5: 402-410

Open Access **Research Article**

Characterisation of Metals in Water and Sediments of Subarnarekha River along the Projects' Sites in Lower Basin, India

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Abstract:

The Subarnarekha River is one of the most important east flowing rivers of India. The river in West Bengal and Odisha is site for the Subarnarekha Barrage Project and Kirtania port respectively. The major objectives of the present study were to investigate the pre-projects level of heavy metals in water and sediments of the Subarnarekha River along the projects' sites. Water and Surface sediments collected from six locations were analysed for Fe, Zn, Cu, Pb, Ni, Cd, Mn and Cr with Atomic Absorption Spectrophotometer. Contamination factor, Contamination degree, Pollution load index (PLI) and Geo-accumulation index (I_{geo}) were used to assess the degree of accumulation of heavy metals in the sediments. The important findings of the study are: (1) Ficklin plot classified all water samples into near neutral low metal class; (2) A close relationship was obtained between organic carbon and metal content in river bed sediments; (3) All sampling sites displayed PLI values ≥ 0 but < 1; (4) I_{geo} values for metals at the sampling sites were ≤ 0. The results indicated river water and bed sediments to be unpolluted and ecologically suitable and sustainable. Absence of major anthropogenic influence in the region was primarily found to be responsible for the unpolluted nature of water and sediments. This study can be used as reference to monitor the status of quality of water and sediments during ongoing projects and post-projects scenario.

Keywords: Box and whisker plot, Contamination factor, Ficklin plot, Geo-accumulation index, Heavy Metals, Pollution load index, Sediment contamination

1. Introduction:

Industrial activities, domestic wastes, urbanisation and land development all contribute to the heavy metal pollution of rivers. The identification and quantification of the heavy metal in water and sediments are important environmental issues (Addo *et al.,* 2011). Presence of heavy metals in water and sediments has attracted researchers because of their persistence nature, toxicity and bio-availability. The metals in the bio-available form can enter the food chain and produce a range of metabolic and physiological disorders. Metals like Hg, Pb and Cd are toxic even at minute concentrations. Though metals like Fe, Zn and Mn are needed for the normal development of living beings they can be detrimental above their threshold values. Metals released into aquatic environments become bound to the particulate matters which in due course settle down and become part of the sediments (Suthar *et al.,* 2009). Heavy metals adsorbed to the sediments have the potential to remobilise under altered environmental conditions like change in pH or organic matter content. Thus, sediments which act as sink of heavy metals can become immediate source of metal pollution of the water bodies. Regular environmental monitoring and assessment of aquatic resources with respect to heavy metals is necessary to study the impact of developmental projects and programmes on their water and sediment quality. Water and sediments both act as habitats of aquatic organisms. Any alteration in the quality of water and sediments is definitely going to affect the aquatic organisms. This may start sequential actions ultimately affecting all forms of life including human beings.

The Subarnarekha River is one of the most important intestate rivers of India having a total catchment area of 19,236 km^2. It flows through three Indian states viz. Jharkhand, West Bengal and Odisha before draining into the Bay of Bengal. A few studies have been undertaken to investigate the pollution of the Subarnarekha River towards its eastern and coastal sides (Senapati and Sahoo, 1996; Panda *et al.,* 2006). Two important developmental projects along the River are in the planning stage. According to the Subarnarekha Barrage Project, a barrage is to be constructed near Bhasraghat (West Medinipore district, West

Bengal) to irrigate 1,14,198 ha of land annually through a left bank canal and its distribution system covering a cultural command area of 96,860 ha (http://pib.nic.in/newsite/...33636). A deep water; all weather port at Kirtania (Balasore district, Odisha) is also being planned and finalised by the Government of Odisha (http://www.orissadiary.com/ShowBussinessNews =3556). Environmental monitoring and assessment of freshwater resources provide feedback on the status of their quality degradation based on performance of developmental programmes and projects. Based on these important developments, the major objective of the present study was to investigate the most recent occurrence and distribution of some heavy metals in water and sediments of the Subarnarekha River along the projects' sites in West Bengal and Odisha during pre-projects period. This study can be used as reference to monitor the status of quality of water and sediments of the Subarnarekha River during ongoing projects and post-projects scenario.

