INTRODUCTION

The study of nutrients and heavy metals in major river basins of the world is centered around their total concentration, chemical form and speciation in the dissolved and particulate forms, their flux to the oceans and their bioavailability. Davies (1992) summarized the present state of trace metal studies as: there is now a plethora of trace metal and nutrient data for soils, sediments, and plants, in contrast with the 1960s. But most of the case studies have taken place in the industrialized world, and the intertropical areas have been particularly neglected.

Nutrients and heavy metals are now extensively researched in life, agriculture, and environmental sciences. Until the 1960s, the major concern was micronutrient research (Davies 1992). Since then, the subject has broadened to encompass excess concentration of both micronutrient and heavy metals. This is primarily because of creeping land and water crises in the pipeline as a rapidly growing human population is dependent on the productivity of the landscapes which are already full of past mismanagement of waste, land and water (floods, water scarcity, water pollution, and water related land fertility degradation). Productivity of these landscapes is being increasingly stressed by over-exploitation of their resources (e.g., ground water, soil) and pollution of air and water (Falkenmark M, in Aswathanarayana 1995). Nutrients and heavy metals, when occur in excess in water and sediments, perturb the environment by disturbing the biogeochemical cycles significantly (Venugopal and Luckey 1975, Fergusson 1990).

Nutrients are chemical elements and compounds in the environment from which living beings synthesize living matters; their body cell and tissues, their genetic material, their energy bearing molecules, and their reproductive cells (Antweiler et al. 1996). Besides carbon, hydrogen and oxygen (the basic component of life), the elements which are required in higher quantities such as, Na, Ca, K, Mg, Si, N, P and
S - are called macro-nutrients. The micro-nutrients are Cu, Fe, Zn, Cl, B, Mo, Co and V etc. Insufficiency of any of these nutrients can limit growth of organisms. However, in the aquatic system, the micro-nutrients are always in excess, and N and P are the two most significant nutrients that commonly limit the productivity (Newbold 1992, Antweiler et al. 1996).

*Heavy metal* is a term represents a group of elements whose lower limiting density is 6 g.cm\(^{-3}\) (Wild 1988). However, to distinguish from alkali and alkaline earth metals, the metals exceeding the atomic mass of calcium are often referred to as 'heavy metals' (Förstner and Wittman 1981). Generally, the term nutrient refers to any inorganic material that is necessary for life and thus most of the heavy metals are nutrients too. In this study *trace elements, trace metals* and micro-nutrients will be treated synonymous with the term *heavy metal*.

**Biogeochemical cycle of nutrients and heavy metals**

Elements, both nutrients and heavy metals are recycled within and between the various components of the geoenvironment (lithosphere, hydrosphere, atmosphere, and biosphere). The recycling processes are energized by solar radiation (ultra-violet, visible and infra-red), mechanical (kinetic and potential), chemical and thermal energy of the earth (part of which is derived from the decay of uranium, thorium and \(^{40}\)potassium) (Aswathanarayana 1995). In most cases, the elements released during the process of weathering are transported and recombined in various ways. The transport mechanisms are depicted in Fig. 01.

The behavior of trace metals and nutrients in the aquatic system is complicated by the interactions between the dissolved and particulate matter under non-equilibrium condition (Fig. 02A-B). They may be precipitated through the interaction of water and solids, mixing with other kinds of waters and loss or addition of gases. The different types of precipitation reactions have been elaborated by Salomons and Förstner (1984).
Fig. 01 Transport mechanisms of metals in the geo-environment (after Fergusson 1990): a. volcanic activity, b. weathering, c. weathering, d. aerosol, e. fallout (solid), f. outgassing, g. gas adsorption, h. evaporation, i. precipitation, j. spray.
Fig. 02A Movement of trace metals in the hydrological cycle (after Salomons and Forstner 1984)

Fig. 02B Major processes and interactions between the dissolved and solid metal species in the surface water (Salomons and Forstner 1984).
The central issues concerning the nutrient cycling have mostly been studied in estuaries (Day et al. 1989) and lakes (Wetzel 1983). Meybeck (1982, 1993) has stressed the importance of rivers in nutrient cycling.

