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The composition of suspended matter from Ganges–Brahmaputra sediment dispersal system during low sediment transport season

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Abstract

Suspended matter supplied from the Ganges–Brahmaputra river system was sampled in waters of the adjacent shelf area in times of low suspended matter load (November/December 1997). The absolute amount of suspended matter ranges between >500 mg/l near the river mouth and <1 mg/l on the outer shelf. Chemical and mineralogical analyses of the suspended matter show changes in its composition with distance from the source. Compositional changes are based on grain size partitioning; quartz and heavy minerals are precipitating sooner, leaving the suspended matter from the outer shelf depleted in quartz, REE, Zr, Hf, Nb, Ta, W, U, Th, Y, and Ca. The other part of the compositional changes is based on the reaction of suspended matter with seawater. Manganese hydroxide is producing coatings on suspended matter grains, and adsorbing elements out of the seawater. As a consequence, enrichment of Mn, Zn, Ni, Cr, Co, Li, Cs, As, Sb, Pb, Tl, and Cd can be observed in the suspended matter from the outer shelf. Arsenic is accumulating on the inner shelf by a factor of 2. Chemical partitioning in sequential leaching experiments shows that arsenic does not follow the enrichment mechanism of manganese and other transition metals. More than 90% of the total arsenic is bound to nearly insoluble phases of the suspended matter (e.g., crystalline iron oxides, residual minerals). For comparison with suspended matter, adjacent surface sediments were sampled and analysed. In this sediment, the dominating phase is the one which settled down during the times of high suspended load, with more quartz and heavy minerals and consequent chemical composition. The enrichment of manganese oxide and adsorbed transition metals gets lost; because of anaerobic conditions in the bottom-near seawater, they dissolve from the sediment and get recycled in seawater. Arsenic is found in nearly insoluble parts of the sediment, as it is the case in the suspended matter © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Suspended matter; Bay of Bengal; Composition; Phase distribution; REE patterns

1. Introduction

The Ganges and Brahmaputra Rivers unite with the Meghna River to form the Meghna Estuary before reaching the coast at the northern end of the Bay of Bengal. The combined load of these two rivers is reported to be the greatest sediment load of any river system (Holeman, 1968; Coleman, 1969; Milliman and Meade, 1983). The sediment load of the Ganges is estimated to be 520×10^6 tons/year and that of the Brahmaputra 540×10^6 tons/year (Milliman and Syvitski, 1992). The Meghna River has negligible impact to the sediment load, contributing only 1% to the total (Coleman, 1969). The strong seasonal changes which follow the annual hydrologic cycle are characteristic for the sediment discharge of the Ganges–Brahmaputra river system, with maximum discharge in August

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and minimum between January and March (Barua et al., 1994). The maximum and minimum annual sediment discharge loads differ by an order of magnitude (Coleman, 1969; Bangladesh Water Development Board, 1972).

The suspended matter discharges into a marine environment characterised by high energy due to strong tidal currents, seasonal monsoons and frequent cyclones. About one third of the discharged sediment remains on the shelf (Kuehl et al., 1997; Michels et al., 1998). The dynamics of the distribution of suspended sediment in the shallow shelf area off the Ganges–Brahmaputra mouth was investigated by Barua et al. (1994). These investigators showed that the main trend of sediment transport was to the south and west, which is in agreement with sedimentation rates of 1–>5 cm/year on the western shelf near "Swatch of No Ground" and less than 0.3 cm/year on the eastern shelf near Chitagong (Kuehl et al., 1997).

The chemical and mineral composition of the sediment load has not previously been investigated. The goal of this paper is to partly close this gap, offering mineral and chemical compositions of the suspended sediment load for the time of the year when sediment supply is low and to report the changes of sediment load composition with distance from the coast.

2. Sampling

The cruise SO126 was performed in November and December 1997 during the time of the year when sediment supply to the Bay of Bengal is at its lowest. Nine samples of suspended matter from the Ganges–Brahmaputra–Meghna River system were collected on the shelf at different distances from the river mouth (water depth between 1 and 50 m), representing the mixing zone (Fig. 1, PZ-samples; Table 1). On this part of shelf the top water layer, about 50 m deep, is composed of warm (>28 °C), oxygen-rich water of low salinity (< 30‰). The bottom water layer is composed of cold (< 20 °C) sea water (salinity 34.5‰) free of oxygen. There is a sharp boundary between these two water layers (Karstensen et al., 1998, cruise report).

The vertical distribution of suspended matter in the water column was detected with multiprobe profiles (Karstensen et al., 1998, cruise report). High concentrations were indicated by changes in light attenuation. From the depth of maximum light attenuation (sampling depth see Table 1), water was collected with a pump (Grundfos Jetsub JS 2-04; max. flow of 1.6 $m^3\ h^{-1}$), and fed through a flow-through centrifuge that separated the solids from water. The flow of water was set at 10 l/min. To separate suspended matter, water was injected into a fast rotating cylinder (20,000 rpm) in which solid particles were accumulated. Depending on the concentration of suspended matter, the sampled volume of water varied between 150 and 1500 l. After centrifugation, the samples were flushed out of the cylinder with deionised water. The suspensions were subsequently filtered through a 0.45-μm filter and dried on board the RV Sonne at 40 °C. The total concentration of suspended matter was calculated from the dry weight. Due to long sampling time (between 15 and 150 min), only a limited number of locations in the Bay of Bengal could be sampled. In all cases, only one of the observed water layers with high suspended water load (multiprobe measurements) was sampled at each location.

To verify uncontaminated sampling, water samples were taken from the water stream leaving the centrifuge. These samples were acidified, stored, and later analysed with ICP-MS/ICP-OES (for details see Analytical methods). From the data of these water samples, it could be shown that the sampling procedure using a flow-through centrifuge unfortunately leads to a contamination of the sampled suspended matter. The elements molybdenum, copper and tin are enriched in the water samples leaving the centrifuge, so that data for these elements could not be used for suspended matter samples gained by centrifugation. A possible contaminating source for these elements is the material of the bearing of the rotating cylinder and its lubrication (MoS_2).

Sediment samples from the sediment/water interface were recovered from locations adjacent to where the suspended matter samples were collected in order to characterise changes in the chemical and mineralogical composition between suspended matter and sediments (Fig. 1, KL-, KH-, and SL-samples). Three sorts of devices were used for sampling sediment: long box corer KH (4 m), gravity corer SL (5 m), and piston corer KL (10–20 m). All the sediments were gained immediately after the sampling device reached the shipboard laboratory. In case of undisturbed surface the "semi-liquid" layer was sampled. Depending on type of sediment the semi-liquid layer was 2 to 3 cm thick. In cases of disturbed sediment surface, larger samples were