2. Materials and Methods:

2.1 Study Area

The Subarnarekha River towards its sub-lower and lower reaches flows through West Medinipore district (21°36' to 22°57' N and 86°33' to 88°11' E, undivided Medinipore) in West Bengal and Balasore district (21°03' to 21°59' N and 86°20' to 87°29' E) in Odisha (Figure a). In West Medinipore Subarnarekha River flows through the upland region of the western portion of the district which is a part of the Chottanagpur plateau covered with hard lateritic stones. The eastern portion is a part of the coastal plain and is formed of alluvial soil (http://paschimmedinipur.gov.in). Balasore district is a part of the eastern coast and is situated on the northern most part of the Odisha state. Bay of Bengal lies towards its east and West Medinipore on its north. Subarnarekha River in Odisha flows from west to east through the inner alluvial plain and the coastal belt before draining into the Bay of Bengal (http://baleswar.nic.in). The climate in this part of India is mostly hot and humid. Major industrial activities are lacking in these regions.

2.2 Sampling Procedure and Preservation

Water and sediment samples were collected from six sites during December, 2011 period. Gopiballabhpur (situated upstream of Bhasraghat in West Bengal) and Kirtania (lying upstream of Odisha coast) were selected as first and sixth sites respectively (Figure a). Water samples were collected at 10-15 cm depth in pre-conditioned and acid rinsed clean polypropylene bottles (Ahdy and Khaled, 2009). The samples were immediately acidified with concentrated nitric acid to a pH below 2.0 to minimise precipitation and adsorption onto container walls (APHA, 2005). Surface sediment samples were taken at a depth of about 5 cm and immediately transferred into pre-cleaned polythene bags. The collected samples were oven dried at 40°C for 48 hours, homogenised, sealed in clean polythene bags and then stored at 4°C for further processing (Yongming *et al.,* 2006; Suthar *et al.,* 2009).