Autotrophs commonly are limited by the availability of certain chemical constituents, including C, N, P, Si, S, K, Mg, Na, Fe, Mn, Zn, Cu, B, Mo, V, and the vitamins thiamin, cyanocobalamin, and biotin (Hutchinson 1967). C, N, P, and Si are the most heavily utilized, and since C is typically abundant in water as dissolved CO$_2$, the usual presumption is that N, P, and Si are of primary importance (Allan 1995).

The various forms of nutrients in solution transform into the particulate phase by physical and chemical processes as well as by metabolic activities. Adsorption is especially important for highly charged ions such as phosphate and is reversible (desorption) depending on ion concentration and environmental conditions. Other important physico-chemical processes are the coalescing of colloidal-size particles into larger aggregates, termed flocculation; and chemical precipitation which occurs under oxidizing conditions and reverses under anaerobic conditions. The principal pathways of cycling of course differ among nutrients, a brief description of which can be seen in Allan (1995).

Large low land rivers often have their nutrient concentrations modified by industrial emission, sewage and agricultural effluent, and they are also likely to have their flow regime regulated by dams. The uniqueness of their catchment area, individual climate and geology make it difficult to generalize the characteristics of the drainage basins. However, studies regarding specific rivers can be seen in Whitton (1975), Davies and Walker (1986), Dodge (1989) etc.

Meybeck (1982, 1993) provided a comprehensive review of the various forms of carbon, nitrogen and phosphorus in world rivers. Natural levels of dissolved phosphorus are very low, around 0.01 mg.L$^{-1}$ for PO$_4^{3-}$ and 0.025 mg.L$^{-1}$ for total dissolved phosphate, which includes the organic form. Natural level of dissolved inorganic nitrogen are also low as compared with that in rivers affected by human activities - about 0.12 mg.L$^{-1}$, and nitrate is the major fraction. Human activities have
profundely altered nutrient levels in many of the world surface waters. Almost invariably, higher nutrient concentrations are found at the lowermost sites along large rivers, due to a variety of human activities. Human population density in the watersheds of 42 major rivers of the world was found to explain 76% of the variation in average annual nitrate concentration (Peierts et al. 1991). Nutrient concentrations often vary seasonally due to influences of hydrology, and changes in anthropogenic inputs (Allan 1995).

Importance of river sediment in nutrients and heavy metals study

The river sediment play an important role in the biogeochemical cycle of nutrients and heavy metals. Virtually all sediments (bed and suspended) in a river channel are supplied either by terrestrial erosion (runoff, mass flows, ice flows, eolian supply) or through the production of organic matter, biogenic carbonate and biogenic opal (Eisma 1993). These deposits reflect the biological, chemical and physical conditions of water body and become a signature to the state of nutrient and heavy metal concentration in the aqueous system. The establishment of nutrient and heavy metal level in sediments can play a key role in detecting their sources.

River sediments are a major sink as well as a source of nutrients and heavy metals. It has been observed that particulate phosphorus (organic and inorganic) represents 95% of phosphorus carried by the rivers (Meybeck 1982). Global particulate nitrogen transport by rivers amounts to 33 × 10^{12} g.N.a^{-1}, more than 80% of which occurs in rivers having high suspended matter concentration such as the Ganges, Brahmaputra, Mekong and Huanghe (Ittekkot and Zhang 1989). On a world average ca. 0.5 gigatons organic carbon is being transported by the rivers annually to the ocean. This transport, in general, is equally distributed between dissolved and particulate fraction of the riverine load (Spitzy and Ittekkot 1991). Again, it has been observed that trace element content in particulate matter is 1,000 to 100,000 times higher than the corresponding metal content in the aqueous phase (Förstner and
Wittman 1981), and more than 97% of the mass transport of metals is associated with sediments (Gibbs 1977). These facts imply that, river sediment plays an essential and significant role in the concentration and transportation of nutrients and heavy metals, and forms an integral part of the global geochemical cycle.

Bed sediments are sometimes called *in-place pollutants* because they represent a potential source of toxic elements and other biological effects which manifest themselves as direct effect on benthic organisms.