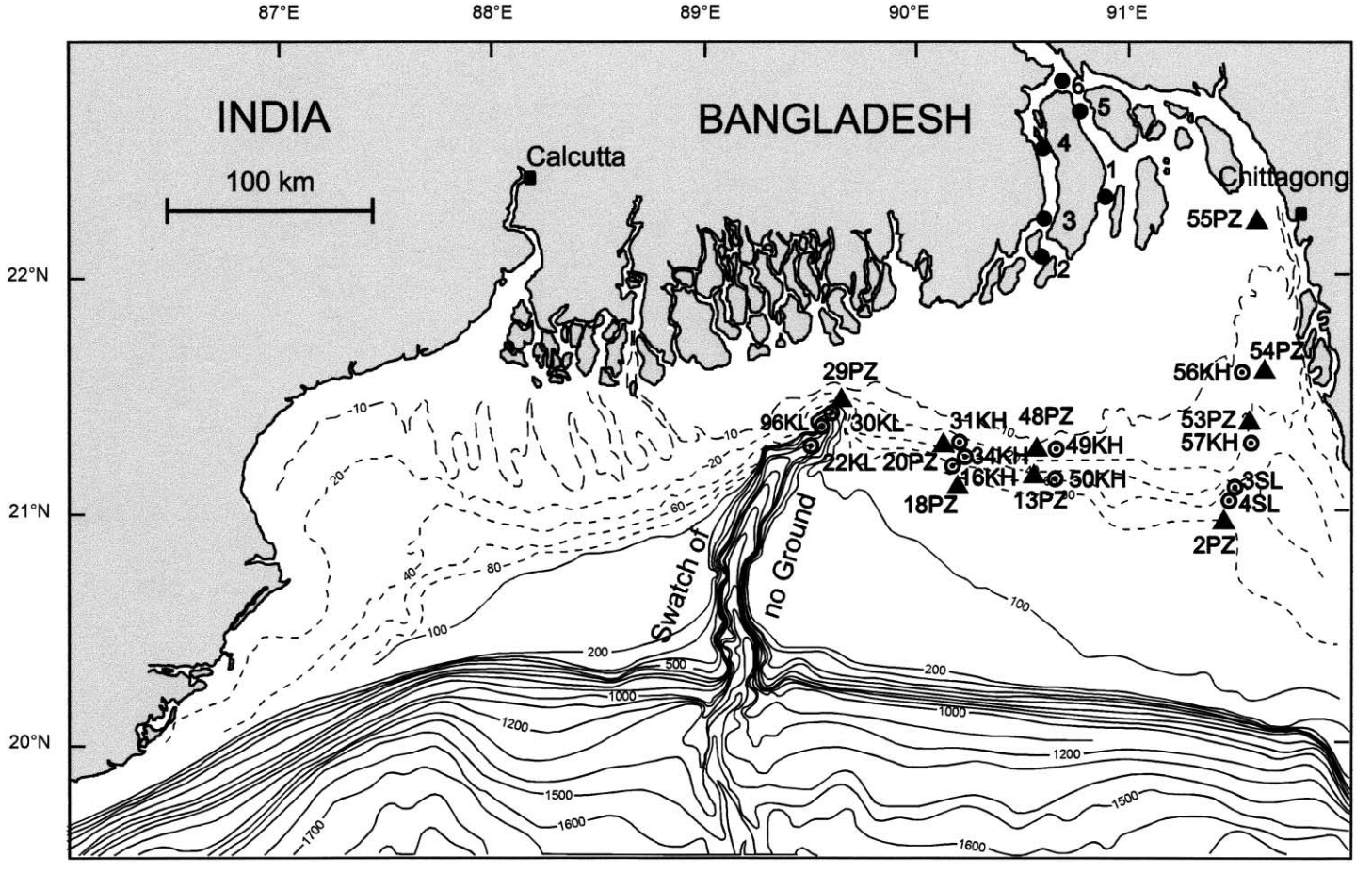


Fig. 1. Map of the sampling area in the Bay of Bengal with sampling positions for suspended matter (PZ) and sediment samples (KH, SL, KL).

Table 1
Suspended matter samples

Sample name	Water depth (m)	Sampling depth (m below surface)	Salinity (‰)	Suspended material concentration (mg/l)	Sampling site characterisation
55 PZ	12	1	18.3	535.4	strong fluvial influence, near Chittagong, high suspended matter load
54 PZ	20	5	29.4	69.3	strong southward current, high suspended matter load
53 PZ	23	5	31.8	9.44	strong southward current, intermediate suspended matter load
02 PZ	58	42	34.9	5.3	near bottom maximum, intermediate suspended matter load
48 PZ	16	13	28.8	2.1	near bottom maximum, intermediate suspended matter load
29 PZ	60	44	34.3	2.3	low oxygen-high salinity water, intermediate suspended matter load
20 PZ	28	22	31.4	0.88	layer of decreasing oxygen, transmission and temperature, low suspended matter load
18 PZ	54	47	34.5	0.91	near bottom maximum, low suspended matter load
13 PZ	45	37	35.3	0.75	near bottom maximum, low suspended matter load

collected that were up to 10 cm thick. In a part of sediment cores, the adjacent layer was also sampled to obtain information about the homogeneity of recent sediment. The sediments were transferred to the laboratory without any pretreatment on board. All sediment samples are of recent age, and the outcropping of drowned Pleistocene beach barriers in the area (Wiedicke et al., 1999; Michels et al., 1998) was well mapped and avoided by sampling.

In addition, six samples of sediments from the Meghna Estuary were supplied by our Bangladesh counterparts.

3. Analytical methods

The semiquantitative determination of mineral composition of the samples was performed with X-ray diffraction. The quantitative determination of quartz was performed following a method described by Meyer and Klosa (1997). The results are listed in Appendix A.

Microscopic investigations of the estuary sediments were performed on bulk samples. In the case of two samples (the one with lowest and the other one with highest heavy mineral content), separated fractions were obtained. Grain size fractions of these two samples, obtained by means of wet sieving, passed further separations to heavy and light mineral fractions, first with a solution of specific weight 3 kg/l, and subsequently with magnetic purification of the light fraction from the remaining composite aggregates. The fractions were investigated microscopically and were analysed with XRF and ICP-MS (see below).

Sediment samples, including fractions from two estuary sediments, were transferred to the laboratory, dried at 40 °C, and milled to less than 40 μm particle size in an agate mill. Bulk chemical analyses of suspended matter and sediment samples were performed with X-ray fluorescence (XRF) using Philips PW 2400 and PW 1480 wavelength dispersive spectrometers. Forty-two major and trace elements were quantitatively analysed after fusion of the samples with lithiummetaborate at 1200 °C for 20 min (sample/$LiBO_2$ = 1/5). Quality of the results was controlled with certified reference materials (CRM) (i.e., BCR, Community Bureau of Reference, Brussels). This procedure ensures an analytical precision better than ±0.5% relative for major elements and 1–10 mg/kg for trace elements of the analysed certified reference materials.

Trace elements and rare earth elements were analysed with ICP-MS (Perkin Elmer Sciex Elan 5000) and ICP-OES (Jobin Yvon JY 166 Ultrace) after wet chemical decomposition of the samples in a closed microwave system (3 ml HF, 1 ml HNO_3 suprapure quality,

0.5 g sample). After decomposition the solutions were treated with $HClO_4$ to remove HF. Quality of the results was controlled using CRM (e.g., NIST CRM 1643d). For major and trace elements, an analytical precision of ± 5% was achieved (results listed in Appendix A).

The phase distribution of elements in the samples was determined in a four-step sequential leaching procedure, which was slightly modified to exclude element interference in the following ICP-OES determination of trace elements (Tessier et al., 1979).

The sequential leaching procedure was performed as follows:

(1) Exchangeable adsorbed elements: 1 g of powdered and dried sample was leached for 1 h with 20 ml 1 M sodium–acetate solution (pH = 7). After leaching the samples were centrifuged and washed with water.

(2) Weak acid leachable elements, carbonates: the residue of step 1 was treated with 20 ml of a 0.1 M acetic acid solution for 5 h. After centrifugation the residue was washed with water.

(3) Easily reducible fraction (including Mn-oxides and co-precipitated elements): the residue of step 2 was treated with 20 ml of a 0.04 M hydroxylamine–hydrochloride solution (NH_2OH/HCl; pH = 2). The sample was shaken for 6 h at a temperature of 95 °C. The residue was again centrifuged and washed with water.

(4) Strong acid soluble elements: the residue of step 3 was treated with 4 ml boiling aqua regia (1 ml HNO_3 + 3 ml HCl suprapure quality).

(5) The so-called "residual" fraction of the samples was analysed with XRF after passing the sequential leaching procedure as described above.

All reagents used were of suprapure grade, and possible contamination of the resulting solutions was checked using reagent blanks. The solutions of the leaching steps were diluted with water to a volume of 50 ml. Element concentrations were analysed using ICP-OES. The standard deviation of ICP-OES measurements was generally better than 10%, determined by multiple extractions of selected samples. The recovery of elements in the sequential extraction procedure ranged between 90% and 110% for major elements and 80% and 120% for trace elements compared to XRF bulk analyses of the samples.

Arsenic was analysed with a hydride generation procedure using a Perkin Elmer Atomic Absorption Spectrometer (type Aanalyst 100) with a flow-injection system (FIAS 400) following a procedure described in the manual. Standard deviation ranged between 5% and 15% relatively; quality was proofed using CRM NIST 1643d.