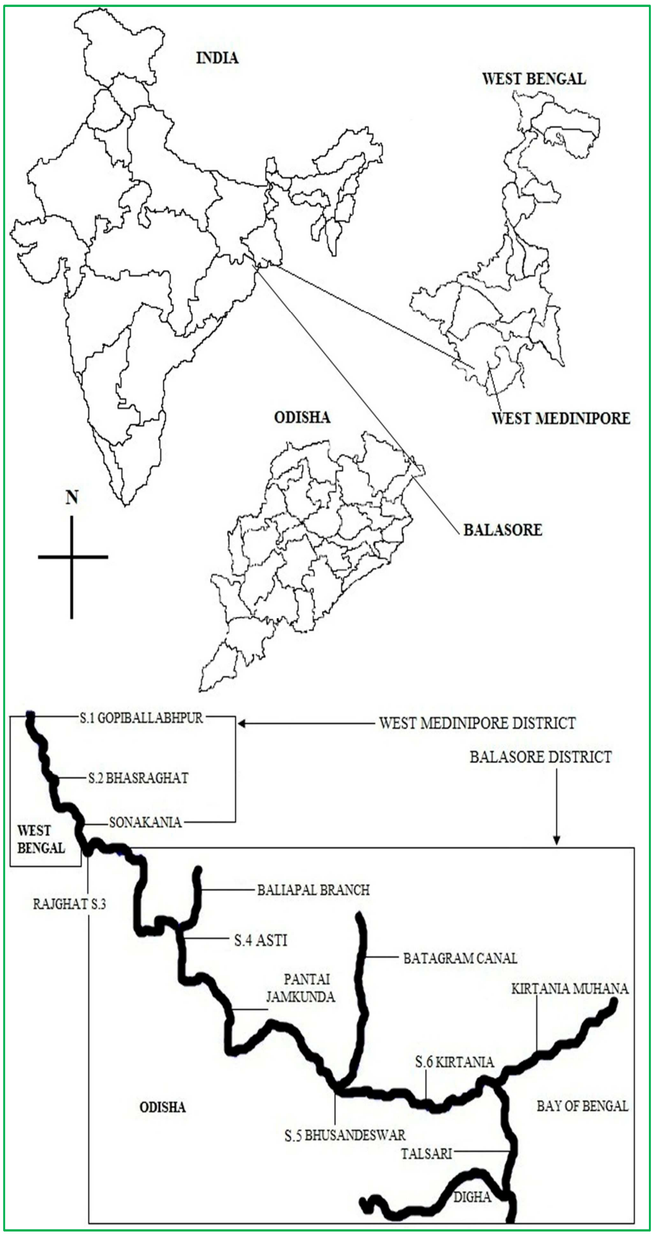

Figure a: Study area and sampling sites

2.3 Sample Analyses

The pH of the water samples was measured at the sampling sites with a digital pH meter (pHTestr 30, Eutech instruments, Oaklon). Organic Carbon (OC) in the sediments was determined by partial oxidation method of Walkley and Black (1934). For the determination of total heavy metals in the samples, extraction procedures as described in APHA (2005) were followed. Hot plate digestion of water and sediment samples was carried out with tri-acid Nitric acid-Sulphuric acid-Perchloric acid (10 part HNO_3 + 1 part H_2SO_4 + 4 part $HClO_4$) mixture. The digested samples were filtered through Whatman No. 42 filters and made up to 25 ml by adding distilled water in a volumetric flask. Heavy metal concentrations (Fe, Zn, Cu, Pb, Cd, Ni, Cr and Mn) were determined using Atomic Absorption Spectrophotometer (GBC Avanta Ver 1.33). Only chemicals and reagents of analytical grades were used throughout the experiment. All glassware were washed with 14% HNO_3 and rinsed thoroughly with double distilled and deionised water prior to use. Only double distilled and deionised water was used for the study.

2.4 Statistical Analyses

Box and whisker plots were used to display the spatial variation and range of heavy metal concentrations. The middle horizontal line of the box represents the median value. Fifty percent of the data points lie within the box. The whiskers show the spread of data and a closed circle represents outliers (Chapman *et al.,* 1999). Pearson correlation coefficient was used to evaluate the strength of association between heavy metals. Regression coefficient was also calculated between heavy metals and OC (%) in sediment samples.

2.5 Determination of Contamination Factor (CF) and Contamination Degree (CD)

CF for each metal was determined as described below

$$CF = \frac{Observed\ metal\ concentration}{Background\ concentration\ of\ the\ same\ metal}$$

CD for each site was calculated as sum of all contamination factors (Ahdy and Khaled, 2009).

2.6 Determination of Pollution Load Index (PLI)

Pollution load index for each site was determined following the method proposed by Tomlinson *et al.,* (1980).

$$PLI = \sqrt[n]{(CF_1 \times CF_2 \times CF_3 \times \ldots\ldots\ldots\ldots\ldots\ldots \times CF_n)}$$

Where, CF is the contamination factor and n is the number of parameters.

2.7 Determination of Geo-Accumulation Index (I_{geo})

The I_{geo} values for different metals were determined following Müller (1979).

$$I_{geo} = log_2 \frac{C_n}{1.5 \times B_n}$$

Where, C_n is the observed concentration of the metal (n) in the sediment, B_n is the geochemical background of the metal (n), 1.5 is the correction factor for the background matrix due to lithogenic effects. Average shale standard for different metals reported by Turekian and Wedepohl (1961) was taken as background concentration throughout the study.

3. Results and Discussion:

3.1 Metals, pH and OC

Concentrations of eight studied heavy metals in water and sediments recorded at different sites along the Subarnarekha river are displayed in Table 1. The concentration of Fe was much higher at most of the locations as compared to other metals in both water and sediments. Based on concentration range and abundance heavy metals in water and sediments can be ranked as Fe > Mn > Zn > Cu > Pb > Ni and Fe > Ni > Mn > Zn > Pb > Cu respectively. One interesting point to notice is that though the concentration of Fe fluctuated in river water, it remained relatively stable in bed sediments. The behaviour of metals displayed similar pattern in both water and sediments. pH ranged from 7.21 to 7.40 showing relatively alkaline nature of water. OC varied from 0.17 to 0.81%.

