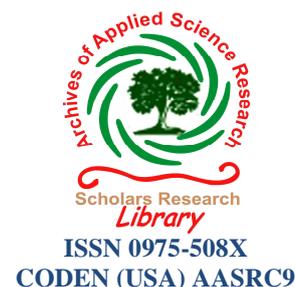




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## Evaluation of Metal Contamination in Freshly Deposited Sediment of Hugli Estuary, India

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### ABSTRACT

Metal contamination in sediment of Hugli estuarine system has often been assessed using datasets that provide dramatic over – or underestimate of actual status of sediment contamination due to lack of stipulated guidelines for collecting sediment with respect to grain sizes. This study has been undertaken to present how the level of metal contamination in sediment can be evaluated. The analytical results focus spatial and temporal conditions of metals in 200-  $\mu\text{m}$  and 63- $\mu\text{m}$  sediment fractions. To better elucidate actual degree of metal pollution, contamination sources, and geochemical processes; statistical analysis are employed to intensive datasets collected from Hugli estuary that discharges into the largest alluvial fan in the world. Multivariate analysis including hierarchical cluster analysis clearly identified influence of anthropogenic and natural activities on metal contamination and geochemical processes controlling level of metal contamination in sediment of Hugli estuary. Normalization of Al indicated relatively high enrichment factors for Pb, Ni, Cu, Co, Cd, Cr and Fe suggesting contamination of these metals due to anthropogenic activities. Strong seasonal change, variable tidal energy level and irregular estuarine geometry in the study area played crucial role in governing metal concentration in freshly deposited sediment. This paper sheds some lights on necessity of refining existing monitoring practice and its importance in estuarine environment management.

**Key words:** Estuary; Metal pollution; sediment; multivariate analysis; monitoring practice.

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### INTRODUCTION

The Hugli estuary supports the world's largest mangrove system, the Sundarbans and is identified as positive estuary in mixohaline region. This complex environment suffers from intense anthropogenic activities that alter estuarine ecosystem mainly due to uncontrolled discharge of persistent pollutants particularly metals to this estuary. Study for better understanding of metal distribution in this estuarine sediment is a major concern in evaluating the effect of anthropogenic influences. Sediments are important carriers of metals in the hydrological cycle and because metals are partitioned with the surrounding areas, they reflecting the quality of aquatic ecosystem [1]. Coastal and estuarine region are the important sinks for metals and that are accumulated in living organisms and bottom sediments [2]. Heavy metals accumulate in the sediments through complex physical and chemical adsorption mechanisms depending on the nature of the sediment matrix and the properties of the adsorbed compounds [3-5]. A high affinity of the metals with organic matter, metal oxides, and clay minerals helps to accumulate them in benthic sediment effectively over time. Their occurrence in the environment results primarily from natural process, such as weathering of rocks and anthropogenic activities[6-8]. Sediments are normally mixtures of several components

including different mineral species as well as organic debris. Sediments represent one of the ultimate sinks for heavy metals discharged into the environment [9-11]. Last few decades the metal contamination of surface water from uncontrolled discharge of industrial waste and erosion of surface soil due to poor land use planning is a major environmental issue. The on-going anthropogenic activities in the upper catchment area cause accumulation of metals in this estuarine zone. The sources of metals in the tidal stretch of Hugli estuary, North-East coast of Bay of Bengal are numerous. The researches have been carried out so far to focus concentration, distribution and possible sources of metals in this estuarine zone restricted to the saline zone [12]. The study of the actual dynamics and possible sources of metals need adequate data and their interpretation for better understanding. Now it is an emergent need to understand the geochemical characteristics of the sediment and the dynamics of metals in freshly deposited sediment over the tidal stretch of Hugli estuary that discharges into the largest alluvial fan in the World.

This paper reports the chemical composition of freshly deposited sediment in this zone to evaluate the geochemical processes controlling the metal concentration including possible anthropogenic influences in response to spatial and temporal condition. This study will also focus the necessity of refining the existing monitoring of metal contamination in sediment.

### **Characteristics of basin and river Ganges**

The relevant information on geologic, geomorphic and hydrologic framework and evolution of the Ganga basin [13] are required for better understanding of the sources and behavior of metallic elements in sediment. The sediment load of the Hugli estuary system consists exclusively of fine sand, silt and clay at their lower reaches within the Bengal basin, and is deposited under uniformly fluctuating, unidirectional energy conditions. The sediments have a close similarity in grain size with the sediments of the surrounding floodplain. The Ganga basin has a good amount of smectite and a lower amount of kaolinite [14] derived from the low-temperature alteration of high-grade crystalline metasediments of the Himalayas by pedogenic processes within the Bengal basin [15,16]. The heavy minerals in the Ganga system are in the order of amphibole-garnet-epidote and dominated by unstable minerals [17,18]. Subramanian [19] recorded 46% illite, 28.7% kaolinite, 22.8% chlorite and 2.5% montmorillonite in the suspended sediments of the Ganges () at Calcutta (presently Kolkata). The high-grade metamorphic terrains are the provenance of 40–46% of heavy minerals, followed by igneous terrains with 21–29% attribution [17,18]. The Ganga is named as Hugli in West Bengal. The sediment load of Hooghly River originating from natural weathering products and from anthropogenic sources is estimated to be  $520 \times 10^6$  T  $\text{yr}^{-1}$  [20-21]. Abbas and Subramanian [22] calculated that at Kolkata (former Calcutta), the Ganga annually supplies  $411 \times 10^6$  T (i.e.,  $328 \times 10^6$  T Sediment +  $83 \times 10^6$  T solute load) of total load to the Hugli estuary.

The hydrodynamic study focused that by discharge ( $15646 \text{ m}^3 \text{ s}^{-1}$ ) Ganga is the fifth largest river in the World [20]. The Ganga enters the basin from the northwest after draining Himalayas and most of north India for about 2500 km. The river after bifurcation below Farakka, flow southwards down the deltaic plain of West Bengal as the river Hugli and then empties into Bay of Bengal. The hydraulic character of the Ganga suddenly changes on entry into the tidal zone of the gangetic delta. The tidal stretch of river Hugli is up to Triveni from the Sagar islands. The tidal influence varies depending on upland flow with maximum amplitude of 5.5 m. The Hugli estuary is a well mixed type and vertically homogenous throughout the year except with slight stratification for a short period during south west monsoon (June – September) due to fresh water discharge [23]. Discharges from Farraka Barrage are  $2975 \pm 1144 \text{ m}^3 \text{ S}^{-1}$  during monsoon with highest value ( $4000 \text{ m}^3 \text{ S}^{-1}$ ) during September,  $1000 \pm 81.6 \text{ m}^3 \text{ S}^{-1}$  during pre-monsoon with minimum value ( $900 \text{ m}^3 \text{ S}^{-1}$ ) during May; and  $1875 \pm 985.5 \text{ m}^3 \text{ S}^{-1}$  during post-monsoon [23]. Surface runoff ( $\text{Km}^3 \text{ month}^{-1}$ ) was reported to be 0.88, 4.02, 18.7, 8.47, 20.47 and 1.93 during May, June, July, August, September and October respectively [24]. The large tidal variation, irregular estuarine geometry, the presence of island and presence of navigation channel separated by shallow zone make the flow quite complicated.

In this soil, the values of total carbon (TC) were reported to be  $8.2 \pm 2.8 \text{ mg g}^{-1}$  and bears good correlation with grain size of the sediments [25]. Sarin *et al.* [26] reported dominance of cation load (Na, Ca) in Ganga sediments. The river waters in the basin are circum-neutral to slightly alkaline [13,27], indicating chemical evolution associated with mineral weathering. The chemistry suggests dominance of carbonate dissolution, with some input of calcic plagioclase and pyroxene weathering [27].

### **3.0 Study area**

The study includes 240-km-long tidal stretch of the river in West Bengal between the mouth of Bay of Bengal and confluence of Churni River (Figure. 1). Thirteen locations were selected (Figure. 1) along the study stretch. The

sampling locations selected in this study stretch are named as Sagar(G1), Auckland(G2), Mud Point(G3), Diamond Harbour(G4), Raichak(G5), Falta(G6), Uluberia(G7), Poojali(G8), Garden Reach(G9), Dakhineswar(G10), Palta(G11), Triveni(G12) and Churni(G13). The station G1 is located at the mouth of Bay of Bengal with maximum tidal influence and G13 at the extreme upstream with negligible tidal influence. All 13 stations are categorized into three groups based on sea water intrusion and tidal influence: i) G1-G5; ii) G6-G11; iii) G12-G13. The first group is located in the lower stretch which depicts sea water intrusion and maximum tidal influence; the second group indicated moderate tidal influence with no sea water intrusion and third group showed negligible tidal influence. The station G5 was included in the first group because of sea water intrusion during dry season. The sea water intrusion was investigated by the level of chloride concentration at all stations over the months. This estuary and its basin encompasses an immense and complex area with diverse river hydrology and basin lithology.