4. Results and discussion

4.1. Mineralogical composition of suspended matter and sediments

The suspended matter contains between 9% and 45% quartz (average 23 ± 13%). Differences can be observed between quartz content of suspended matter sampled near the river mouth (19–45%; average 28 ± 11%) and remote suspended matter (9–15% quartz; average 12 ± 3%). Generally, the quartz content of suspended matter decreases with distance from the river mouth. The content of quartz is used as an indicator for the grain size distribution of the sample material, because the limited amount of suspended matter samples did not allow grain size analyses. Other mineral components of the suspended matter include feldspar, muscovite and chlorite.

The recent surface inner shelf sediment contains between 21% and 49% quartz (average 30 ± 9%), i.e., the quartz content is higher than in the suspended matter. Other mineral components occur in similar amounts in these sediments as in the suspended matter.

The sediment sampled in Meghna Estuary contains significantly higher amounts of quartz (average 51 ± 3%) and feldspar, and significantly less muscovite–illite compared with suspended matter and shelf sediments. Hornblende is present in Meghna Estuary sediments, whereas the existence of kaolinite has not been verified. Chlorite could be detected at trace levels. Dolomite was not observed.

The following grain size results were obtained for two Meghna Estuary samples (sample 4 with lowest heavy mineral content and sample 6 with highest heavy mineral content):

Grain size	355–200 μm	200–112 μm	112–63 μm	<63 μm
Sample 4	26%	48%	19%	7%
Sample 6	21%	40%	26%	13%

Consequently, these results classify them as fine sands.

The microscopy of Meghna Estuary sediments shows that they reveal minimum damage from transport. This is normal for the silt size fraction, but in the sand fraction, which is typically more sensitive to transport damages, only very few of the grains are weakly rounded. The main minerals of the Meghna Estuary sediments are quartz (40–55%), plagioclase (14–21%), alkali feldspar (2–10%), muscovite (illite) (2–7%), clay minerals like chlorite (kaolinite) and illite (4–10%), and heavy minerals (5–25%). The usually untwinned plagioclase shows a wide range of An-contents, with maximum An-35 and an average of An-25 (calculated from the XRF data of light mineral concentrates >20 μm). Alkali feldspar is also mostly untwinned, with occasional microcline or perthite twinning.

The heavy mineral assemblage of Meghna Estuary sediment samples contains high amounts of mixed green amphiboles, hereafter referred to as green hornblende. Following the amphiboles, the next most common minerals are members of the epidote family and colourless to reddish garnet. Occasionally biotite could be found in higher amounts. Moreover, the following accessory (<5%) or trace minerals were observed: opaques (ilmenite, magnetite), OPX, CPX, zircon, tourmaline, apatite, rutile, brookite, sphene, allonite, staurolite, and kyanite. Metamorphic and intermediate to acid magmatic rocks that occur within the drainage area are the main source of the minerals observed in Meghna Estuary sediments. Evidence of moderate to weak weathering could be observed on pyroxenes, the minerals from the estuary assemblage which are most prone to weathering. Other more weathering-resistant minerals were fresh in appearance.

We explain the differences in the mineral composition between suspended matter and shelf sediments on the one side, and estuary sediments on the other side with different energetic levels of the environment, causing differences in grain size sorting of particles. The composition of the shelf sediment samples is not identical with the suspended matter because sedimentation during the high supply season is the dominant factor in deposition of the shelf sediment. The amount of suspended matter from the low supply season is significantly lower (1% of the annual discharge for the one month frame of our expedition due to determination of Barua et al., 1994). The suspended matter from the high supply season is expected to have higher amounts of quartz and other coarse detritus because it was transported during high water input (i.e., during higher energetic levels).

The Meghna Estuary sediment could be termed as "residual sand". The coarser mineral grains are concentrated, and the clay minerals were washed out or not deposited at all. As the minerals from the source rocks are well preserved, the transport must have been short or gentle. Chemical weathering is not advanced either. It is known from the analyses of dissolved Sr and Li isotopes that the weathering of the Himalaya was so extreme that it changed the isotopic signals of seawater via river transport (Edmond, 1992; Krishnaswami et al., 1992; Huh et al., 1998). This chemical weathering must have taken place preferentially in the clay mineral fraction and shows only little effect on the fine sand that accumulates in the arms of the estuary.

4.2. Partition of suspended matter and surface sediments based on their geographic position

The suspended matter and the adjacent surface sediments in the Bay of Bengal can be divided in different groups according to sampling distance from the river mouth, as follows.

4.2.1. Suspended matter and surface sediments sampled close to the river mouth (suspended matter samples 54PZ, 55PZ, sediment top of core 56KH, 57KH)

This area is characterised by a high suspended matter load (up to 500 mg/l); the composition of this suspended matter is similar to the original river particulate matter (Subramanian et al., 1985; Schmitz, 1987). The analyses of river particular matter have not been published yet for the Ganges–Brahmaputra system; Martin and Meybeck (1979) calculate their "geochemical flux of elements associated with RPM" for the river Ganges with the analysis of one "river bank", i.e., "freshly deposited sediment" near Allahabad. Subramanian et al. (1985) and Schmitz (1987) draw their conclusions from the composition of river sediments from the Ganges by using the analysis of the river bank sediment of Martin and Meybeck (1979). Therefore, we cannot conclude with confidence that our samples of suspended matter close to the river mouth are identical with river suspended matter, but they do exhibit large similarity with the river bank near Allahabad (Martin and Meybeck,

Table 2
Composition of suspended matter

Element	Average composition of suspended river matter (mg/kg) (Martin and Meybeck, 1979)	Composition of suspended matter sampled at the river mouth (mg/kg) (this study)
Ag	0.07	(0.1)
Al	94,000	101,000 ± 500
As	5	15 ± 1.5
Ba	600	470 ± 15
Be	–	(3)
Bi	–	(1)
Ca	21,500	7600 ± 700
Cd	(1)	(0.1)
Co	20	21 ± 1
Cr	100	120 ± 3
Cs	6	11 ± 1
Fe	48,000	56,000 ± 1000
Ga	25	23 ± 1
Hf	6	3.2 ± 0.1
K	20,000	27,500 ± 200
Li	25	67 ± 4
Mg	11,800	19,200 ± 200
Mn	1050	940 ± 25
Na	–	11,000 ± 2000
Nb	–	17.3 ± 0.2
Ni	90	69 ± 4
Pb	100	30 ± 3
Rb	100	178 ± 2
Sb	2.5	(0.8)
Sc	18	21.5 ± 0.5
Sr	150	97 ± 1
Ta	1.25	1.7 ± 0.1
Th	14	11 ± 2
Ti	–	5300 ± 10
Tl	–	(0.9)
U	3	2 ± 0.2
V	170	132 ± 2
W	–	9 ± 4
Y	30	35 ± 1
Zn	250	120 ± 10
Zr	–	124 ± 2
REE		
La	45	19 ± 2
Ce	95	26 ± 2
Pr	(8)	6 ± 0.6
Nd	35	21 ± 2
Sm	7	4 ± 0.4
Eu	1.5	1 ± 0.1
Gd	(5)	4 ± 0.4
Tb	1	0.7 ± 0.05
Dy	–	4 ± 0.3
Ho	(1)	0.8 ± 0.05
Er	(3)	2.5 ± 0.2
Tm	(0.4)	0.4 ± 0.02
Yb	3.5	2.5 ± 0.15
Lu	0.5	0.4 ± 0.02

1979), and with average upper continental crust (Taylor and McLennan, 1981).

4.2.2. Suspended matter and surface sediment sampled near the coast (suspended matter samples 2PZ, 29PZ, 48PZ, 53PZ, surface sediments of cores 3SL, 4SL, 96KL, 22KL, 30KL, 49KH)

The suspended matter load of the water in this area ranges between 2 and 10 mg/l. During transport from the river mouth to the inner shelf, the suspended matter load of the water decreases by a factor of 50–250.

4.2.3. Suspended matter and surface sediment sampled far from the coast (suspended matter samples 13PZ, 18PZ, 20PZ, surface sediment of cores 50KH, 16KH, 31KH, 34KH)

With increasing distance from the river mouth, the suspended matter content decreases down to concentrations below 1 mg/l.

4.3. Chemical composition of suspended matter

The average bulk chemical composition of suspended matter from the river mouth is very close to the worldwide average composition of suspended river material (Martin and Meybeck, 1979; Martin and Whitfield, 1983) (Table 2). Our data suggest that during transport through the large drainage area of the river system with its different geological zones, the mixing process equalises the composition of the suspended matter load so effectively that at the river mouth it is similar to the worldwide average (i.e., it is a representative mixture of well homogenised continental upper crust rocks; Schmitz, 1987).