Bottom sediments of aquatic system become contaminated with toxic substances through a series of fate and transport processes acting on the contaminants introduced to the water column from external sources. Unlike many of the more conventional pollutants, toxic chemicals tend to have a strong affinity for particulate matters in aquatic system. Thus, depending on the chemical properties and characteristics of the receiving water body, much of the introduced contaminants are sorbed by biotic and abiotic suspended matter and settle from the water column and become a part of the bed sediments. Historically, this long term accumulation of contaminants in bottom sediments was considered a safe repository of these relatively insoluble toxic substances. However, recent studies have demonstrated that, when external loads to a water body have been eliminated, the recovery of the system is not govern strictly by washout from the water column. There is a much slower response controlling the long-term recovery of the system that is governed by the interaction of contaminated bottom sediments with the overlying water. There are numerous examples of this scenario in both freshwater and estuarine sediments (for detail see DePinto et al. 1994).

*River sediment load and variabilities*

The annual transport of water and sediments by the world rivers to the ocean has been estimated to be ca. $35 \times 10^3$ km$^3$ (Milliman 1991) and 15-20 billion tons (Milliman 1992) respectively. The physiographical and climatic features over the individual river
drainage basins are important factors controlling their discharge pattern. For instance, of the total water and sediment discharge by the world rivers, about 65% water and 80% sediment comes from the tropical and subtropical rivers draining S-E Asia, part of Oceania, and N-E South America (Milliman 1991) (Fig. 03). Half of the world river sediment is derived from the Himalayan region and its environs (Meade 1996). Among the Himalayan rivers, the Ganges-Brahmaputra river system carries one of the largest load of river sediment in the world. These rivers and the other Peninsular Indian rivers contribute annually about $2 \times 10^8$ tons (Milliman and Meade 1983) to $1.6 \times 10^9$ tons (Subramanian 1993a) of sediments into the Bay of Bengal. Milliman and Syvitsky (1992) observed that, when considering mainly large rivers, the contribution from numerous smaller rivers is underestimated. Material eroded in small river basins has a better chance to reach the sea than that in large river basins with flood plains and deltas. They estimated that prior to dam construction about $20 \times 10^{15}$ g.y$^{-1}$ of sediment was transported to sea. About half of this was supplied by rivers with basins smaller than 10,000 km$^2$.

The spatial and temporal variabilities in river dissolved load has been described by Walling and Webb (1983). River input of water and sediments to the ocean exhibits seasonal as well as spatial variability which is contributed by geological, climatological and anthropogenic factors. For example, influence of monsoon brings periodic high and low discharge conditions in the rivers, sometimes leading to massive floods. Seasonality associated with rising and falling stages of the river water resuspends and stores large quantities of fine grained sediments. This storage and resuspension are related to the mean slope of the flood wave on the river surface (Meade et al. 1985). Catastrophic events, e.g., tropical storms, may increase the quantity of sediment transported by small rivers, by one to several order of magnitude in a short period of time (Milliman 1991). The inputs from rivers of Indian subcontinent exhibit seasonal variations, with about 85-90% of it occurring during monsoon months (Ittekkot et al. 1986; Sarin et al. 1989). During the monsoon months of high discharge the Peninsular Indian rivers carry upto 95% of their annual sediment load (Vaithiyanathan et al. 1988),
Fig. 03 Annual fluvial sediment flux from large drainage basin areas to the oceans. Number in million of tons (per year); arrows proportional to the numbers (from Milliman and Meade 1983).
whereas the monsoon contribution to the Himalayan rivers, though very significant, is not necessarily that high in comparison (Subramanian 1993a).

Spatial variability of sediment load is evident in large river basins, such as the Ganges-Brahmaputra basin, primarily in response to the lithological and geomorphological variations (Subramanian 1993a). Meade (1994, 1996) has observed that in case of the Amazon and Orinoco rivers, suspended sediment concentrations are spatially heterogeneous because of the hydraulics of sediment transport and because inputs from tributaries that carry sediments in concentrations that differ from those of other tributaries or those of the mainstem river itself. Meybeck (1977) has studied the particle transport as a function of surface runoff and relief (Table 01). However, Eisma (1993) has pointed out that, the spatial variation in sediment transport in the Colorado river and the Nile are strongly influenced by dams, while the Yellow river and many of its tributaries receive very much sediment from a loess area of medium to low relief. Subramanian (1993a) observed that the dominant contribution of sediment load to the Godavari and the Mahanadi rivers come from sedimentary rocks and the easily weatherable igneous rocks (Deccan trap) though they may not occupy vast areas of the drainage basins. It has also been observed that the rivers in the Indian subcontinent show pronounced spatial variation in sediment load primarily in response to the river-bed slope and the gradual build-up of the urban areas (viz. the Ganges).