### MATERIALS AND METHODS

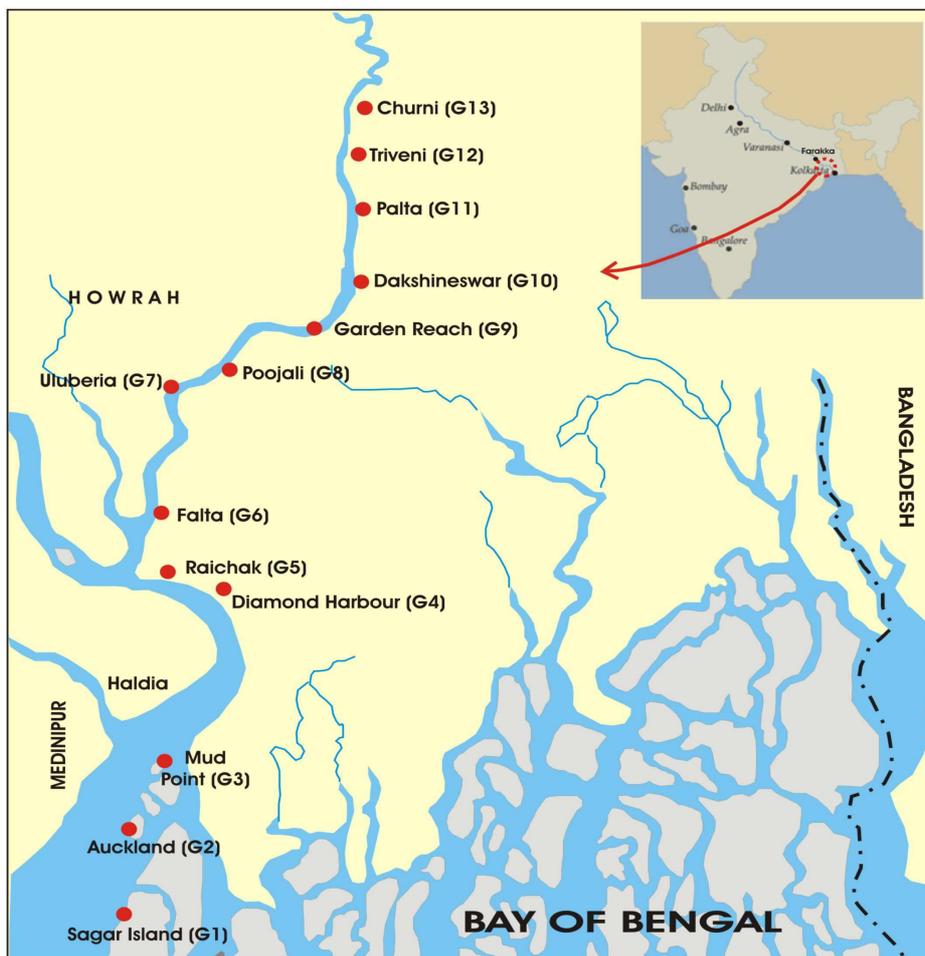
Sediment samples were collected in the period of August (to represent wet season) and February (to represent dry season) from 30 sampling sites of 13 locations of the Hugli Estuarine system in India. Samples were collected from left bank, right bank and mid bank of 13 locations. Samples could not be collected from all the mid banks of 13 sampling sites as collected materials were often full of sand. Sediment samples were collected from On-Board marine vessel (Meena M.V.) using Van-Veen Grab sampler. At each station five sediment samples were collected to maintain the accuracy of the findings. All the samples were stored separately in precleaned polyethylene bags and frozen in order to prevent changes in chemical composition of the sediment and then transported to the laboratory. Samples were air dried in a dust free environment and grounded with pestle and mortar and sieved through a 200- $\mu\text{m}$  sieve and 63- $\mu\text{m}$  sieve and then kept sealed in screw cap polyethylene bottles.

For measurement of total metals in sediment, samples were digested with sodium chloride and a mixture of nitric acid and Perchloric acid. Five samples were digested by Microwave digestion (CEM MDS-2000) using 5 ml aqua regia as per manufacturer's recommendation. The analyses of the entire pretreated sample were performed using Flame AAS of GBC (Avanta PM) equipped with a hydride vapor system. Calibration standards were prepared from multielement standard stock solutions. For minimizing interferences, we prepared a multi-element standard stock solution in which ratios of metals in this multiple element calibration standards were analogous to their ratios in the samples. These multi-element standards and blanks were prepared in the same matrix to minimize matrix effects and for background correction [28]. For Calibration, the standards traceable to NIST were used. Recalibration check was performed at regular interval. As a quality control, duplicate analyses were performed on five selected samples. The total metal concentrations showed good agreement with Microwave digestion method. The relative standard deviations of the means of duplicate measurement were less than 5%. The accuracy of the total metals was evaluated using the certified reference material (MBSS) prepared by Akademie der Wissenschaften der DDR, Institut für Meereskunde and certified by analyzing in 42 laboratories in 18 states.

The measurement uncertainties (MU) of all the metals were estimated taking into account all relevant sources of uncertainty during the analytical process starting from the preparation of standards to the analysis by AAS in accordance with guide to the expression of uncertainty in measurement [29]. The estimated values of MU ( $C_{\text{sample}} \pm UC_{\text{sample}}$ ) were  $8.4 \pm 0.34 \mu\text{g Pb g}^{-1}$ ,  $330 \pm 7.7 \mu\text{g Mn g}^{-1}$ ,  $1.4 \pm 0.24 \mu\text{g Cd g}^{-1}$ ,  $104 \pm 3.5 \mu\text{g Zn g}^{-1}$ ,  $2.30 \pm 0.34 \mu\text{g Hg g}^{-1}$ ,  $24000 \pm 84.1 \mu\text{g Fe g}^{-1}$ ,  $31400 \pm 75, 5 \mu\text{g Al g}^{-1}$ ,  $14 \pm 0.7 \mu\text{g Ni g}^{-1}$ ,  $22.1 \pm 0.78 \mu\text{g Cu g}^{-1}$ ,  $1.4 \pm 0.07 \mu\text{g As g}^{-1}$ ,  $24.5 \pm 1.7 \mu\text{g Cr g}^{-1}$ ,  $21 \pm .72 \mu\text{g Co g}^{-1}$ .

The levels of organic matter (Org-C) in each sample were determined using loss on ignition at  $555^{\circ}\text{C}$ . The calcimetry method [30] was used to determine the content of carbonate ( $\text{CaCO}_3$ ) in the sediments. In this method, the amount of  $\text{CO}_2$  released from the reaction is dependent on the amount of  $\text{CaCO}_3$  in the sample. The estimation of percentage of sand, silt and clay in sediments were carried out in accordance with method described by Gee and Bauder [31]. All results are expressed on dry-weight basis.

Figure - 1: Study Stretch of River Ganga



### Statistical Applications

In this study, statistical methods including mean, median, standard deviation (SD), coefficient of variation (CV), Pearson's Correlation Analysis, FA (Factor Analysis) and HCA (*Hierarchical Cluster Analysis*) were performed in order to obtain information about the relationships and behavior of the metals, and for the determination of the background concentrations. Multivariate techniques can help to simplify and organize large data sets and to make useful generalization that can lead to meaningful insight [32]. The rotation of the principal component was carried out by the Varimax normalized method. The Varimax rotation was performed to secure increased principal components of chemical/environmental significance. The hierarchical method of the cluster analysis used in this study has the advantage of not demanding any prior knowledge of the number of clusters, which is a prerequisite of the non-hierarchical method [32]. The normality of the distribution of each element was previously checked (Kolmogorov–Smirnov test). In this paper, distribution of metal concentration was normal, since the p-values in Kolmogorov–Smirnov test were higher than 0.05 in all the cases.

Enrichment Factors (EF) were computed utilizing the metal content in sediment and continental shale [33] as background value to understand the contamination state of the metals in sediment. The following equation was used for calculating the enrichment factors:

$$EF = (M/AI)_{\text{Sediment}} / (M/AI)_{\text{Background}}$$

## RESULTS AND DISCUSSION

### Distribution of metals, texture, organic-C and carbonate

General sediment characteristics of all the sampling sites are presented in Table-1 and Table-2 to have clear picture on the occurrence of metals in <63- $\mu\text{m}$  and <200 $\mu\text{m}$  sediment fraction of 30 sampling sites in 13 locations of Hugli estuary during dry season and wet season respectively. Table-1 and Table-2 revealed that concentrations of all metals were relatively high and consistent in <63- $\mu\text{m}$  sediment fraction compared to the occurrences of metals in <200 $\mu\text{m}$  sediment fraction over the sampling sites. Distribution of metal content in <200 $\mu\text{m}$  fraction of sediment is more or less similar to the distribution of org-C, carbonate and texture. Elevated level of metal concentration in <63- $\mu\text{m}$  sediment fraction indicated high affinity with fine grain sizes. Increasing metal concentrations in the finer grain sizes has also been reported by earlier studies [7, 8, 16].

Regular monitoring of metal contamination in sediment mainly reports level of metal contamination without specifying grain sizes of the sediment. Therefore total metal concentrations in sediment will obviously vary with the variation of the grain sizes and thereby evaluation of spatial and temporal variation without relating them with the grain sizes will mask actual variability of metal concentration in sediment. To justify the investment of vast human and financial resources in different monitoring programs, this aspect must be taken care off. This study clearly revealed the necessity of focusing the role of grain sizes even in regular monitoring activities. Several researchers reported interrelation of metals with sediment properties. A huge literature is available in different science journals focusing relation between grain sizes and level of metal concentrations in sediment. But It is also not always feasible to incorporate study of environmental behavior of metal in different grain sizes of sediment in regular monitoring programme. Considering the above fact, data interpretation was done to focus how to overcome the shortcomings in reporting the metal contamination in sediment. For this purpose, initially spatial trends of metals, texture,  $\text{CaCO}_3$  and organic matter were evaluated by Men-Kandall test and that showed no systematic trend (inclined / declined) over the sampling sites. Now the assembled analytical results of all the sampling sites in 13 locations ( Table – 1 and Table – 2) were processed to estimate the values of mean, median, SD, CV, maximum, minimum, skewness and kurtosis of metals, organic matter, carbonate and sediment texture (sand, silt and clay) and presented in Table-3. The mean values of all the metals in <63  $\mu\text{m}$  sediment fraction were consistently and prominently higher than that in <200- $\mu\text{m}$  sediment fraction in both seasons. These mean values clearly indicated that prevailing fluctuation of metal concentration over the sampling sites was mainly controlled by the grain sizes. This fluctuation cannot be considered as spatial variation during both seasons. The CV revealed that magnitude of variation of metal concentration among the stations in <63- $\mu\text{m}$  sediment fraction were remarkably decreased (15.4-37.7) compared to that (42.8-61.5) in <200- $\mu\text{m}$  sediment fraction. The Minimum-maximum values and First quartile –Third quartile values of metals in <63- $\mu\text{m}$  sediment fraction clearly indicated narrow range of metal concentrations in study area. This variability is decreased mainly due to differences in the grain sizes of the sample. Good agreement between mean and median of the metal concentrations (Table-3) exhibits normal distribution of the datasets in the study area. Skewness value less than 0.5 in all the metals except As (0.8) in <200- $\mu\text{m}$  sediment fraction indicated that distribution is symmetrical. Kurtosis values indicates a platykurtic to very leptokurtic distribution of metals in the sediments.