The chemical composition of suspended matter from the river mouth changes with increasing distance from the source. The depletion of quartz during transport of suspended matter into regions far from coast (as discussed above) is the main reason for changes in

the elemental composition of suspended matter; the changes are initialised by grain size fractionation during sedimentation.

To characterise geochemical reactions leading to enrichment or depletion of elements in the suspended matter, the element concentrations of the quartz-free fraction of suspended matter were calculated from the bulk analytical results. The factor f was calculated:

$$f = \frac{\text{element concentration of samples (calculated free of quartz)}}{\text{element concentration of estuary sediments (calculated free of quartz)}}$$

The results are shown in Fig. 2.

Compared to estuarine sediment, the suspended matter is enriched as follows: Mn, Zn ≫ Ni>Li, Cs, As, Sb, Pb, Tl, Cr, Co, Cd (Fig. 2).

The enrichment of most of these elements in the non-quartz fraction can be explained by the growth of Mn-oxy-hydroxide coatings on the surface of suspended matter particles. For suspended matter sampled far from the coast (white column in Fig. 2), manganese is enriched by a factor of 7 compared to the estuary sediment. The concentration of manganese in suspended matter sampled near to the river mouth shows no difference compared to the estuary sediments. The transition metals Zn, Ni, Cr, Co, Pb, and Cd follow a similar enrichment mechanism. The phase distribution of these elements and manganese determined in sequential leaching experiments (Fig. 3a: manganese; Fig. 3b zinc) changes with distance from the river mouth and parallel to the longer residence time of particles in the water column. The transport time of particles from the estuary to the outer shelf enables the growth of manganese oxy- hydroxides on their surfaces in an oxic environment (Karstensen et al., 1998). The indication for the described enrichment mechanism is the complete change of the phase distribution of manganese in suspended matter from the river mouth to those sampled far from the coast. In samples from the river mouth, 60% of the manganese is found in a soluble form while 40% occurs in the residual fraction (Fig. 3a). In suspended matter sampled far from the coast, nearly 100% of manganese occurs in a soluble form. Of the total soluble content, 70% can be mobilised under reducing conditions. Other metals behave similarly, suggesting that manganese coatings on the surfaces of suspended matter particles act as trace metal adsorbers.

Arsenic is enriched in the suspended matter samples by a factor of 1.5–2 (Fig. 2), but the arsenic enrich-

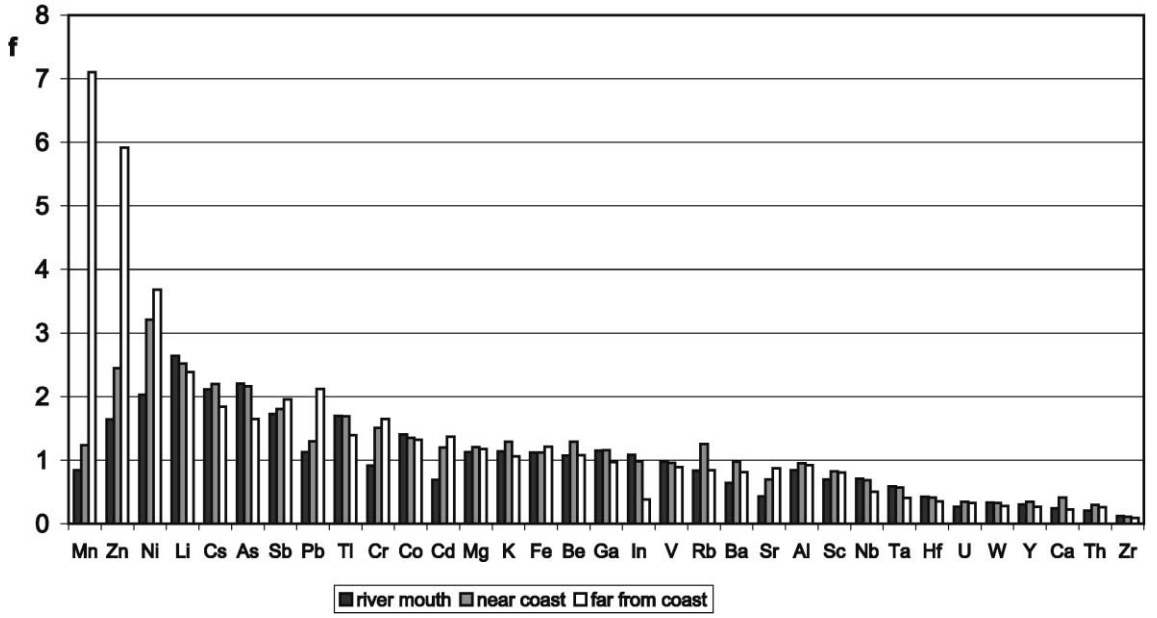


Fig. 2. Average enrichment/depletion of elements in the suspended matter against average Meghna Estuary sediment composition. All the chemical compositions are calculated on a quartz-free basis. The suspended matter is divided into three groups: (a) sampled close to the river mouth, (b) sampled near the coast, (c) sampled far from the coast.

ment does not follow the enrichment mechanism of manganese and other transition metals (Zn, Ni, Cr, Co,). In contrast to these elements, arsenic is found to be hosted in the insoluble, residual phase of the suspended matter matrix (Fig. 3c). The phase distribution of Arsenic is more similar to those of iron (Fig. 3d), with no changes due to different redox conditions in the water column.

Another group of elements, including Mg, K, Fe, Be, Ga, In, V, Rb, Ba, Sr, and Al, is not influenced during the transport process. No changes in their concentration with increasing distance from the river mouth could be observed (Fig. 2). Results from the sequential leaching procedure indicate, that the chemical behaviour of these elements is constant during the transport of material from the estuary to the open sea (Fig. 3d, Fe). Our data suggest that these elements do not take part in reactions between particles and the water column in the Bay of Bengal even under changing redox conditions.