3.2 Ficklin Plot

Cationic species tend to be less mobile under high pH conditions and more mobile under low pH conditions. Since, predominant charge on most of the cations (Zn, Cu, Pb, Cd, Mn) is +2, their mobility and bioavailability is enhanced under low pH (acidic) conditions (Smith, 2007). The Ficklin plot is a scatter diagram in which the sum of the base metals is plotted against pH (Ardejani *et al.,* 2011). Ficklin plot is an easy and effective mean of classifying water samples based on differences in the sum of base metal concentrations. In the present study Zn, Cu, Pb, Cd and Mn were selected for the construction of Ficklin plot rather than more commonly occurring metal Fe. Value of Cd was taken as 0 as it was not detected in water samples. Differences in the sum of base metal concentrations allow differentiating between different geological controls on water composition and chemistry (Kelepertzis *et al.,* 2010). The sum of base metals (Zn, Cu, Pb, Cd and Mn) plotted as a function of pH is displayed in Figure b. The Ficklin plot classified all water samples as near neutral low metal waters. The pH of the Subarnarekha River was found to be alkaline (7.21 – 7.40). Sum of base metals were higher at S.2, S.3 and S.6 and lowest sum of base metals was recorded at S.1.

Table 1: Concentrations of metals in water (mg/l) and sediments (mg/kg), pH and OC (%)

Parameters	Sampling sites						
	S.1	S.2	S.3	S.4	S.5	S.6	Shale standard#
Fe (mg/l)	0.006	8.105	7.814	6.475	0.002	11.319	
Fe (mg/kg)	516.485	632.965	617.035	640.110	643.955	687.410	47200
Zn (mg/l)	0.005	0.215	0.159	0.102	0.014	0.096	
Zn (mg/kg)	6.155	5.110	4.930	8.295	16.335	17.695	95
Cu (mg/l)	0.021	0.003	0.068	0.054	0.038	0.021	
Cu (mg/kg)	0.345	0.305	1.180	1.840	3.295	4.175	45
Pb (mg/l)	0.012	0.039	0.028	0.023	0.019	0.015	
Pb (mg/kg)	3.390	4.620	5.340	7.915	6.880	8.745	20
Ni (mg/l)	0.007	0.018	0.021	0.015	0.013	0.016	
Ni (mg/kg)	16.360	23.380	21.575	26.690	29.385	37.540	68
Cd (mg/l)	ND	ND	ND	ND	ND	ND	
Cd (mg/kg)	ND	ND	ND	ND	ND	ND	0.3
Mn (mg/l)	0.004	0.313	0.249	0.151	0.028	0.454	
Mn(mg/kg)	18.605	67.84	71.990	85.890	130.08	161.205	850
Cr (mg/l)	ND	ND	ND	ND	ND	ND	
Cr (mg/kg)	ND	ND	ND	ND	ND	ND	90
pH of water	7.40	7.28	7.21	7.38	7.30	7.26	
OC (%)*	0.17	0.32	0.36	0.61	0.81	0.53	