**Fate of sediments in the coastal zone**

Meade (1996) has pointed out that, although the sediment discharges of the world's large rivers are commonly reported in terms of delivery to the oceans (e.g. Milliman and Meade 1983), this practice can be misleading. A river usually delivers different quantities of sediment to its delta, to the coastline, to the continental shelf, and to the deep sea. Of the approximately $10 \times 10^8$ tonnes that the Amazon river deliver annually to its delta, about $2 \times 10^8$ tonnes are deposited in the delta, $6 \times 10^8$ tonnes are deposited on the continental shelf, and the remaining $2 \times 10^8$ tonnes are transported.
Table 01: Relation of particle transport \((Ts = \text{bed load} + \text{suspended matter in} \ t.km^{-2}.yr^{-1})\) with surface runoff \((q \ \text{in} \ l.s^{-1}.km^{-2})\) and relief (qualitative).
(After Meybeck 1977, quoted in Eisma 1993)

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<td>High runoff</td>
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<td>Low runoff</td>
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<td>High relief</td>
<td>Ts &gt; 100</td>
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<td>(Magdalena, alpine rivers)</td>
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<td>Medium relief</td>
<td>10 &lt; Ts &lt; 100</td>
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<td>Low relief</td>
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<td>(Quebec)</td>
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northwest ward along the coasts of the Guyanas; virtually no Amazon sediment reaches the deep sea under present condition. Of the $11 \times 10^6$ tonnes that the combined Ganges and Brahmaputra rivers deliver to Bangladesh during an average year, about $6 \times 10^8$ tonnes are deposited in the delta, about $3 \times 10^8$ tonnes are deposited on the continental shelf, about $1 \times 10^8$ tonnes may be transported across the shelf to be deposited in the deep sea. Of the $11 \times 10^8$ delivered by the Yellow river during a average year, $9 \times 10^8$ tonnes are deposited within the delta, and the remaining $2 \times 10^8$ tonnes are transported alongshore or deposited on the continental shelf, virtually all within the Gulf of Bohai (Meade 1996). Milliman (1991) also observed that for the ten largest rivers (which collectively contribute 36% of the fluvial load reaching the oceans) probably less than 25-30% escapes the shelf break; rather most remains in coastal areas. Of the ten rivers, only the Mississippi and the Ganges-Brahmaputra at present appear to discharge significant percentages of their sediment beyond the shelf break.

Studies regarding the transport and distribution of suspended sediments in the estuarine coastal region of the Bengal basin and on the distal part of the Bay of Bengal has been carried out by a number of authors (Emmel and Curray 1985, Kuehl et al. 1989, Barua 1990, Barua et al. 1994). It has been observed that during low discharge period the transport of suspended sediments is largely restricted to an area landward of about 20-isobath; however, during high discharge period the dominant transport direction is south-southwest (Barua et al. 1994). This sediment load may bypass the shelf and be deposited directly onto the adjacent slope, rise and fan (Kuehl et al. 1989). Emmel and Curray (1985) have observed, however, that the sediment load may even bypass the deepsea fan, implying that there could be a possible sediment sink at the outer (subareal) Bengal delta (Milliman 1991). Studies of sediments in the Bengal Fan at ODP Leg 116 cores at $1^\circ$S suggest that the sediments are first cycle detritus derived from high rate of denudation and rapid transformation to the distal Bengal Fan, and have been ascribed to a significant pulse of uplift and rapid erosion in the Himalayas (Stow et al. 1990).
While the physical aspects of the fate of suspended sediments of the GBM system in the Bay of Bengal is fairly known, the chemical aspects have not been studied at all. Studies from Hooghly estuary (Subramanian et al. 1987, Subramanian and Jha 1988, Subramanian et al. 1988, Subramanian 1993b) shows that the suspended sediments are enriched in heavy metals relative to that of the bed sediments. Moreover, the chemically mobile fractions such as organic and metallic hydroxide coatings have large proportion of heavy metals. Particle water exchanges and remobilization from sediments, are the two major mechanisms in the process of fresh water and salt water end members (Bewers and Yeats 1989). If this is also the case with the suspended sediments in the Bangladesh coastline, their release in water by constant agitation and by coastal physicochemical activities may have drastic and immediate consequences on the biological environment.