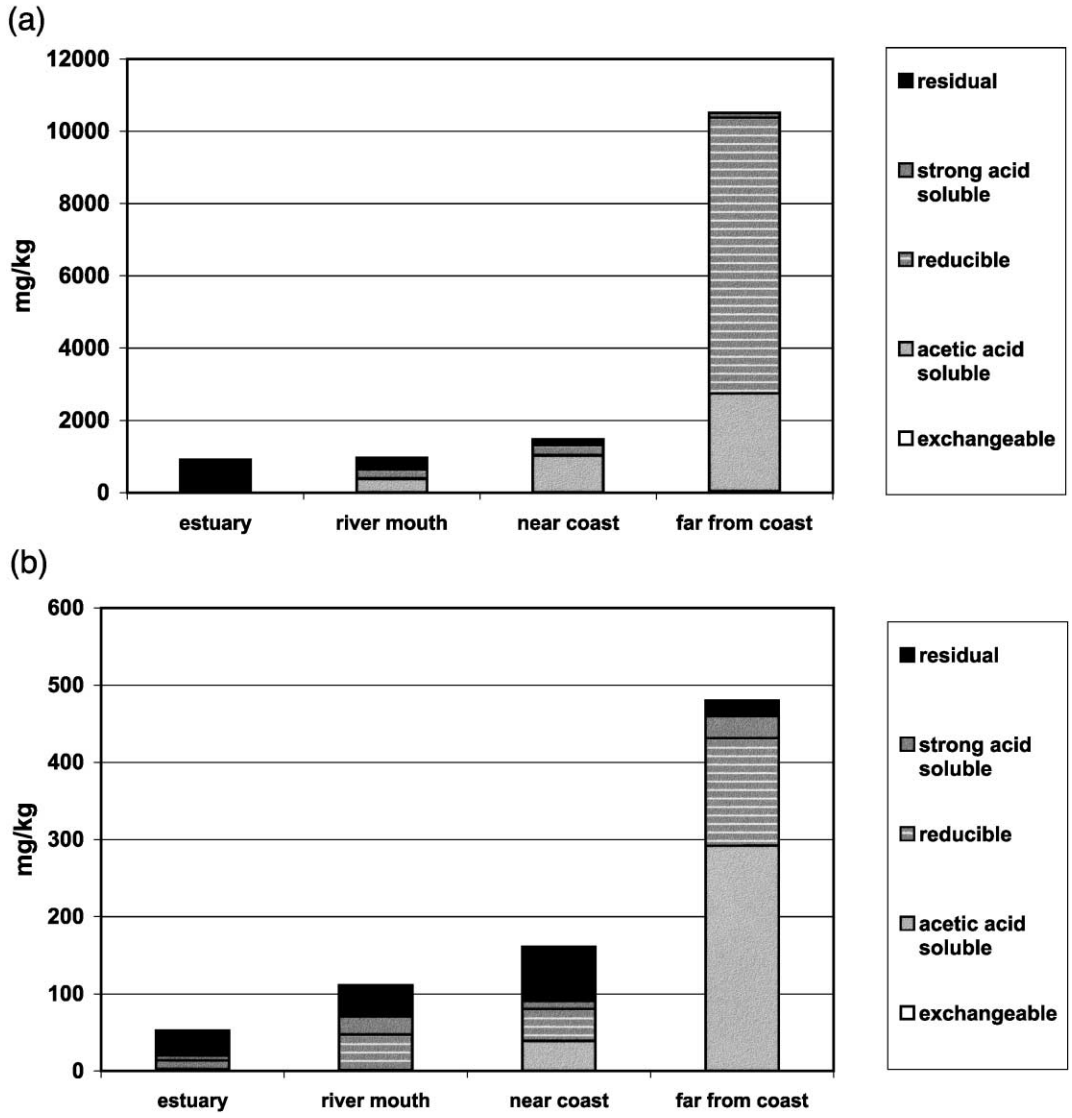


Fig. 3. Phase distribution of selected elements in suspended matter samples with increasing distance from the river mouth: (a) manganese, (b) zinc, (c) arsenic, (d) iron.

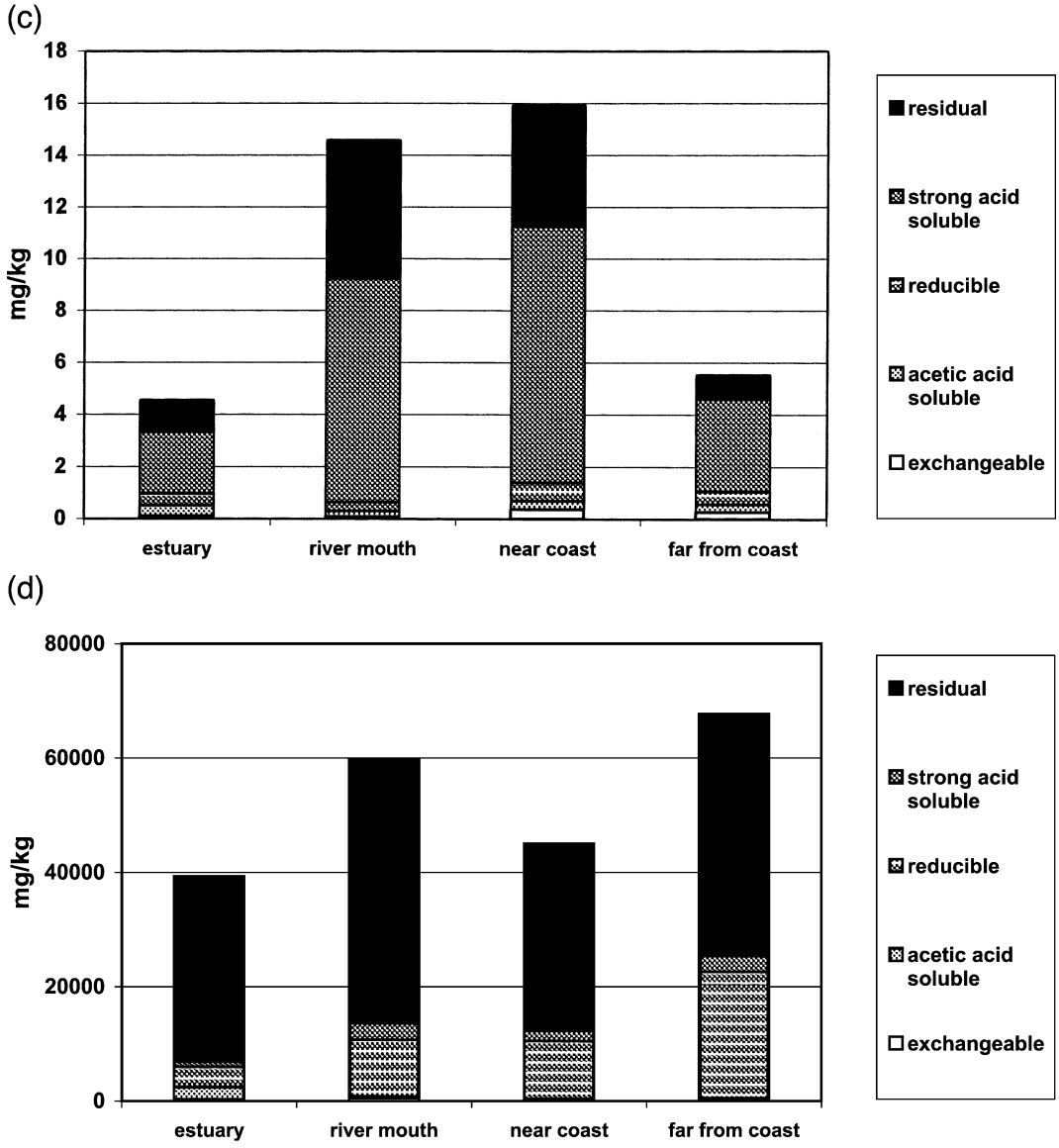


Fig. 3 (*continued*).

The third group of elements, containing Zr, Hf, Nb, Ta, W, Th, and U, are being removed from the suspended matter in the course of transport from the estuary to the open ocean (Fig. 2). These elements, incorporated in the heavy mineral phase of the suspended matter, are not taking part in any chemical reactions, and therefore are associated with the residual insoluble fraction. Their host phases (i.e., heavy minerals) are mechanically separated from the suspended matter early in the transport process.

4.4. Chemical composition of surface sediments

The concentration ratio of elements in shelf sediments to elements in the estuary sediment (on quartz-free basis) is shown in Fig. 4. Copper, Li, Cs, and As are

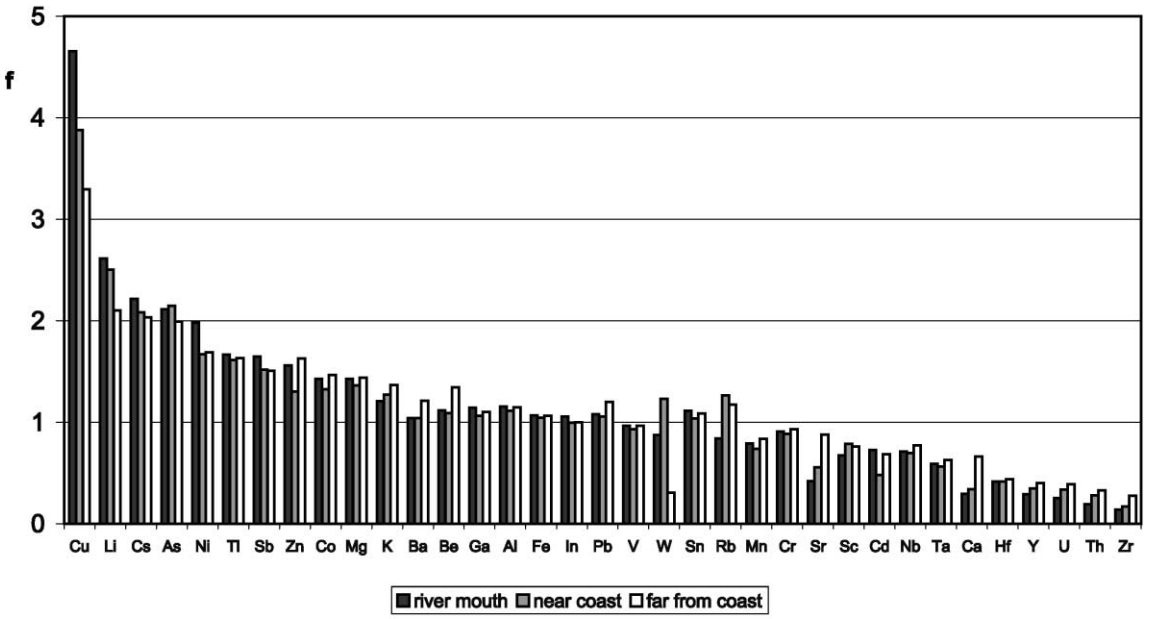


Fig. 4. Average enrichment/depletion of elements in sediment tops against average Meghna Estuary sediment composition. All chemical compositions are calculated on a quartz-free basis. The sediments are divided into three groups following the sampling positions of suspended matter.

significantly enriched in shelf sediments by more than a factor 2. We suggest that this is caused by an enrichment of the fine fraction in marine sediments compared to estuary sediments. More than 50% of the amount of enriched elements are insolubly integrated in the fine fraction (e.g., Fig. 5a, Cu).