* and # for sediments samples; # Turekian and Wedepohl (1961); ND = not detected

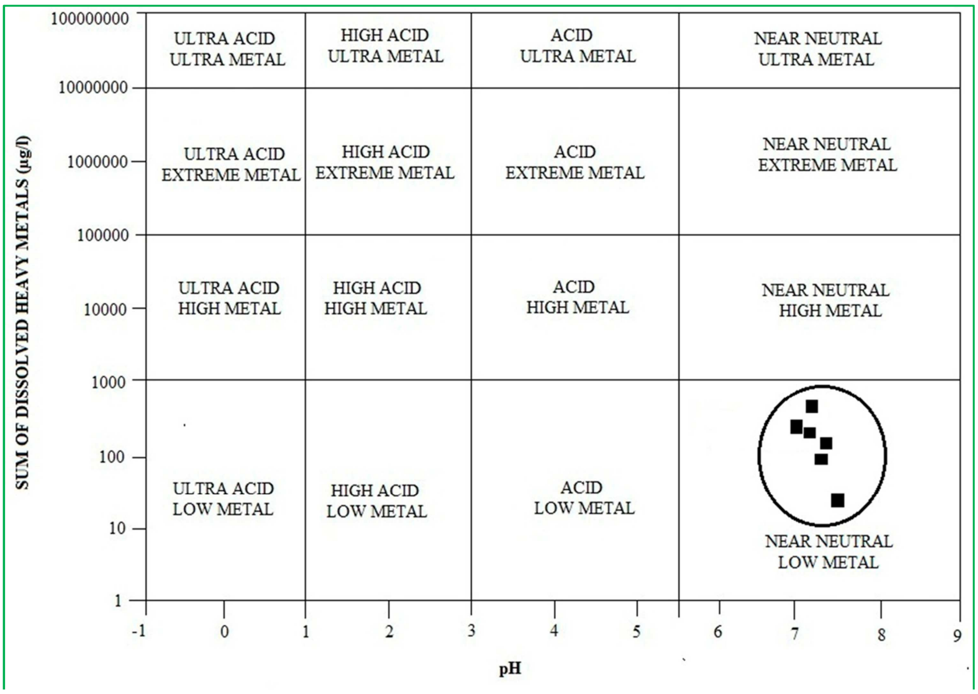

Figure b: Ficklin plot displaying sum of dissolved metals as a function of pH (Modified from Ardejani *et al.,* 2011)

3.3 Statistical Analyses

Box and whisker plots summarising concentrations of metals in water and sediments of the Subarnarekha River are presented in Figure c. It is observed from these plots that most of the metals displayed variations in their presence in water and sediments. Correlation analysis between metals in water samples (Table 2) showed very strong relationship between Fe-Ni-Mn and Ni-Mn. Strong association was also noted between Zn-Cu and Zn-Ni. Pb was found to be interrelated with Ni. It is very interesting to note in correlation analysis of sediments (Table 3) that all metals showed strong association with each other. Heavy metals in environment usually have complicated relationships among them (Li *et al.,* 2012). Heavy metals showing very high correlation may indicate same origin and controlling factors (Rafiei *et al.,* 2010).