The distribution pattern of texture, carbonate and organic carbon is more or less identical with metal distribution pattern over the sampling sites. However, magnitude of variation with respect to CV of these parameters is relatively high in <200- $\mu\text{m}$  sediment fraction. The minimum and maximum concentration of metals along with organic carbon and sediment texture are also presented in Table-3 to show the ranges of their concentrations. The organic matter in sediments ranges between 0% and 0.9%, with an average of 0.3% during dry season and between 0% and 3.1%, with an average of 0.8% during wet season (Table 1). The carbonate in sediments ranges between 0% and 4.7%, with an average of 2.0% during dry season and between 0.4% and 5.5%, with an average of 3.2% during wet season. The clay in sediments ranges between 0% and 52.6%, with an average of 13.3% during dry season and between 0.5% and 27.9%, with an average of 12% during wet season (Table 1).

**Table-1 : Chemical Composition in <63- μm (s) and <200 μm(us) sediment fractions during dry season**

| Stn | Bank | Pb-s              | Pb-us     | Ni-s | Ni-us | Cu-s | Cu-us | Mn-s | Mn-us | Zn-s | Zn-us | Co-s | Co-us | Cd-s | Cd-us | Cr-s              | Cr-us | Hg-s | Hg-us       | As-s | As-us | Fe-s | Fe-us | Al-s | Al-us | Org.C us | CaCO3 us | Sand us | Silt us | Clay us |   |
|-----|------|-------------------|-----------|------|-------|------|-------|------|-------|------|-------|------|-------|------|-------|-------------------|-------|------|-------------|------|-------|------|-------|------|-------|----------|----------|---------|---------|---------|---|
|     |      | μgg <sup>-1</sup> |           |      |       |      |       |      |       |      |       |      |       |      |       | mgg <sup>-1</sup> |       |      |             |      |       |      |       |      |       | (%)      |          |         |         |         |   |
| G1  | RB   | 27                | 12        | 33   | 20    | 23   | 19    | 672  | 346   | 43   | 19    | 11   | 5     | 0.36 | 0.18  | 42                | 23    | 0.25 | 0.06        | 3.4  | 1.1   | 35   | 14    | 43   | 18    | 0.11     | 1.2      | 54.8    | 30      | 15      |   |
|     | LB   | 28.9              | 25.2      | 45   | 40    | 31   | 28    | 590  | 524   | 82   | 69    | 21   | 17    | 0.3  | 0.3   | 64                | 48    | 0.2  | <b>0.2</b>  | 2.0  | 1.4   | 28   | 21    | 35   | 28    | 0.2      | 2.0      | 22.1    | 26.4    | 50.4    |   |
| G2  | RB   | 33                | 33        | 49   | 42    | 36   | 32    | 966  | 833   | 59   | 50    | 18   | 12    | 0.50 | 0.38  | 66                | 56    | 0.27 | 0.24        | 3.2  | 3.1   | 37   | 34    | 45   | 45    | 0.56     | 3.6      | 25.1    | 71      | 4       |   |
| G3  | LB   | 26                | 10        | 37   | 10    | 27   | 14    | 692  | 217   | 33   | 14    | 13   | 3     | 0.24 | 0.06  | 35                | 7     | 0.19 | 0.04        | 1.4  | 0.9   | 34   | 12    | 44   | 15    | 0.06     | 0.8      | 86.9    | 11      | 12      |   |
|     | MB   |                   | <b>10</b> |      | 8     |      | 11    |      | 232   |      | 14    |      | 3     |      | 0.04  |                   | 5     |      | 0.03        |      | 0.8   |      | 9     |      | 15    | 0.01     | 0.7      | 93.1    | 6.8     | 0.1     |   |
|     | RB   | 20                | 11        | 31   | 17    | 17   | 12    | 571  | 328   | 23   | 13    | 7    | 3     | 0.26 | 0.12  | 51                | 17    | 0.12 | <b>0.06</b> | 1.5  | 0.5   | 21   | 15    | 27   | 19    | 0.10     | 0.3      | 85      | 12      | 3       |   |
| G4  | LB   | 30                | 26        | 48   | 42    | 32   | 29    | 603  | 545   | 86   | 71    | 22   | 18    | 0.32 | 0.30  | 66                | 51    | 0.20 | <b>0.17</b> | 2.1  | 1.5   | 29   | 22    | 36   | 28    | 0.21     | 2.1      | 19.8    | 28      | 53      |   |
|     | RB   | 35                | 13        | 47   | 16    | 38   | 15    | 855  | 349   | 57   | 36    | 13   | 9     | 0.36 | 0.09  | 54                | 14    | 0.21 | <b>0.05</b> | 2.5  | 0.5   | 44   | 18    | 55   | 23    | 0.11     | 1.9      | 70.4    | 24      | 5.2     |   |
| G5  | LB   | 26                | 25        | 36   | 33    | 20   | 19    | 542  | 500   | 96   | 76    | 20   | 17    | 0.22 | 0.16  | 40                | 33    | 0.24 | 0.06        | 1.9  | 0.6   | 23   | 20    | 28   | 27    | 0.33     | 3        | 30      | 58      | 13      |   |
|     | RB   | 30                | 18        | 42   | 29    | 32   | 23    | 983  | 613   | 65   | 50    | 19   | 13    | 0.36 | 0.24  | 66                | 40    | 0.23 | 0.13        | 2.1  | 1.1   | 46   | 32    | 58   | 42    | 0.75     | 1.1      | 29.2    | 58      | 13      |   |
| G6  | LB   | 21                | 19        | 31   | 21    | 25   | 23    | 655  | 493   | 44   | 34    | 10   | 10    | 0.30 | 0.18  | 51                | 40    | 0.27 | 0.15        | 2.1  | 2.5   | 29   | 24    | 37   | 32    | 0.42     | 3.5      | 19.5    | 63      | 17      |   |
|     | RB   | 29                | 22        | 40   | 34    | 36   | 19    | 846  | 613   | 97   | 72    | 16   | 15    | 0.36 | 0.00  | 73                | 38    | 0.18 | 0.14        | 1.7  | 0.8   | 36   | 23    | 44   | 30    | 0.51     | 3.5      | 45.8    | 53      | 1.7     |   |
| G7  | LB   | 27                | 23        | 29   | 28    | 25   | 19    | 617  | 448   | 58   | 39    | 18   | 10    | 0.22 | 0.14  | 54                | 40    | 0.18 | 0.15        | 1.5  | 1.1   | 28   | 24    | 36   | 30    | 0.41     | 3.2      | 34.3    | 60      | 5.4     |   |
|     | RB   | 30                | 24        | 33   | 29    | 28   | 20    | 780  | 486   | 83   | 62    | 18   | 16    | 0.38 | 0.20  | 49                | 36    | 0.21 | 0.17        | 3.5  | 1.5   | 36   | 26    | 44   | 34    | 0.54     | 3.2      | 18.6    | 62      | 19      |   |
| G8  | LB   | 29                | 19        | 40   | 26    | 35   | 19    | 763  | 434   | 57   | 46    | 19   | 10    | 0.28 | 0.22  | 59                | 40    | 0.16 | 0.12        | 1.4  | 1.1   | 24   | 21    | 30   | 28    | 0.23     | 1.3      | 49.1    | 43      | 7.6     |   |
|     | RB   | 28                | 22        | 33   | 26    | 21   | 16    | 554  | 414   | 44   | 34    | 13   | 8     | 0.30 | 0.24  | 44                | 33    | 0.21 | 0.14        | 2.4  | 1.6   | 31   | 23    | 39   | 30    | 0.88     | 3.3      | 13.9    | 64      | 22      |   |
| G9  | LB   | 29                | 26        | 36   | 30    | 30   | 28    | 571  | 461   | 50   | 38    | 13   | 9     | 0.28 | 0.17  | 57                | 51    | 0.23 | 0.16        | 1.9  | 1.7   | 29   | 25    | 37   | 33    | 0.58     | 3.4      | 17.1    | 65      | 18      |   |
|     | RB   | 24                | 20        | 33   | 30    | 23   | 19    | 545  | 458   | 46   | 33    | 13   | 9     | 0.26 | 0.17  | 42                | 35    | 0.21 | 0.19        | 3.4  | 1.6   | 46   | 28    | 56   | 38    | 0.41     | 3.2      | 32.6    | 50      | 17      |   |
| G10 | LB   | 16                | 12        | 21   | 12    | 19   | 10    | 287  | 157   | 53   | 27    | 10   | 6     | 0.21 | 0.07  | 35                | 15    | 0.11 | 0.03        | 1.2  | 0.3   | 13   | 6     | 16   | 7     | 0.08     | 0.3      | 88.5    | 8.4     | 3.1     |   |
|     | MB   |                   | 2.5       |      | 6.6   |      | 3     |      | 53    | 35   | 10    |      | 3     |      | 0.01  |                   | 4     |      | 0.12        | 0.03 | 1.1   | 0.4  |       | 5    |       | 6        | 0.11     | 0       | 98.5    | 1.5     | 0 |
|     | RB   | 33                | 24        | 41   | 31    | 35   | 22    | 707  | 448   | 74   | 59    | 19   | 13    | 0.39 | 0.21  | 64                | 43    | 0.19 | 0.15        | 3.5  | 1.4   | 32   | 24    | 39   | 32    | 0.45     | 2.9      | 20.4    | 63      | 16      |   |
| G11 | LB   | 32                | 24        | 37   | 31    | 34   | 26    | 711  | 511   | 66   | 45    | 17   | 11    | 0.28 | 0.16  | 59                | 49    | 0.22 | 0.17        | 2.4  | 1.8   | 31   | 24    | 40   | 32    | 0.45     | 3        | 24.7    | 49      | 26      |   |
|     | MB   |                   | 7         |      | 7.4   |      | 5     |      | 153   |      | 16    |      | 4     |      | 0.14  |                   | 8     |      | 0.19        | 0.11 | 1.9   | 1.2  |       | 5    |       | 6        | 0.03     | 0       | 99.5    | 0.5     | 0 |
|     | RB   | 27                | 23        | 36   | 32    | 31   | 23    | 630  | 505   | 67   | 46    | 16   | 12    | 0.26 | 0.22  | 61                | 49    | 0.17 | 0.13        | 1.9  | 2.4   | 29   | 22    | 37   | 29    | 0.34     | 3.1      | 22.9    | 57      | 20      |   |
| G12 | LB   | 25                | 19        | 30   | 21    | 28   | 22    | 585  | 428   | 23   | 15    | 10   | 4     | 0.12 | 0.08  | 49                | 40    | 0.23 | 0.19        | 3.1  | 2.8   | 40   | 29    | 50   | 38    | 0.28     | 3        | 37.7    | 49      | 14      |   |
|     | MB   |                   | 2         |      | 6.5   |      | 2     |      | 91    |      | 9     | 4    | 2     |      | 0.03  |                   | 7     |      | 0.09        | 0.03 | 1.1   | 0.8  |       | 6    |       | 8        | 0.02     | 0       | 99.8    | 1.2     | 0 |
|     | RB   | 29                | 24        | 33   | 30    | 31   | 22    | 568  | 448   | 51   | 42    | 14   | 10    | 0.23 | 0.13  | 59                | 42    | 0.19 | 0.14        | 1.7  | 1.6   | 25   | 23    | 32   | 31    | 0.31     | 2        | 29.2    | 54      | 17      |   |
| G13 | LB   | 26                | 14        | 37   | 16    | 40   | 10    | 832  | 288   | 38   | 20    | 18   | 5     | 0.20 | 0.09  | 64                | 28    | 0.11 | 0.07        | 1.7  | 0.9   | 41   | 16    | 53   | 20    | 0.01     | 0.5      | 82.1    | 13      | 5.2     |   |
|     | MB   |                   | 8.5       |      | 6.5   |      | 2     |      | 123   |      | 11    |      | 3     |      | 0.08  |                   | 3     |      | 0.10        | 0.03 | 0.8   | 0.5  |       | 7    |       | 9        | 0.02     | 0       | 99.4    | 0.6     | 0 |
|     | RB   | 29                | 28        | 43   | 39    | 37   | 32    | 672  | 616   | 68   | 65    | 18   | 14    | 0.21 | 0.05  | 68                | 54    | 0.3  | 0.2         | 3    | 3     | 35   | 32    | 44   | 43    | 0.39     | 4.7      | 11.8    | 68      | 21      |   |