Arsenic is, as manganese, a redox-sensitive element which can be transferred from As(V) (arsenate) to As(III) (arsenite) within a reducing sediment (Nickson et al., 2000). Arsenic is enriched even in the suspended matter under oxic conditions and in the sediment top under reducing conditions; obviously there is no cycling of arsenic in the water column nor in the reducing sediments on the Bangladesh shelf. In both types of samples As is found in the acid soluble and residual phase of the material. The behaviour of As is quite similar to the behaviour of iron (Fig. 5c,d). This is, on the one hand, in agreement with the fact that arsenic off Bangladesh groundwater is not bound to manganese oxides but hosted to iron oxides. On the other hand, the behaviour of arsenic and iron in suspended matter and sediment top samples is in contradiction with the observation that both elements are released together from Bangladesh groundwater under reducing conditions (Brannon and Partrick, 1987; Peterson and Carpenter, 1986).

The lack of enrichment in Mn, Zn, Ni, Pb, Cr, Cd in the sediment top with increasing distance from the coast suggests that the sediment settles within a reducing environment. The manganese oxide coatings, which already stained the ''far from coast'' suspended matter, are thus dissolved after sedimentation (Fig. 5b, Mn). This is in accordance with the oceanographic investigations from Karstensen et al.(1998), which shows on two profiles from the general study region that the seawater is void of oxygen at depths greater than 50 and 35 m, respectively. The high Mn/Fe ratios observed in the suspended matter sampled far from the coast indicate that the major part of the enriched metals originates from remobilisation from the sediments. Only a minor part is formed by the hydrogenous Mn-oxide precipitation process known from deep-sea areas with low sedimentation rate (Elderfield, 1977).

Another group of elements (i.e., K, Ba, Be, Ga, Al, Fe, In, Pb, and V) occur in a nearly insoluble form,

therefore the concentrations of these elements compared to estuarine samples do not change with increasing distance to the coast (i.e., Fig. 5d Fe).

The elements Zr, Th, U, Y, and Hf exhibit the greatest depletion in sediments from the shelf when compared to estuary sediments (Fig. 4). This likely reflects the effect of grain size differences, as in the case of the suspended matter. The heavy minerals, which host these elements, are enriched in estuary sediments. In the marine sediments, they are diluted by the fine sediment fraction with distance from the river mouth.

The elements Ca and Sr are also depleted in sediments from the shelf when compared with estuary sediment, and, although their depletion is not as pronounced as in the case of the heavy mineral hosted elements (Zr, Th, U, Y, and Hf), the differences

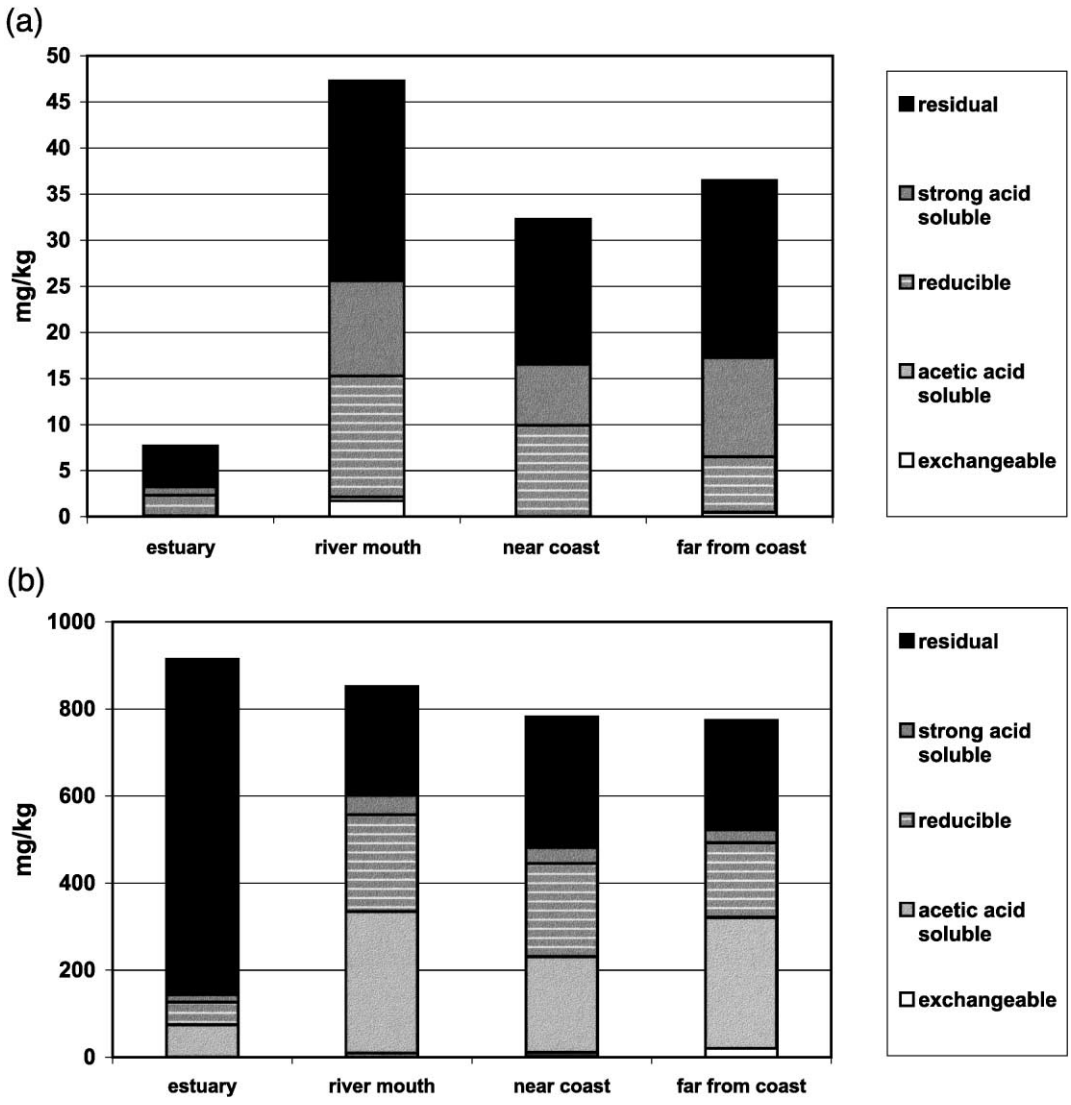


Fig. 5. Phase distribution of selected elements in sediment top samples with increasing distance from the river mouth: (a) copper, (b) manganese, (c) arsenic, (d) iron.