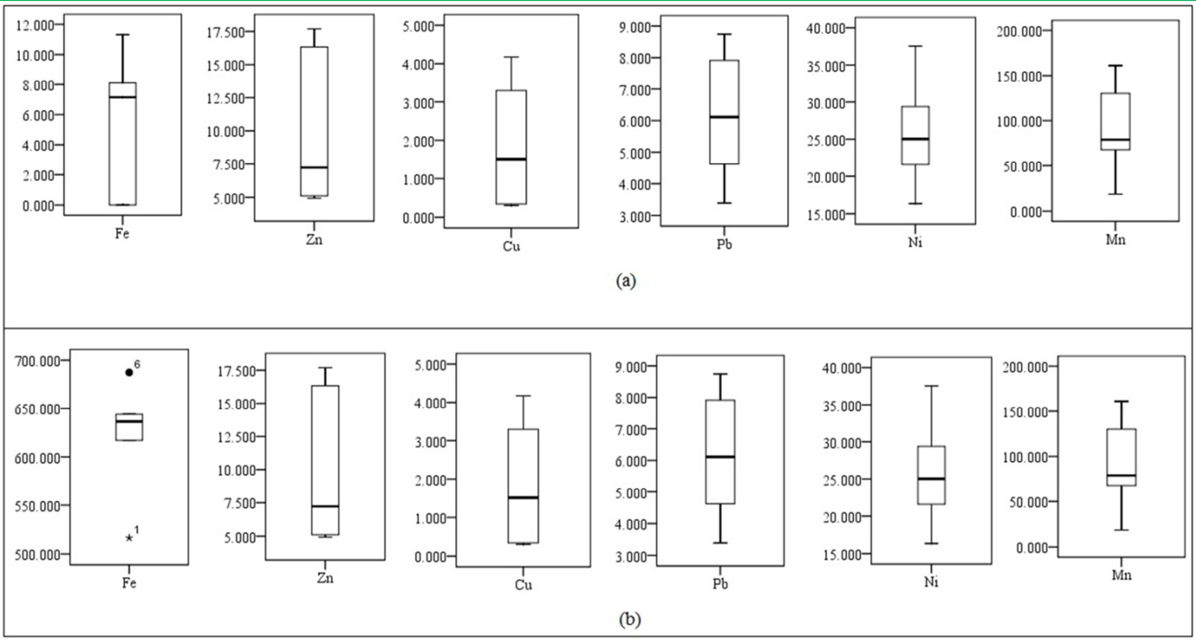

Figure c: Box and whisker plots for different metals; (a) = river water, (b) = river sediments

Table 2: Correlation coefficients between metals in river water

		Fe	Zn	Cu	Pb	Ni	Mn
Fe	Correlation	1	0.350	-0.016	0.393	0.746	0.962
	significance		0.496	0.976	0.441	0.089	0.002
Zn	Correlation		1	0.625	0.408	0.719	0.253
	Significance			0.185	0.422	0.107	0.629
Cu	Correlation			1	-0.081	0.339	-0.205
	Significance				0.879	0.511	0.697
Pb	Correlation				1	0.709	0.337
	Significance					0.115	0.514
Ni	Correlation					1	0.677
	Significance						0.140
Mn	Correlation						1

Table 3: Correlation coefficients between metals in river sediments

		Fe	Zn	Cu	Pb	Ni	Mn
Fe	Correlation	1	0.600	0.724	0.847	0.891	0.895
	significance		0.208	0.104	0.003	0.017	0.016
Zn	Correlation		1	0.957	0.741	0.864	0.886
	Significance			0.003	0.092	0.027	0.019
Cu	Correlation			1	0.869	0.923	0.947
	Significance				0.025	0.009	0.004
Pb	Correlation				1	0.911	0.879
	Significance					0.012	0.021
Ni	Correlation					1	0.975
	Significance						0.001
Mn	Correlation						1

We also correlated the OC% values with metal concentrations determined at different sampling sites. The investigation revealed all metals under study displaying a close relationship with OC%

(Figure d). Similar results have also been reported by Suthar *et al.,* (2009) for the Hindon River in India. Presence of organic matter can influence the accumulation of heavy metals in the bed sediments (Suthar *et al.,* 2009; Mohiuddin *et al.,* 2010; Iwuoha *et al.,* 2012) as organic carbon can be bounded with some metal fractions in tight forms (Suthar *et al.,* 2009). Our study further strengthens above arguments that organic matter load can increase metal enrichment in river sediments and under changed environmental conditions these bound metals can become bio-available to enter the food chain.

3.4 Contamination Factor and Contamination Degree

Contamination factor and Contamination degree are used to assess the pollution load of the sediments with respect to heavy metals. CF values for heavy metals recorded at different sampling locations are presented in Table 4. Hakanson (1980) has provided four grade ratings of sediments based on CF values. All sites along the Subarnarekha's lower plain displayed low CF (CF < 1, Class I) indicating the unpolluted nature of sediments. CD is simply the sum of all CF values of a particular site. Ahdy and Khaled (2009) described CD in terms of four classes. It is noted from the determined CF values that CD in bed sediments ranged from 0.516 to 1.472 (Table 4) which further indicate the unpolluted nature (CD < 6, Class I) of Subarnarekha sediments towards its lower plains.