Source of river sediments

Sediments are the product of physical and chemical weathering on the continents (see for detail, Holland 1978). They begin their life cycle at the first contact between rocks and weathering processes, in which the water promotes chemical changes and provide physical transport (Golterman 1984). They may originate from igneous, metamorphic and sedimentary rocks, by physical and chemical weathering over the exposed surface. The addition of the water molecule restructures the rockmass into material having the capacity to adsorb; water converts primary rock material into a 'spongy mass'. Part of this process takes place during transport. Rivers not only transport sediments, they alter and add to sediment properties. The transfer of denudation product from the place of its origin to the river channel takes place at different span of time. Meade and Parker (1985) and Meade et al. (1990) observed that 90% of the soil eroded in the uplands in the United States are kept stored mostly in hillslopes, floodplain and other parts of stream valley somewhere between erosion sites and sea, and only 10% of the total sediments eroded from uplands are delivered to the ocean by the rivers. Thus in
a contemporary timescale the sediments lying at different storage sites along the way between the uplands and estuaries form the major and important source for rivers (Meade 1982). These sediments enter a river channel through overland flow (Statham 1977).

**Source of nutrients and heavy metals**

There are several sources of nutrients and trace metals in the environment, both natural and man-made. They may be soil parent material (rocks), commercial fertilizers, liming materials, sewage sludge, animal wastes, coal combustion residues, metal smelting industries, auto-emissions and others (Adriano 1986). With the exception of the parent material, all are anthropogenic in nature. The first seven sources are the primary input sources of agro-ecosystem. The rest may impact on natural ecosystem, as well as on urban and rural areas. From all these sources, nutrients and heavy metals enter river channel primarily by surface runoff and atmospheric fall-out.

Thus nutrients and heavy metals in aquatic ecosystem eventually get accumulated in sediments, where they may adversely affect the benthic biota, become a source of contamination in the water column, accumulate in biological tissues, and enter pelagic and human food chains. Contaminated sediments now appear to be the main source of toxins in many bays, lakes and rivers. Because of their potential adverse impact, the long periods of time associated with natural assimilation of many in-place contaminants, and the high cost of mitigation, sediments have become a focus of concern for many research and regulatory programs (Thomas 1994).

**Studies on nutrients and heavy metals in the Subcontinent**

Direct measurement of nutrients - C, N and P in suspended and dissolved loads - are rather limited in Indian rivers (Subramanian and Ittekkot 1991, Ittekkot et al. 1985).
However, nature and flux of organic matter in Indian rivers have been studied by Ittekkot and Arain (1986), Ittekkot et al. (1985), and Subramanian and Ittekkot (1991), Gupta (1996). Subramanian (1984), Vaithiyanathan et al. (1989) and Ramesh et al. (1995) studied the nature and flux of carbon and phosphorus by the major Indian rivers. Several studies have been carried out on the heavy metal distribution in the sediments of Indian rivers (Sarin et al. 1979, Borole et al. 1982, Subramanian et al. 1987, Ramesh et al. 1989, Subramanian et al. 1989, Ramesh et al. 1990, Jha et al. 1990, Biksham et al. 1991, Chakrapani and Subramanian 1993, Gupta and Subramanian 1994, Chakrapani and Subramanian 1995, Ramanathan and Subramanian 1995 and others). Most of the studies have reported only the bulk sediment concentrations, and metal speciation studies are scarce. So far, no attempt has been made to quantify the extent of metal pollution in the Indian rivers.