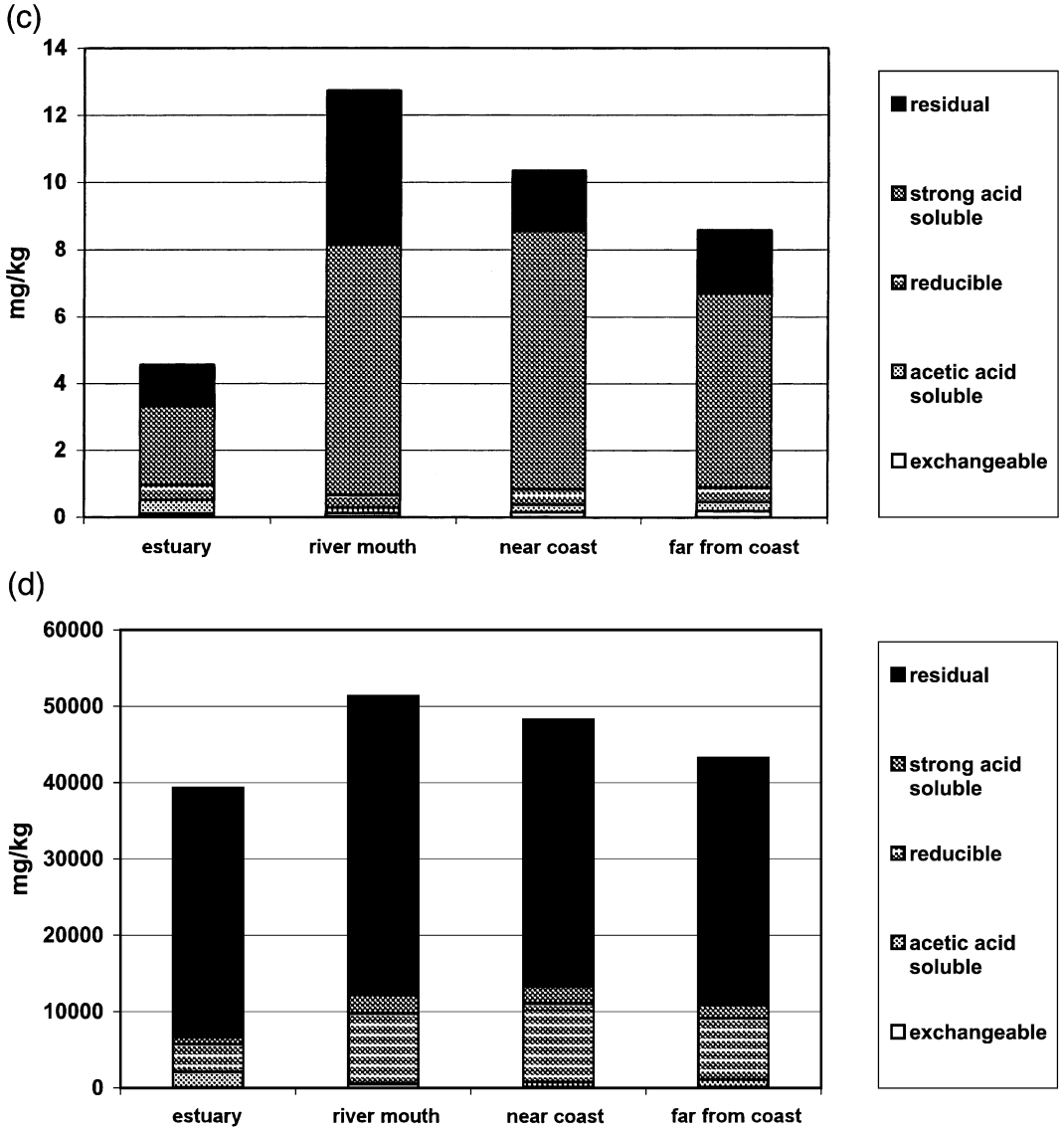
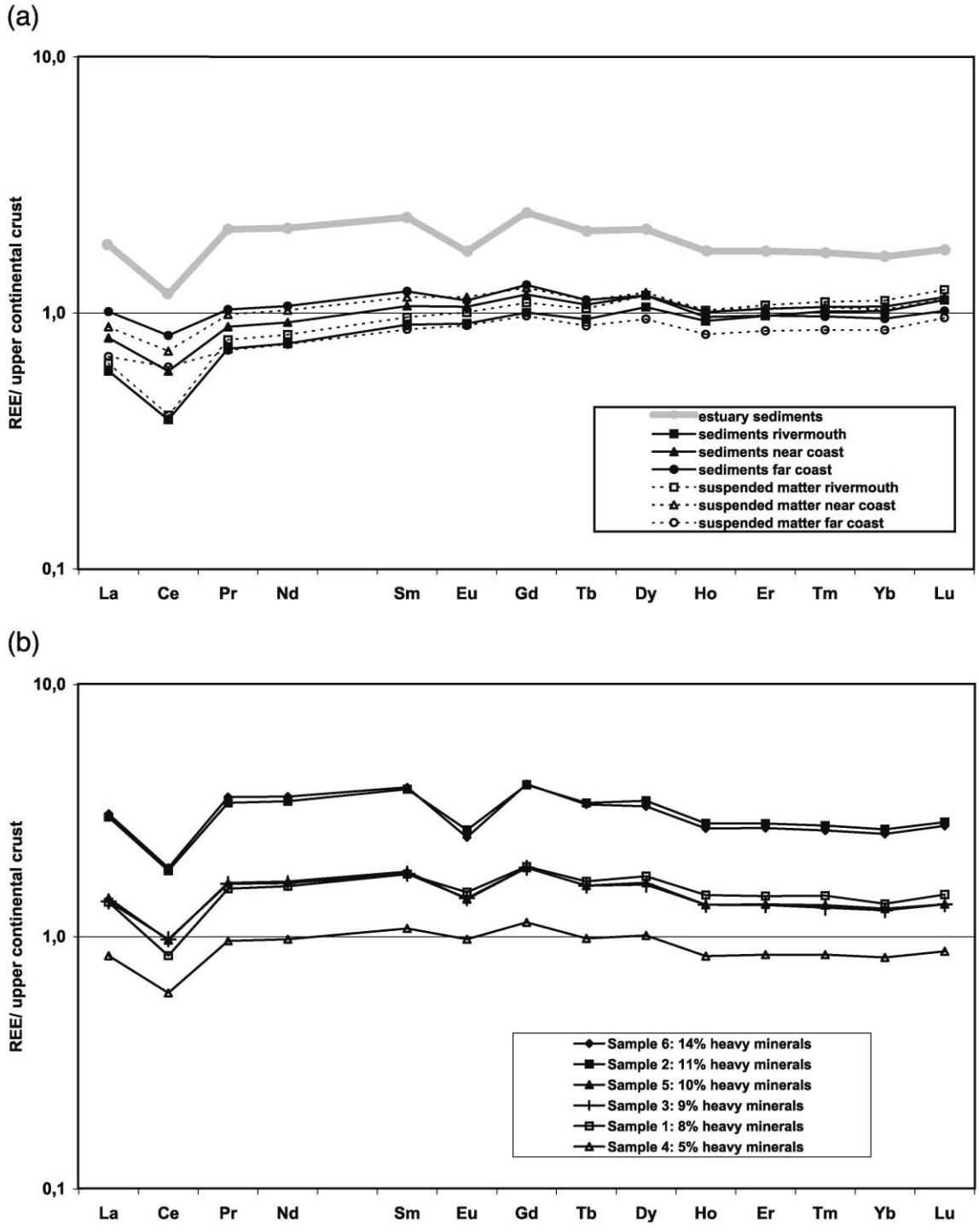


Fig. 5 (*continued*).

between near-coast marine sediments and sediments further away from coast, is substantial. Ca and Sr are present in the estuary sediments as part of the insoluble phase, bound to rock forming minerals because carbonate is missing (see analyse of heavy mineral fractions in the Appendix A). In marine sediments the significant amount of Ca and Sr occur in weakly soluble parts (i.e., in these sediments some carbonate planktonic tests are present) and their quantity increases with distance from the coast because the sedimentation rate of the detrital component decreases. Still, there is a deficiency of bulk Ca and Sr in marine sediments. The amount of Ca and Sr originating in planktonic tests in areas distant from the river mouth is still lower than the amount of Ca and Sr in the rock-forming minerals concentrated in the estuary sediment.