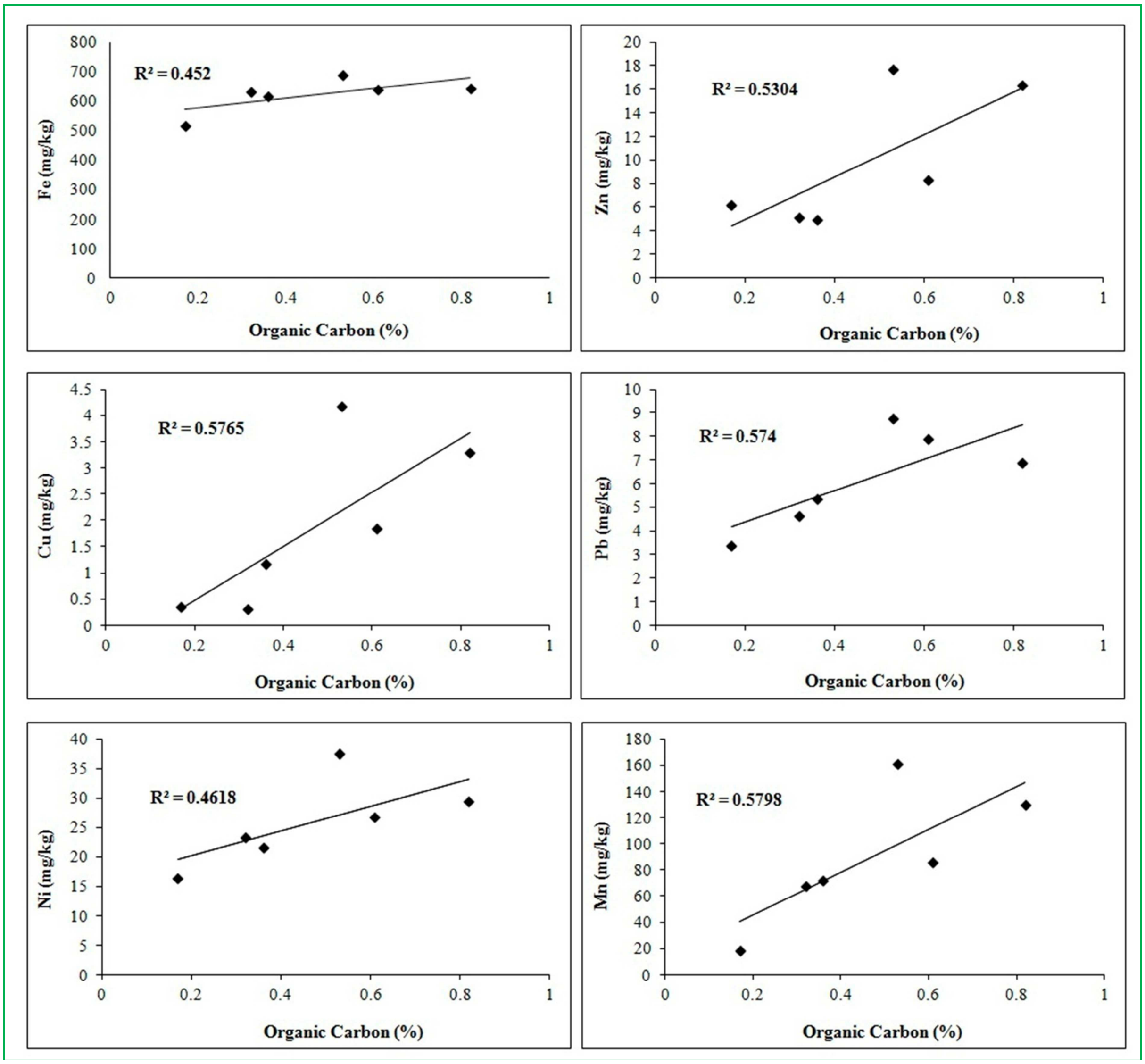

Figure d: Plots displaying correlation between different metals and organic carbon (%)

Table 4: Contamination factor, contamination degree and PLI values

Parameters	Contamination factor S.1	S.2	S.3	S.4	S.5	S.6
Fe	0.011	0.013	0.013	0.014	0.014	0.015
Zn	0.065	0.054	0.052	0.087	0.172	0.186
Cu	7.67×10^{-3}	6.78×10^{-3}	0.026	0.041	0.073	0.093
Pb	0.169	0.231	0.267	0.396	0.344	0.437
Ni	0.241	0.344	0.317	0.393	0.432	0.552
Mn	0.022	0.079	0.085	0.101	0.153	0.189
Contamination degree	0.516	0.728	0.760	1.032	1.188	1.472
PLI	0.041	0.056	0.071	0.096	0.125	0.151