**Table-2 : Chemical Composition in <63- μm(s) and <200 μm(us) sediment fractions during wet season**

| Stn | Bank | μgg <sup>-1</sup> |      |       |      |       |      |       |      |       |      |       |      |       |      |       |      | mgg <sup>-1</sup> |      |       |      |       |      | (%)   |      |          |                       |         |         |         |
|-----|------|-------------------|------|-------|------|-------|------|-------|------|-------|------|-------|------|-------|------|-------|------|-------------------|------|-------|------|-------|------|-------|------|----------|-----------------------|---------|---------|---------|
|     |      | Pb-us             | Pb-s | Ni-us | Ni-s | Cu-us | Cu-s | Mn-us | Mn-s | Zn-us | Zn-s | Co-us | Co-s | Cd-us | Cd-s | Cr-us | Cr-s | Hg-us             | Hg-s | As-us | As-s | Fe-us | Fe-s | Al-us | Al-s | Org.C-us | CaCO <sub>3</sub> -us | Sand-us | Silt-us | Clay-us |
| G1  | LB   | 22                | 29   | 35    | 43   | 33    | 40   | 656   | 730  | 65    | 75   | 17    | 20   | 0.31  | 0.34 | 50    | 68   | 0.21              | 0.25 | 1.8   | 2.3  | 37    | 44   | 46    | 59   | 0.97     | 3.9                   | 8.6     | 67.1    | 24      |
|     | RB   | 21                | 30   | 29    | 37   | 20    | 28   | 651   | 702  | 49    | 69   | 12    | 16   | 0.2   | 0.2  | 44    | 46   | 0.2               | 0.29 | 1.5   | 2.0  | 26    | 32   | 33    | 43   | 3.1      | 4.1                   | 10      | 74.4    | 15      |
| G2  | LB   | 19                | 29   | 23    | 27   | 17    | 26   | 451   | 587  | 46    | 54   | 10    | 13   | 0.3   | 0.3  | 33    | 43   | 0.18              | 0.23 | 1.7   | 2.2  | 23    | 29   | 29    | 39   | 3        | 3.2                   | 13      | 69.8    | 17      |
|     | MB   | 18                | 27   | 21    | 28   | 18    | 31   | 376   | 641  | 46    | 63   | 11    | 16   | 0.3   | 0.6  | 41    | 43   | 0.15              | 0.28 | 1.6   | 2.9  | 21    | 35   | 27    | 47   | 3.1      | 2.9                   | 41      | 50.5    | 8.4     |
| G3  | LB   | 11                | 16   | 12    | 21   | 3     | 6    | 321   | 540  | 19    | 24   | 3     | 6    | 0.1   | 0.2  | 7     | 12   | 0.01              | 0.02 | 0.7   | 0.8  | 10    | 12   | 12    | 16   | 0.6      | 0.9                   | 89      | 9.2     | 1.6     |
|     | RB   | 18                | 20   | 24    | 31   | 18    | 24   | 425   | 576  | 42    | 56   | 7     | 10   | 0.2   | 0.3  | 33    | 40   | 0.16              | 0.20 | 1.4   | 1.8  | 22    | 26   | 27    | 34   |          | 2.6                   | 36      | 44.1    | 20      |
| G4  | LB   | 16                | 21   | 20    | 26   | 16    | 16   | 456   | 578  | 26    | 42   | 10    | 15   | 0.2   | 0.3  | 29    | 45   | 0.11              | 0.14 | 2.1   | 2.6  | 21    | 25   | 26    | 0    | 2.2      | 4.2                   | 13      | 71.1    | 15      |
|     | MB   | 12                | 21   | 16    | 39   | 9     | 16   | 329   | 675  | 21    | 59   | 9     | 16   | 0.04  | 0.21 | 25    | 35   | 0.11              | 0.34 | 1.3   | 3.8  | 12    | 34   | 15    | 69   | 0.42     | 1.1                   | 67      | 29      | 4       |
| G5  | LB   | 22                | 27   | 37    | 43   | 33    | 39   | 660   | 725  | 64    | 73   | 17    | 20   | 0.29  | 0.32 | 53    | 71   | 0.21              | 0.27 | 2.3   | 2.8  | 37    | 43   | 48    | 47   | 0.43     | 4.2                   | 8.2     | 67.9    | 24      |
|     | MB   | 9                 |      | 3     |      | 2     | 0    | 120   |      | 16    |      | 4     |      | 0.00  | 0.00 | 8     |      | 0.06              | 0.00 | 0.5   |      | 7     |      | 9     | 0    | 0.08     | 0.7                   | 95      | 4.3     | 0.6     |
| G6  | LB   | 22                | 30   | 42    | 52   | 34    | 38   | 680   | 715  | 66    | 69   | 19    | 21   | 0.35  | 0.42 | 55    | 3    | 0.25              | 0.35 | 1.9   | 2.4  | 33    | 38   | 40    | 40   | 0.48     | 5.1                   | 6.6     | 65.5    | 28      |
|     | MB   | 5                 |      | 6     |      | 5     | 0    | 132   |      | 20    |      | 6     |      | 0.08  |      | 12    |      | 0.07              | 0.00 | 0.6   |      | 12    |      | 14    | 0    | 0.09     | 1.3                   | 98      | 1.8     | 0.2     |
| G7  | LB   | 22                | 28   | 36    | 38   | 28    | 31   | 490   | 515  | 55    | 62   | 16    | 17   | 0.12  | 0.30 | 48    | 57   | 0.21              | 0.25 | 2.1   | 2.4  | 26    | 29   | 32    | 30   | 0.49     | 4.8                   | 35      | 50.6    | 14      |
|     | MB   | 8                 |      | 9     |      | 3     | 0    | 137   |      | 30    |      | 5     |      | 0.08  |      | 8     |      | 0.13              | 0.00 | 0.5   |      | 11    |      | 13    | 0    | 0.03     | 1.1                   | 86      | 10.4    | 3.7     |
| G8  | LB   | 16                | 25   | 27    | 47   | 24    | 39   | 414   | 655  | 37    | 83   | 11    | 17   | 0.20  | 0.23 | 33    | 47   | 0.21              | 0.27 | 1.5   | 2.2  | 30    | 41   | 37    | 65   | 0.37     | 4.1                   | 51      | 36.7    | 13      |
|     | MB   | 4                 |      | 3     |      | 4     | 0    | 86    |      | 11    |      | 7     |      |       |      | 7     |      | 0.05              | 0.00 | 0.7   |      | 10    |      | 13    | 0    | 0.10     | 0.8                   | 95      | 4.7     | 0       |
| G9  | LB   | 24                | 28   | 37    | 51   | 31    | 40   | 535   | 695  | 59    | 73   | 16    | 17   | 0.42  | 0.39 | 47    | 56   | 0.22              | 0.28 | 2.5   | 3.0  | 38    | 40   | 46    | 52   | 0.49     | 3.7                   | 19      | 58.3    | 23      |
|     | MB   | 2                 |      | 2     |      | 2     | 0    | 132   |      | 11    |      | 6     |      | 0.06  |      | 7     |      | 0.05              | 0.00 | 0.2   |      | 9     |      | 12    | 0    | 0.07     | 0.4                   | 96      | 3       | 0.6     |
| G10 | LB   | 30                | 32   | 54    | 58   | 40    | 40   | 575   | 750  | 62    | 72   | 18    | 23   | 0.30  | 0.42 | 59    | 71   | 0.09              | 0.12 | 3.4   | 4.4  | 35    | 41   | 43    | 54   | 0.57     | 3.1                   | 14      | 78.9    | 7.1     |
|     | MB   | 13                | 26   | 10    | 43   | 6     | 41   | 175   | 590  | 16    | 79   | 8     | 17   | 0.11  | 0.39 | 18    | 73   | 0.12              | 0.00 | 0.7   | 0.0  | 12    |      | 16    | 0    | 0.05     | 3.0                   | 86      | 11      | 3.6     |
| G11 | LB   | 21                | 29   | 36    | 52   | 29    | 41   | 435   | 625  | 54    | 56   | 17    | 20   | 0.21  | 0.36 | 46    | 56   | 0.22              | 0.27 | 2.6   | 3.2  | 34    | 40   | 41    | 58   | 0.36     | 4.9                   | 36      | 42.5    | 21      |
|     | MB   | 25                | 26   | 44    | 50   | 42    | 46   | 590   | 750  | 96    | #    | 20    | 22   | 0.33  | 0.42 | 54    | 63   | 0.22              | 0.30 | 2.1   | 2.6  | 36    | 41   | 45    | 52   | 0.48     | 5.5                   | 11      | 66      | 23      |
| G12 | LB   | 18                | 28   | 33    | 53   | 23    | 41   | 365   | 595  | 75    | 99   | 13    | 18   | 0.21  | 0.27 | 31    | 58   | 0.18              | 0.26 | 1.7   | 2.7  | 27    | 41   | 33    | 67   | 0.23     | 3.9                   | 29      | 65.5    | 5.9     |
|     | MB   | 22                | 24   | 46    | 49   | 41    | 42   | 540   | 590  | 73    | 78   | 21    | 23   | 0.26  | 0.36 | 63    | 69   | 0.26              | 0.28 | 3.4   | 3.9  | 41    | 43   | 50    | 47   | 0.87     | 4.5                   | 10      | 69.6    | 20      |
| G13 | LB   | 21                | 28   | 27    | 43   | 21    | 35   | 574   | 888  | 43    | 62   | 11    | 14   | 0.09  | 0.18 | 34    | 37   | 0.24              | 0.34 | 3.7   | 5.0  | 34    | 43   | 44    | 67   | 0.20     | 5.1                   | 63      | 22.5    | 14      |
|     | RB   | 4                 |      | 4     |      | 2     | 11   | 71    | 213  | 20    | 12   | 6     | 11   |       |      | 10    | 32   | 0.04              | 0.05 | 0.4   | 0.5  | 11    | 8    | 13    | 23   | 0.03     | 0.5                   | 90      | 9.2     | 0.5     |