The Hooghly estuary have been studied for its geochemical and nutrient characteristics by a number of authors, important among them are Subramanian and Jha (1988), Modak et al. (1992), Subramanian (1993b). It is to be noted here that all these studies are limited to the Indian sector of the Bengal basin. Studies regarding texture and mineralogy, ion chemistry and particle-bound elemental and nutrient chemistry are either scarce or absent for the Bangladesh part of the Bengal basin which incidently covers more than 80% of the basin area and allows the passage of main channels of the Ganges-Brahmaputra-Meghna river system to the Bay of Bengal.

Morgan and McIntire (1959) briefly studied the grain-size distribution of the flood plain and adjoining terrace sediments of the basin and a short report on the grain-size distribution of the suspended sediments from some major rivers in Bangladesh is available in Kranck et al. (1993). Coleman (1969) studied the river morphology of the Brahmaputra in the Bengal basin. Jahan et al. (1990) have worked on grain-size distribution on bed sediments from two stations of the river Brahmaputra (Jamuna). Sarin et al. (1989) have reported on the clay mineralogy from two stations of the region.
The solute load of the Bengal basin, Bangladesh has received very little attention. Almost all the earlier works have focussed on the middle and upper region of the Ganges-Brahmaputra river system in the Indian part of the subcontinent or Tibetan region of the Brahmaputra (e.g., Hu et al. 1982, Sarin and Krishnaswami 1984, Subramanian 1987, Sarin et al. 1989, 1990 etc.). However, there are reports based on single station sampling - one each from the Padma and the Jamuna (the Bangladesh section of the Ganges and the Brahmaputra respectively) - on the nature and variability of the dissolved and particulate organic matter (Ittekkot et al. 1985, 1986), and on the major ion and isotope chemistry (Sarin et al. 1989, 1990) in Bangladesh. Carroll et al. (1993) studied the behavior of Ba and 226Ra in the Ganges-Brahmaputra mixing zone. No other geochemical studies have reported for Bengal basin, Bangladesh as yet.

Objectives

The Bengal basin represents one of the geologically youngest and tectonically active denudation regime of the world (Morgan and McIntire 1959, Valdiya 1984) and includes the second largest delta - the Bengal delta (Coleman 1981) - and the highest sediment dispersal system - the Ganges-Brahmaputra-Meghna (G-B-M) river system (Kuehl et al. 1989). The G-B-M river system (the lower segments of the Ganges and the Brahmaputra in Bengal basin, Bangladesh are called the Padma and the Jamuna respectively) has deep-sea connection through the Bengal Fan (Emmel and Curray 1985, Kuehl et al. 1989, Rea 1992, Barua et al. 1994) and has the potential to influence the tropical marine system (Ittekkot et al. 1986). The basin allows annually passage to an estimated 1330 km³ of water, more than 1060 million tons of total suspended solid (TSS) and more than 173 million tons of total dissolved solid (TDS) to the Bay of Bengal (Milliman et al. 1995). This region has low level of industrialization but is intensively cultivated and the population density is one of the highest in the world (400 - 1200 people per km²) (Milliman et al. 1989).
The size distribution of sediments, their mineralogy, and the hydro- and geochemical character of the basin are the major variables to be considered in a study related to the sediment and their environmental implications. There is a general dearth of knowledge about the biogeochemical characteristics of the Bengal basin, more so for the Bangladesh part of the basin. The present study is an attempt to generate enough baseline data for a proper biogeochemical evaluation of the Bengal basin and to understand the physical and chemical behavior of particle bound nutrients and heavy metals in the basin.

The following are the important aspects of the present study which aim at contributing to the broad objective of having a comprehensive understanding of the overall environmental biogeochemistry of the Bengal basin.

1. The texture and mineralogy of the bed and suspended sediments and their source to the river channels
2. The water chemistry and its controlling factors
3. Study of the elemental chemistry of bed, suspended and core sediments and their controlling factors
4. Distribution of C, N and P in the river bed sediments and their controlling factors
5. Fractionation of P in the bed sediments and its plant availability
6. Chemical and physical fractionation studies of heavy metals in bed sediments, and
7. Environmental Impact Assessment with respect to heavy metal enrichment in the basin