Table 5: Geo-accumulation index (Igeo) values for the Subarnarekha river sediments

Sampling sites	Geo-accumulation index values Fe	Zn	Cu	Pb	Ni	Mn
S.1	-7.099	-4.434	-7.612	-3.146	-2.640	-6.098
S.2	-6.806	-4.802	-7.790	-2.699	-2.125	-4.232
S.3	-6.843	-4.854	-5.838	-2.489	-2.241	-4.147
S.4	-6.789	-4.103	-5.197	-1.922	-1.934	-3.892
S.5	-6.781	-3.126	-4.357	-2.124	-1.796	-3.293
S.6	-6.687	-3.010	-4.015	-1.778	-1.442	-2.984

Table 6: Index classifications of sediment quality

CF values Hakanson (1980)	Class	Sediment quality
CF < 1	1	Low CF
1 ≤ CF < 3	2	Moderate CF
3 ≤ CF < 6	3	Considerable CF
CF ≥ 6	4	Very high CF

CD values Ahdy and Khaled (2009)	Class	Sediment quality
CD < 6	1	Low CD
6 ≤ CD < 12	2	Moderate CD
12 ≤ CD < 24	3	Considerable CD
CD ≥ 24	4	Very high CD

I_{geo} values Müller (1981)	Class	Sediment quality
≤ 0	0	Unpolluted
0-1	1	Unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	Moderately to strongly polluted
3-4	4	Strongly polluted
4-5	5	Strongly to extremely polluted
≥ 6	6	Extremely polluted

3.5 Pollution Load Index

Pollution load index is an effective instrument for assessing and comparing the sediment quality. According to Mohiuddin *et al.,* (2010) PLI = 0 indicates perfection; PLI = 1 points to only baseline levels of pollutants present and PLI values > 1 represent progressive deterioration of sites. All sites display PLI values close to zero (Table 4) reflecting unpolluted nature of the sediments. This study further show both CD and PLI complement each other and can be used in combination to evaluate the environment of sediments. These indices are easy to understand and can help the public and the policy makers for ascertaining the pollution load of an area to take necessary remedial measures.

3.6 Geo-Accumulation Index

Geo-accumulation index consists of a seven grade classification system of sediments from unpolluted to extremely polluted (Müller, 1981) nature. Class 6 reflects 64 fold enrichment above the background value (Singh *et al.,* 2005). I_{geo} values obtained for metals in sediments are presented in Table 5. All sampling sites display I_{geo} values < 0 for each heavy metal taken in this study suggesting the sediment quality is unpolluted ($I_{geo} \leq 0$, Class 0) for the region. Classification systems described for sediment quality by different authors is provided in Table 6.

4.0 Conclusions:

Ficklin plot classified the Subarnarekha River water along the projects' sites into near neutral low metal category. All sites displayed CF < 1 (Class I) and CD < 6 (class I). The river bed further showed PLI values ≥ 0 but < 1. I_{geo} value < 0 (class 0) was noted for all metals. These arguments clearly indicate the quality of the Subarnarekha river water and bed sediments along the projects' sites to be unpolluted and ecologically suitable and sustainable. Subarnarekha River basin in its sub-lower and lower reaches is characterised by the absence of major industrial activities and populated urban centres as compared to upstream regions like Ranchi, Jamshedpur and Moubhandar-Ghatsila in Jharkhand state. Absence of major anthropogenic influence suggests geogenic activities to be principally responsible for the presence of heavy metals in the river water and bed sediments. Continuous monitoring and assessment will keep checking the pollution status of the river water during and after completion of the projects. This study can be used as reference to monitor the status of quality of water during ongoing projects and post-projects scenario.

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