Table-3 : Descriptive statistics of Metals in <63- µm(s) and <200 µm(us), Org-C, Carbonate and Texture in <200 µm sediment during dry and wet season

|                   | Pb-s              | Pb-us | Ni-s | Ni-us | Cu-s | Cu-us | Mn-s  | Mn-us | Zn-s  | Zn-us | Co-s | Co-us | Cd-s | Cd-us | Cr-s | Cr-us | Hg-s | Hg-us | As-s | As-us | Fe-s              | Fe-us | Al-s | Al-us | Org.C | CaCO3 | Sand | Silt | Clay |  |
|-------------------|-------------------|-------|------|-------|------|-------|-------|-------|-------|-------|------|-------|------|-------|------|-------|------|-------|------|-------|-------------------|-------|------|-------|-------|-------|------|------|------|--|
|                   | µgg <sup>-1</sup> |       |      |       |      |       |       |       |       |       |      |       |      |       |      |       |      |       |      |       | mgg <sup>-1</sup> |       |      |       |       | (%)   |      |      |      |  |
| <b>Dry season</b> |                   |       |      |       |      |       |       |       |       |       |      |       |      |       |      |       |      |       |      |       |                   |       |      |       |       |       |      |      |      |  |
| Mean              | 27.5              | 18.1  | 36.7 | 23.9  | 29.4 | 18.3  | 671.9 | 403.8 | 57.7  | 38.0  | 14.9 | 9.0   | 0.3  | 0.1   | 54.8 | 31.6  | 0.2  | 0.1   | 2.1  | 1.3   | 32.1              | 19.6  | 40.3 | 25.9  | 0.3   | 2.0   | 48.7 | 38.4 | 13.3 |  |
| Std Dev           | 4.2               | 7.7   | 6.6  | 11.2  | 6.5  | 8.5   | 150.8 | 178.5 | 20.4  | 21.1  | 4.5  | 5.0   | 0.1  | 0.1   | 10.7 | 17.0  | 0.1  | 0.1   | 0.8  | 0.7   | 7.8               | 8.5   | 9.7  | 11.2  | 0.2   | 1.4   | 31.8 | 24.7 | 13.0 |  |
| CV                | 15.4              | 42.7  | 18.1 | 46.8  | 22.0 | 46.3  | 22.4  | 44.2  | 35.3  | 55.6  | 30.5 | 55.1  | 27.6 | 61.5  | 19.6 | 53.9  | 29.4 | 54.7  | 37.7 | 55.7  | 24.3              | 43.2  | 24.2 | 43.1  | 78.0  | 68.9  | 65.3 | 64.3 | 98.1 |  |
| Max               | 34.9              | 32.5  | 48.8 | 42.0  | 39.6 | 32.4  | 982.8 | 833.0 | 96.8  | 75.7  | 21.5 | 17.6  | 0.5  | 0.4   | 72.8 | 56.2  | 0.3  | 0.2   | 3.5  | 3.1   | 45.7              | 34.2  | 58.4 | 44.8  | 0.9   | 4.7   | 99.8 | 70.9 | 52.6 |  |
| Min               | 16.0              | 2.0   | 20.5 | 6.5   | 16.8 | 2.4   | 287.0 | 53.2  | 23.0  | 9.0   | 4.0  | 2.0   | 0.1  | 0.0   | 34.6 | 2.6   | 0.1  | 0.0   | 0.8  | 0.3   | 13.2              | 4.9   | 16.4 | 6.4   | 0.0   | 0.0   | 11.8 | 0.5  | 0.0  |  |
| 1st quartile      | 25.6              | 12.0  | 32.5 | 15.8  | 25.2 | 12.5  | 571.2 | 298.2 | 44.2  | 16.6  | 12.5 | 4.4   | 0.2  | 0.1   | 49.1 | 15.1  | 0.2  | 0.1   | 1.5  | 0.8   | 28.1              | 14.3  | 35.8 | 18.2  | 0.1   | 0.7   | 22.3 | 12.2 | 3.3  |  |
| 3rd Quartile      | 29.5              | 23.5  | 40.6 | 31.0  | 34.8 | 22.8  | 763.0 | 504.0 | 67.7  | 50.1  | 18.0 | 12.8  | 0.4  | 0.2   | 64.2 | 43.0  | 0.2  | 0.2   | 2.5  | 1.6   | 36.0              | 24.0  | 44.4 | 32.1  | 0.4   | 3.2   | 84.3 | 59.6 | 17.6 |  |
| Skew              | -0.9              | -0.5  | -0.1 | -0.2  | -0.3 | -0.4  | 0.1   | -0.2  | 0.3   | 0.3   | -0.6 | 0.1   | 0.5  | 0.5   | -0.4 | -0.4  | 0.1  | 0.1   | 0.5  | 0.8   | -0.2              | -0.4  | -0.2 | -0.4  | 0.6   | -0.1  | 0.6  | -0.4 | 1.6  |  |
| Kurt              | 1.3               | -0.6  | 0.2  | -1.1  | -0.9 | -0.5  | 1.0   | 0.1   | -0.5  | -1.1  | -0.3 | -1.2  | 1.0  | 0.0   | -0.8 | -1.2  | 0.5  | -0.9  | -0.8 | 0.0   | 0.2               | -0.7  | 0.4  | -0.7  | -0.1  | -1.3  | -1.4 | -1.5 | 3.4  |  |
| Median            | 28.5              | 19.5  | 36.4 | 27.0  | 30.7 | 19.2  | 655.2 | 448.0 | 57.0  | 37.0  | 16.3 | 9.4   | 0.3  | 0.2   | 56.6 | 36.6  | 0.2  | 0.1   | 1.9  | 1.2   | 31.3              | 21.7  | 39.2 | 28.1  | 0.3   | 2.1   | 33.5 | 49.4 | 12.8 |  |
| <b>Wet season</b> |                   |       |      |       |      |       |       |       |       |       |      |       |      |       |      |       |      |       |      |       |                   |       |      |       |       |       |      |      |      |  |
| Mean              | 26.1              | 17.1  | 41.0 | 25.8  | 27.2 | 20.5  | 641.0 | 413.8 | 65.4  | 43.2  | 17.3 | 12.2  | 0.3  | 0.2   | 50.7 | 35.3  | 0.2  | 0.2   | 2.4  | 1.6   | 35.4              | 24.4  | 37.0 | 30.1  | 0.8   | 3.2   | 44.3 | 43.7 | 12.0 |  |
| Std Dev           | 3.7               | 6.8   | 9.7  | 13.8  | 15.3 | 12.3  | 75.9  | 176.7 | 16.7  | 20.7  | 3.7  | 5.2   | 0.1  | 0.1   | 17.1 | 17.1  | 0.1  | 0.1   | 0.9  | 0.7   | 7.4               | 9.8   | 23.4 | 12.3  | 0.9   | 1.5   | 32.6 | 25.2 | 8.8  |  |
| CV                | 14.2              | 39.9  | 23.7 | 53.6  | 56.1 | 59.9  | 11.8  | 42.7  | 25.6  | 48.0  | 21.4 | 43.0  | 35.4 | 50.3  | 33.7 | 48.6  | 60.7 | 42.5  | 36.9 | 47.0  | 20.8              | 40.4  | 63.3 | 40.7  | 115.6 | 47.2  | 73.6 | 57.7 | 73.5 |  |
| Max               | 31.5              | 30.0  | 58.1 | 53.6  | 46.4 | 42.2  | 779.8 | 680.0 | 112.0 | 96.0  | 22.7 | 20.5  | 0.6  | 0.4   | 72.5 | 61.5  | 0.4  | 0.3   | 4.4  | 3.4   | 44.3              | 37.8  | 68.9 | 47.5  | 3.1   | 5.5   | 98.0 | 78.9 | 27.9 |  |
| Min               | 16.0              | 2.0   | 21.0 | 1.7   | 0.0  | 2.3   | 515.0 | 85.5  | 24.0  | 10.5  | 6.5  | 3.1   | 0.0  | 0.0   | 2.6  | 6.6   | 0.0  | 0.0   | 0.0  | 0.2   | 11.8              | 7.2   | 0.0  | 9.1   | 0.0   | 0.4   | 6.6  | 1.8  | 0.0  |  |
| 1st quartile      | 24.8              | 12.4  | 36.5 | 15.6  | 17.8 | 10.1  | 580.0 | 323.0 | 57.0  | 23.8  | 15.8 | 7.5   | 0.3  | 0.1   | 42.6 | 26.0  | 0.1  | 0.1   | 2.1  | 1.1   | 32.3              | 14.0  | 19.4 | 17.6  | 0.3   | 2.2   | 13.5 | 24.1 | 3.8  |  |
| 3rd Quartile      | 28.3              | 21.9  | 48.0 | 36.3  | 39.6 | 29.4  | 702.0 | 538.8 | 74.0  | 60.9  | 20.0 | 16.9  | 0.4  | 0.3   | 63.2 | 49.5  | 0.3  | 0.2   | 2.9  | 2.1   | 41.3              | 33.2  | 53.5 | 40.8  | 0.8   | 4.2   | 71.1 | 65.9 | 19.0 |  |
| Skew              | -1.0              | -0.6  | -0.6 | -0.2  | -0.9 | -0.1  | 0.2   | -0.4  | 0.0   | 0.2   | -1.2 | -0.1  | -0.5 | -0.2  | -1.2 | -0.4  | -0.8 | -0.6  | -0.5 | 0.2   | -1.6              | -0.4  | -0.7 | -0.3  | 1.7   | -0.5  | 0.5  | -0.4 | 0.1  |  |
| Kurt              | 0.8               | -0.2  | -0.5 | -0.8  | -0.7 | -1.1  | -1.1  | -0.8  | 2.6   | -0.2  | 1.8  | -1.3  | 2.0  | -0.6  | 1.8  | -1.0  | -0.9 | -0.8  | 1.9  | -0.1  | 3.4               | -1.3  | -0.9 | -1.3  | 1.7   | -0.9  | -1.3 | -1.2 | -1.4 |  |
| Median            | 27.0              | 18.5  | 43.1 | 27.7  | 32.4 | 20.7  | 625.0 | 435.0 | 69.0  | 47.0  | 17.3 | 11.5  | 0.3  | 0.2   | 56.2 | 36.7  | 0.3  | 0.2   | 2.4  | 1.7   | 37.9              | 26.4  | 44.3 | 32.4  | 0.5   | 3.5   | 35.7 | 49.9 | 12.4 |  |

The silt content in sediments ranges between 0.5% and 70.9%, with an average of 38.4% during dry season and between 1.8% and 78.9%, with an average of 43.7% during wet season. It has been reported that surface runoff bring huge amount of silt during monsoon season [34] in addition to other pollutants from upland. The sand content in sediments ranges between 11.8% and 99.8%, with an average of 48.7% during dry season and between 6.6% and 98%, with an average of 44.3% during wet season. The skewness values indicate normal distribution of data and Kurtosis values indicate a platykurtic to very leptokurtic distribution of metals in the sediments. The Org-C in sediments is mainly related to the decomposition of aquatic organisms and surface runoff from the basin. The geochemistry of sediment in this estuary is dominated by their texture irrespective of their origin. The river sediments are predominantly composed of fine sand, very fine sand and silt along with minor amount of clay fractions. As reported by Chakrapani *et al.*, [35] suspended sediments of the Ganges River consist predominantly of coarse to medium silt (62 – 15  $\mu\text{m}$ ) with dominance of mica as clay minerals. A general decrease of metal content in coarse fraction of the sediments is due to the dilution effect of an increase in quartz and feldspar contents [36]. This study exhibits distribution pattern of these parameters more or less similar to the distribution pattern reported by other researchers [7,16]. The metal concentrations were found less where org-C content were low which indicates that metal content in sediment originates partly from the organic materials. Therefore relatively high concentrations in sediments of few spots, organic materials may take the metal ions out of solution and contribute to the sediment causing fluctuations of metal concentrations. Further increase of concentrations in few locations despite more or less same concentrations of org-C may be due to sorption of metals on to clay which is negatively charged making its surface capable to absorb the metal ions by their outer sheaths of hydroxyl group.

Though differences of concentrations of metals between dry season and wet season were not prominent but concentrations Ni, Zn, Cr, Co, Cr As and Fe in <63  $\mu\text{m}$  fraction were found little high during wet season compared to that of dry season and concentrations of Pb, Mn and Al were marginally low during dry season. Magnitude of difference in metal concentrations observed between dry season and wet season cannot be explained in response to temporal condition.

Based on above discussion it may be inferred that grain size of sediment play crucial role in governing the level of metal concentrations in the sediments. In this context it may be mentioned that influence of grain size on the level of metal concentrations is so prominent that spatial and temporal condition cannot be simply evaluated. Therefore datasets need to be further processed to evaluate the status of contamination and interrelation among metals and other sediment properties in clear statistical terms.

#### **Level of Contamination and Enrichment Factor**

The anthropogenic activities and soil erosion going on in vast catchments area of Hugli were the major source of pollutants. Therefore distribution pattern alone is unable to explain the input of metals from natural and anthropogenic sources. The mean concentration of metal (N=60) in the study area are compared with background concentration of metals, guideline values and other estuary to evaluate the prevailing status of metal contamination. The mean concentrations of Pb was higher than Pb in World Shale value (Table -4). The measured metal concentrations decrease in the following order in accordance with reported concentrations of these metals in crustal average [37]: Mn > Zn > Cr > Ni > Cu > Pb > Co > Cd. The order of prevalence (ranking) of these metals is identical to the order of continental Shale. Many authors prefer to express the metal contamination with respect to average shale value to quantify the extent and degree of metal pollution [38-39]. In the present study enrichment factor was used to assess the level of contamination and the possible anthropogenic impact in sediments of estuary. To identify anomalous metal concentration, geochemical normalization of the heavy metals data to a conservative element, such as Al, Fe, and Si are employed. Several authors have successfully used Al to normalize metals contaminants. In this study Al has been used as a conservative element to differentiate natural from anthropogenic components. In this study, the background concentrations were taken from Turekian and Wedepohl (33). Generally EF values between 0.5 and 1.5 indicate the metal is entirely from crustal materials or natural processes, whereas EF values greater than 1.5 suggest that the sources are more likely to be anthropogenic. The results of the present study show that Pb, Ni, Cd and Cr were significantly enriched in sediments of Hugli estuary since EF values of these four metals are greater than 2.0 and EF of Cu, Co and Fe were moderately enriched in sediments since EFs were greater than 1.5 (Table- 5). This suggests that these metals are partly originated from anthropogenic inputs. It is pertinent to mention that lead has a higher mean concentrations than sediments from other estuary reported by Hema Achyulthan *et al* [40] and the crustal average, suggesting Pb contamination. In addition to Pb contamination, EFs suggest contamination of Ni, Cd, Cr, Cu, Co, and Fe in sediment of Hooghly estuary. This observation is clear indicative of anthropogenic contamination though not in alarming level.

Table-4 Mean Concentration of metals compared with Background, and Sediment guidelines and other estuary

|                                                                          |     | Pb   | Ni   | Cu   | Mn  | Zn  | Co   | Cd    | Cr   | Hg    | As   | Fe | Al   |
|--------------------------------------------------------------------------|-----|------|------|------|-----|-----|------|-------|------|-------|------|----|------|
| This Study                                                               | Wet | 26.2 | 41.8 | 33.3 | 646 | 67  | 17.5 | 0.34  | 51.2 | 0.25  | 2.58 | 36 | 46   |
|                                                                          | Dry | 27.5 | 36.7 | 29.4 | 672 | 59  | 15.3 | 0.29  | 54.8 | 0.20  | 2.27 | 32 | 40   |
| Shale value <sup>1</sup>                                                 |     | 20   | 68   | 45   | 850 | 95  | 19   | 0.3   | 90   | 0.4   | 13   | 47 | 81   |
| Crust ave. <sup>1</sup>                                                  |     | 13   | 75   | 55   | 950 | 70  | 25   | 0.2   | 100  | 0.08  | 1.5  | 50 | 81.3 |
| Adyar Estuary                                                            |     | 2    | 426  |      | 345 | 168 | 10   | -     | 318  | -     | -    |    |      |
| NOAA (National Oceanic and Atmospheric Administration, USA) <sup>2</sup> |     |      |      |      |     |     |      |       |      |       |      |    |      |
| TEL                                                                      |     | 35   | 18   | 35.7 | -   | 123 | -    | 0.596 | 37.3 | 0.174 | 5.9  | -  | -    |
| PEL                                                                      |     | 91.3 | 35.9 | 197  | -   | 315 | -    | 3.530 | 90   | 0.486 | 17   | -  | -    |
| CCME (Canadian Council of Ministers for Environment) <sup>3</sup>        |     |      |      |      |     |     |      |       |      |       |      |    |      |
| ISQG                                                                     |     | 35   | -    | 35.7 | -   | 123 | -    | 0.6   | 37.3 | 0.17  | 5.9  | -  | -    |
| PEL                                                                      |     | 91.3 | -    | 197  | -   | 315 | -    | 3.5   | 90   | 0.486 | 17   | -  | -    |
| CBSQGs (Consensus Based Sediment Quality Guidelines) <sup>4</sup>        |     |      |      |      |     |     |      |       |      |       |      |    |      |
| Threshold Effect Concentration                                           |     |      |      |      |     |     |      |       |      |       |      |    |      |
| Tel                                                                      |     | 35   | 18   | 35.7 | -   | 123 | -    | 0.596 | 37.3 | 0.174 | 5.9  | -  | -    |
| LEL                                                                      |     | 31   | 16   | 16   | -   | 120 | -    | 0.6   | 26   | 0.2   | 6    | -  | -    |
| ERL                                                                      |     | 35   | 30   | 70   | -   | 120 | -    | 5     | 80   | 0.15  | 33   | -  | -    |
| Probable Effect Concentration                                            |     |      |      |      |     |     |      |       |      |       |      |    |      |
| PEL                                                                      |     | 91.3 | 36   | 197  | -   | 315 | -    | 3.53  | 90   | 0.486 | 17   | -  | -    |
| SEL                                                                      |     | 250  | 75   | 110  | -   | 820 | -    | 10    | 110  | 2     | 33   | -  | -    |
| ERM                                                                      |     | 110  | 50   | 390  | -   | 270 | -    | 9     | 145  | 1.3   | 85   | -  | -    |

<sup>1</sup> = K.K. Turkian, K.H. Wedepohl. Bull. Geol. Soc. Am. 1961, 72:175

<sup>2</sup> = National Oceanic and Atmospheric Administration (NOAA), (1999), NOAA screening quick reference tables (SQiRTs), HAZMAT REPORT 99-1, and Updated Feb 2004. Washington.

<sup>3</sup> = CCME, 2002. Canadian Council of Ministers for the Environment 2002. Canadian Sediment Quality guidelines for the Protection of the Protection of Aquatic life: updated, 2002. Winnipeg, Canada.

<sup>4</sup> = Wisconsin Department of Natural Resources (WDNR), 2003. Consensus-based sediment quality guidelines (CG-SQGs) recommendations for use and application interim guidance developed by the contaminated sediment Standing Team WT-732 2003.

Table-5 : Summary of Statistics of Enrichment Factors (EFs) with respect to World Shale Value and Upper Crust

|                                                             | Pb   | Ni    | Cu   | Mn   | Zn   | Co   | Cd   | Cr   | Fe   |
|-------------------------------------------------------------|------|-------|------|------|------|------|------|------|------|
| EF with respect to World Shale Value( Present study)        | 2.73 | 2.89  | 1.75 | 1.42 | 1.22 | 1.59 | 5.86 | 3.08 | 2.62 |
| EF with respect to Upper Crust (Present study)              | 2.47 | 3.63  | 1.66 | 2.02 | 1.63 | 3.03 | 5.89 | 3.9  | -    |
| Adyar Estuary, SE Coast of India (0-10.5 cm interval)       | 0.10 | 21.12 | -    | 0.57 | 2.35 | 0.99 | -    | 8.99 | -    |
| SE Coast of India – Muttukadu (0-5 cm interval) Tidal Zones | 0.06 | 2.81  | -    | 0.78 | 1.10 | 0.92 | -    | 1.57 | -    |

### Interrelationship of metals, texture, org-C, and carbonate

The variability of metal concentration in the sediment is found more or less identical when compared with distribution pattern of organics and sediment texture. Considering this fact the Pearson's correlation analysis at significance level of  $p < 0.001$  were employed to establish their interrelationship among metals, texture and organic. Pearson's correlation coefficients among the contents of Pb, Ni, Cu, Mn, Zn, Co, Cd, Cr, Hg, As, Fe, Al, Org-C, carbonate, sand, silt and clay in sediments of Hugli estuarine are presented in Table 4. The strong positive linear correlations among metal, Org-C, carbonate and texture were clearly observed. The strong positive linear correlations among Pb, Ni, Cu, Mn, Zn, Co, Cd, Cr, Hg, As, Fe, and Al indicates that these metals are associated with each other. The strong linear correlations between the metal concentrations indicate similar origins of these elements and there are some common factors controlling their variability. The correlation coefficient explored the significant positive correlation of metals with organic, carbonate, clay, and silt whereas negative correlation with sand. Positive correlation reveals the affinity of these metals to organic matter, carbonate, silt and clay. Singh et.al.[41] advocated that textural concentrations of river sediment played an important role in the study of bioavailability of metals and toxicity of these sediments as these metals are not homogeneously distributed over various grain sizes. Strong correlation of these metals with clay, silt, carbonate and organic matter indicated that the land derived to the metals in the sediments are not concentrated in oxides and other accessory non-aluminous silicate materials which is contradictory to report of Selvaraj et al.[1]. Positive correlation of Al with organic matter and the total Cr would imply that fine particle fractions enriched in Al adsorb organic matter, which scavenges the Cr [2]. In this study area sand exhibits negative correlation with  $\text{CaCO}_3$  indicating that shell materials present in the sediment are originated from the sand.

Table-6 : Correlation Coefficient among metals ,textural, org-C and carbonate in Sediment of Hugli Estuary

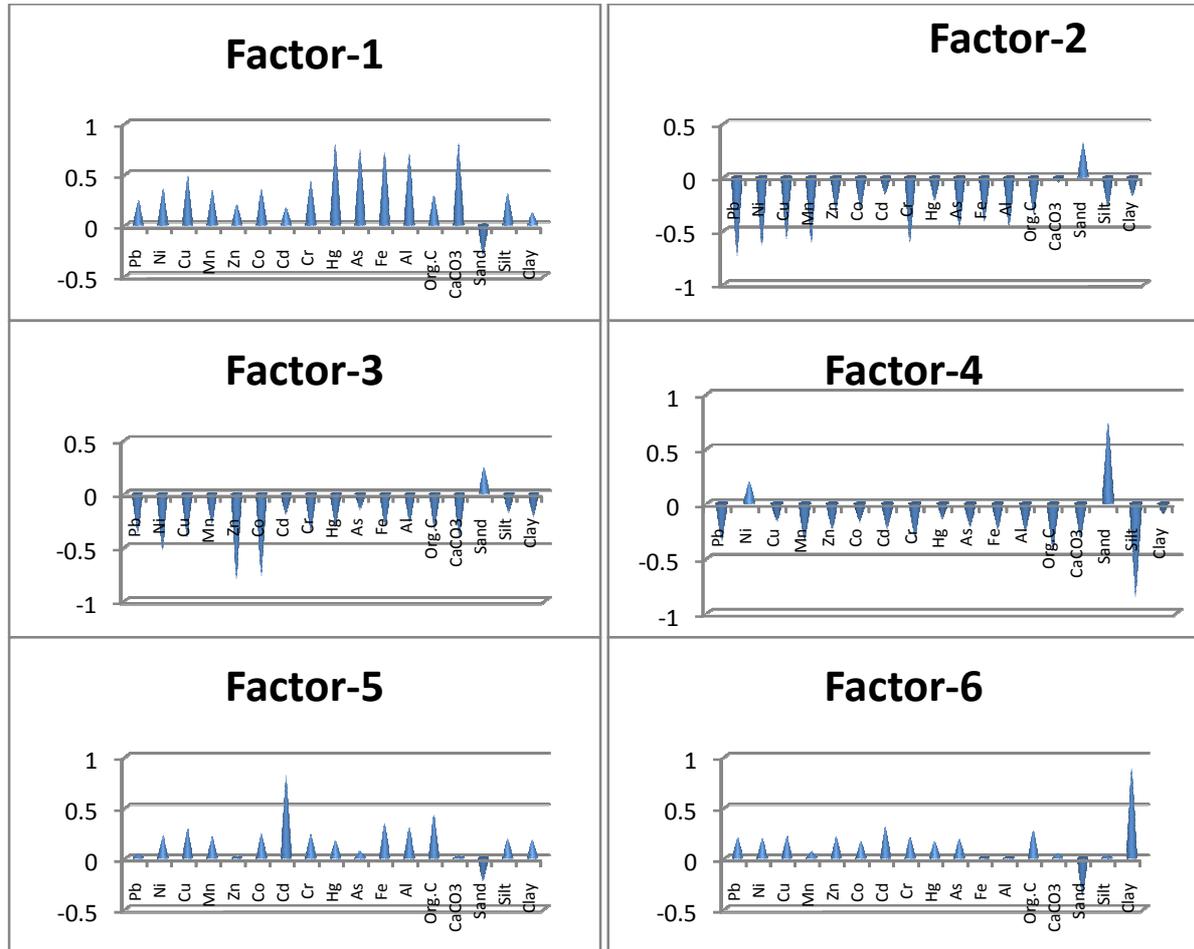
|       | Pb   | Ni   | Cu   | Mn   | Zn   | Co   | Cd   | Cr   | Hg   | As   | Fe   | Al   | Org.<br>C | CaCO<br>3 | San<br>d | Silt | Clay |
|-------|------|------|------|------|------|------|------|------|------|------|------|------|-----------|-----------|----------|------|------|
| Pb    |      |      |      |      |      |      |      |      |      |      |      |      |           |           |          |      |      |
| Ni    | 0.86 |      |      |      |      |      |      |      |      |      |      |      |           |           |          |      |      |
| Cu    | 0.76 | 0.92 |      |      |      |      |      |      |      |      |      |      |           |           |          |      |      |
| Mn    | 0.77 | 0.76 | 0.7  |      |      |      |      |      |      |      |      |      |           |           |          |      |      |
| Zn    | 0.71 | 0.83 | 0.75 | 0.64 |      |      |      |      |      |      |      |      |           |           |          |      |      |
| Co    | 0.69 | 0.87 | 0.8  | 0.64 | 0.89 |      |      |      |      |      |      |      |           |           |          |      |      |
| Cd    | 0.47 | 0.57 | 0.58 | 0.51 | 0.52 | 0.55 |      |      |      |      |      |      |           |           |          |      |      |
| Cr    | 0.84 | 0.89 | 0.88 | 0.75 | 0.7  | 0.78 | 0.57 |      |      |      |      |      |           |           |          |      |      |
| Hg    | 0.59 | 0.67 | 0.72 | 0.61 | 0.54 | 0.65 | 0.44 | 0.75 |      |      |      |      |           |           |          |      |      |
| As    | 0.57 | 0.66 | 0.68 | 0.55 | 0.33 | 0.44 | 0.37 | 0.67 | 0.65 |      |      |      |           |           |          |      |      |
| Fe    | 0.67 | 0.8  | 0.86 | 0.76 | 0.65 | 0.73 | 0.57 | 0.8  | 0.72 | 0.74 |      |      |           |           |          |      |      |
| Al    | 0.71 | 0.8  | 0.85 | 0.77 | 0.64 | 0.71 | 0.54 | 0.81 | 0.81 | 0.75 | 0.99 |      |           |           |          |      |      |
| Org.C | 0.55 | 0.62 | 0.61 | 0.58 | 0.48 | 0.6  | 0.43 | 0.67 | 0.56 | 0.39 | 0.67 | 0.68 |           |           |          |      |      |
| CaCO3 | 0.54 | 0.61 | 0.63 | 0.57 | 0.58 | 0.67 | 0.36 | 0.63 | 0.79 | 0.63 | 0.77 | 0.75 | 0.48      |           |          |      |      |
| Sand  | 0.72 | 0.71 | -0.7 | 0.68 | -0.7 | -0.7 | -0.6 | -0.8 | -0.6 | -0.6 | -0.7 | -0.7 | -0.59     | -0.59     |          |      |      |
| Silt  | 0.65 | 0.62 | 0.62 | 0.65 | 0.56 | 0.54 | 0.51 | 0.69 | 0.51 | 0.56 | 0.67 | 0.69 | 0.64      | 0.59      | 0.91     |      |      |
| Clay  | 0.44 | 0.48 | 0.56 | 0.36 | 0.45 | 0.46 | 0.45 | 0.49 | 0.41 | 0.3  | 0.31 | 0.33 | 0.17      | 0.25      | -0.6     | 0.23 |      |

To obtain more reliable information about the relationships among the variables, the factor analysis was applied [42-43]. Six significant components, whose eigenvalues are higher than one accounting for 90% of the cumulative variance, were distinguished for the analyzed data and shown in Figure-2. Factor one accounted for 23.6% of the total variance and is mainly characterized by high levels of carbonate, Hg, As, Fe, and Al. Factor two accounted for 19.5% of the total variance. It is characterized by strong negative loadings on Pb and moderated negative loading on Ni, Cu, Mn, and Cr. Factor three accounted for 19.5% of the total variance is mainly characterized by negative loadings on Co and Zn. Factor four is characterized by high positive loading on sand and negative loading on silt which explains the significant role of silt in binding certain metals with silt. Factor five shows high positive loadings on Cd. This may be attributed to independent behavior of Cd with little influence on total variability of metal. Factor six shows high positive loadings to clay. Generally, the results of correlation analysis and factor analysis coincide with each other and establish the role of org-C, carbonate and textural composition with metals in sediment.

Cluster analysis of data rendered a dendrogram (Fig. 3) summarizing seventeen variables (Pb, Ni, Cu, Mn, Zn, Co, Cd, Cr, Hg, As, Fe, Al, Org-C, carbonate, sand, silt and clay) for 60 sediment samples. The clustering resulted in two major groups (Groups A and B) and five subgroups, which were selected from the dendrogram of the cluster analysis (Figure 6). The subgroups were chosen from the dendrogram using similarity. First group includes Pb, Ni, Cu, Cr, Zn, Co, Cd and Clay and second groups includes Hg, CaCO<sub>3</sub>, As, Mn, Fe, Al, Org-C and Silt. These two clusters ultimately joins with sand. Pattern of joining clearly indicated influence of anthropogenic and natural activities on sediment contamination. The first subgroup of Pb, Ni, Ca and Cr and second subgroup of Zn and CO

are the indicative of anthropogenic practices (discharge of industrial waste and municipal waste) going on the adjacent landmass and upper catchment area.

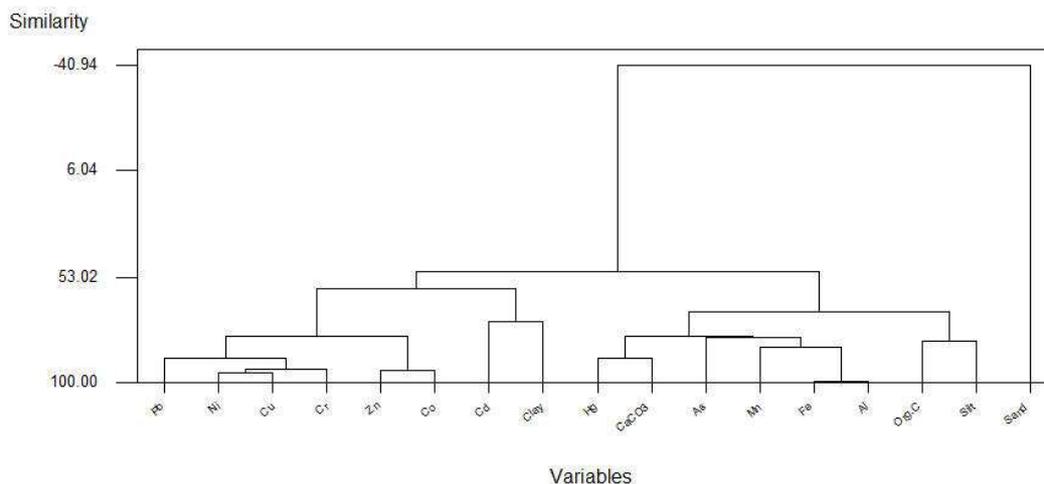
**Figure – 2 : Loading plot of Factor analysis showing relationship among metals, Org-C, carbonate and texture of Sediments**



The association of Cd and clay exhibits high affinity of Cd with Clay followed by moderate affinity of the metals in first and second subgroups. Association of these metals with Clay suggests that these metals are accumulated in the sediments by a process of complexation with clay present in the system. A high positive and significant linear correlations observed between these metals and clay (Table-4) confirm this finding. Second group clearly indicates the close association of Al, Mn, Fe, As, Hg, organic carbon and silt. The striking feature in third subgroup is that Hg is closely associated with carbonate indicating accumulation of Hg in carbonate debris present in the sediment. Fourth subgroup exhibited the close association of Al, Fe and Mn indicating their common origin and mainly from natural processes. Association of As with Fe, Mn and Al suggests that use of ground water for agriculture and domestic purposes in alluvial zone enhances their accumulation in sediment. This finding is the clear evidence of adverse impact of arsenic contaminated ground water in the adjacent landmass of Hugli estuarine system. Fifth subgroup shows close association between organic carbon and silt. Close association of Al, Fe and Mn and their high affinity with organic carbon and silt are the clear indication of their enrichment in sediment by natural process such as rock weathering. Linkage of carbonate with other metals in second group also suggests that moderate amount of shell/skeletal fragment act as scavengers for Fe, Al, Mn and As. Negative similarity of all these variables

in first and second group with sand revealed that accumulation of metals in estuarine sediment are governed by the sand. Level of metal concentrations would be linearly decreased with the increase of sand content in sediment.

**Fig-3 Dendrogram showing parameters groups formed by cluster analysis**



### CONCLUSION

Evaluation of heavy metal sources and their complex dynamics in sediments in Hugli estuary that supports the world's largest mangrove system, the Sundarbans, is an important environmental issue. The reporting of total metal concentrations appears to be misleading for formulation of environment management programme if interpretation of metal datasets is not done in relation with other sediment properties. The present study presents useful methods, for the evaluation of sediment contamination. This study will help to provides pertinent information to policy makers for framing environment management programme. Statistical techniques used in this study will provide guidelines for processing of data on chemical composition of sediments. Multivariate analyses, including distribution of data, the correlation matrix analysis, cluster analysis and factor analysis used in this study provide an important tool for better understanding the complex dynamics of pollutants. The correlation analysis of data shows strong positive correlation among the metal, organic carbon, texture and carbonate indicating their close association and their common origin. Multivariate analysis also helps to understand the geochemical processes governing the level of metal concentration in sediments and to partition the sources of metal (anthropogenic and natural). The mean concentration of the present study showed only lead contamination but enrichment factors indicated enrichment of Pb, Ni, Cd, Cu, Co and in sediments of Hugli estuary. The loadings of FA demonstrated that the variability of the metals in this zone was mainly governed by the sediment properties. Therefore the textural composition, carbonate and organic matter of freshly deposited sediments play crucial role in the sorption and complexation of transition metals.

The level of metal contamination in sediments clearly reflects the poor land use planning leading to soil erosion and resultant enrichment metals in sediments. The inherent physical condition of tidal dominated estuary accelerates the transportation of metal contaminated sediment to sea. This The input of sediment to coast may cause irreparable loss of coastal ecosystem . For actual evaluation of metal contamination in sediment, existing monitoring programme must be designed to include all the relevant parameters to focus the trend of metal contamination in sediment. Once the proper guidelines are prepared based on this study in the backdrop of other studies all over the world will answer the fundamental questions confronting enforcement agencies in cleaning the river Ganges.

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