(a)
10,0
1,0
0,1
REE/ upper continental crust
La
Ce
Pr
Nd
Sm
Eu
Gd
Tb
Dy
Ho
Er
Tm
Yb
Lu
estuary sediments
sediments rivermouth
sediments near coast
sediments far coast
suspended matter rivermouth
suspended matter near coast
suspended matter far coast
(b)
10,0
1,0
0,1
REE/ upper continental crust
La
Ce
Pr
Nd
Sm
Eu
Gd
Tb
Dy
Ho
Er
Tm
Yb
Lu
Sample 6: 14% heavy minerals
Sample 2: 11% heavy minerals
Sample 5: 10% heavy minerals
Sample 3: 9% heavy minerals
Sample 1: 8% heavy minerals
Sample 4: 5% heavy minerals

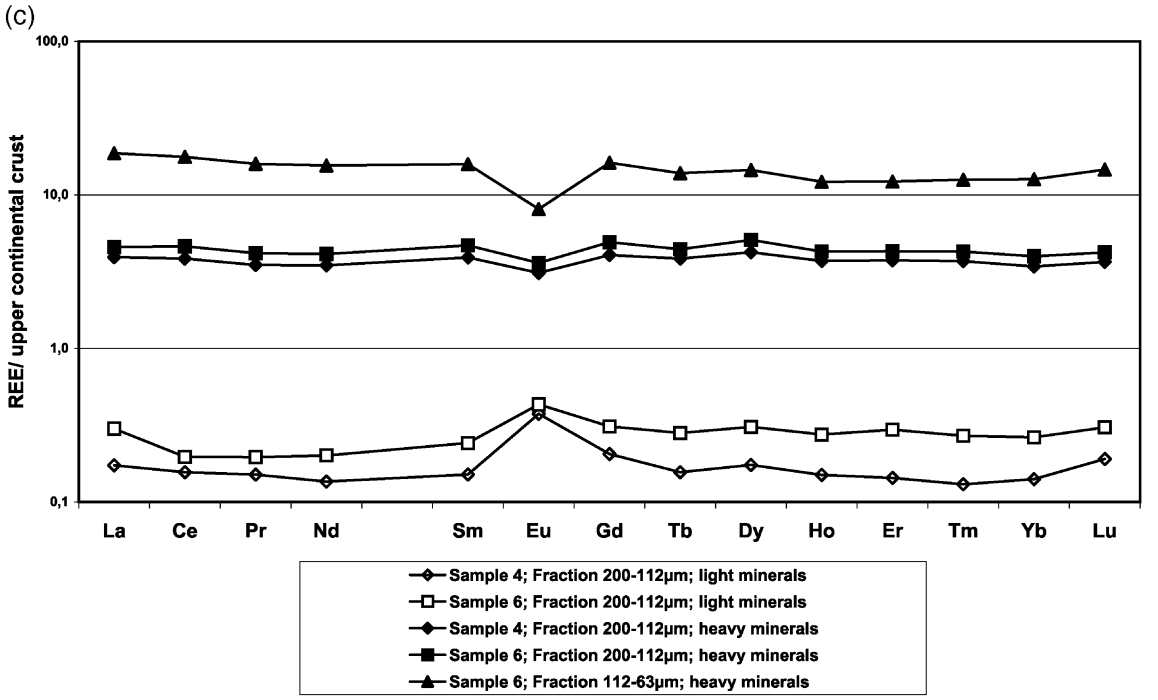


Fig. 6. REE normalised on upper continental crust (Taylor and McLennan, 1981) for different groups of suspended matter and sediments. (a) Average values for sediments and suspended matter near the river mouth, close to the coast, and far from the coast; average values for Meghna estuary sediments. (b) Individual REE plots for six samples from the Meghna estuary sediment. The samples are listed in order of their microscopically determined heavy mineral contents. (c) Individual REE plots for mineral fractions gained from Meghna estuary sediments.

4.5. Rare earth elements

Normalised to average upper crust (Taylor and McLennan, 1981), the rare earth elements of the suspended matter samples are all situated near 1, except for the negative cerium anomaly (Fig. 6a). This is in agreement with the results of Table 2, pointing out that the suspended matter on the shelf off the Brahmaputra–Ganges mouth is well mixed and similar to average upper continental crust. The average negative cerium anomaly is greatest in the suspended matter near the river mouth ($Ce/Ce^* = 0.56$) and decreases with the distance from the coast ($Ce/Ce^* = 0.76$ and 0.88, respectively). This indicates that with distance from the coast, Ce is increasing in comparison to other REE.

Douglas et al. (1999) saw similar negative Ce anomalies in suspended matter from the Murray–Darling River system, and reported that the anomalies were closely related to the Fe abundance in the colloidal fraction (as Fe_2O_3 concentration decreased, the Ce/Ce* became progressively more negative). Douglas et al. (1999) suggested that the Ce anomalies reflected Ce scavenging by Fe oxyhydroxides during the weathering process (e.g., Braun et al., 1990), and thus not necessarily reactions occurring during river transport.

Suspended matter delivered by the Ganges–Brahmaputra river system reach the coast with a distinct negative Ce anomaly. With distance from the coast Ce anomaly is decreasing (i.e., Ce is getting enriched compared to other REE). Parallel to Ce enrichment a Mn-oxyhydroxide phase is progressively accumulating on suspended matter particles. Therefore, the enrichment of Ce in suspended matter off coast can be best explained by considering the arguments of Braun et al. (1990) and Ohta and Kawabe (2001), who suggest that during precipitation of MnO_2, Ce^{3+} is catalytically oxidised on the surface of Mn-hydroxide to insoluble CeO_2. The oxidation of Ce^{3+} to insoluble CeO_2 causes the enrichment of Ce in comparison to other REE, culminating in large positive Ce anomalies. This process of catalytical oxidation was observed only on Mn-

Appendix A
Suspended matter and sediments from the Bay of Bengal

Analytical methods: 1 Quartz determination (Meyer and Klosa, 1997)
2 XRF
3 ICP-MS
4 HG-AAS
5 ICP-OES

Bulk samples

Samples		Quartz %	SiO_2 %	Ti mg/kg	Al mg/kg	Fe mg/kg	Mn mg/kg	Mg mg/kg	Ca mg/kg	Na mg/kg	K mg/kg	P_2O_5 %	(SO_3) %	(Cl) %	(F) %	Ag mg/kg	As mg/kg	Ba mg/kg	Be mg/kg	Bi mg/kg	Cd mg/kg	Co mg/kg	Cr mg/kg
Method		1	2	2	2	2	2	2	2	2	2	2	2	2	2	3	4	3	3	3	3	3	3
3SL	0–5 cm	26	53.62	5102	96,637	53,437	821	19,057	8219	16,617	27,229	0.13	0.09	0.007	0.064	0.07	13.7	446	3.6	0.89	0.057	21.0	136
3SL	5–10 cm	23	53.00	5096	97,537	53,927	774	19,057	8147	16,692	27,478	0.133	0.11	0.012	0.055	0.07	13.9	442	3.8	0.93	0.061	21.1	134
4SL	TOP	28	59.78	4706	81,978	48,261	782	16,343	10,149	17,063	23,493	0.126	0.17	0.01	<0.020	0.06	14.3	415	3.2	0.63	0.050	18.3	115
50KH	0–10 cm	43	70.70	4179	60,809	29,167	511	11,097	19,011	17,211	18,512	0.152	0.09	0.014	<0.020	0.03	4.7	367	2.8	0.24	0.054	11.4	76
49KH	0–10 cm	41	68.55	3813	64,566	37,350	596	13,087	15,580	17,359	20,837	0.124	0.11	0.012	<0.020	0.08	9.6	393	2.3	0.31	0.040	15.2	68
57KH	0–5 cm	21	54.25	5114	94,997	52,878	860	18,515	7719	16,617	26,150	0.124	0.09	0.008	0.061	0.07	14.9	442	3.1	0.86	0.059	20.4	125
57KH	5–10 cm	25	55.14	5108	94,468	52,458	898	17,489	7004	15,727	25,320	0.125	0.08	0.006	<0.020	0.07	14.5	417	2.9	0.81	0.048	19.6	122
56KH	TOP	25	55.23	4964	93,727	51,339	852	18,273	9077	16,766	24,323	0.125	0.10	0.015	0.073	0.10	13.2	449	3.1	0.84	0.066	20.0	117
56KH	0–10 cm	25	54.12	5048	94,785	52,668	867	18,756	8862	16,988	25,320	0.128	0.13	0.011	<0.020	0.11	13.7	454	3.4	0.89	0.12	20.7	116
96KL	0–3 cm	25	53.26	4952	96,373	52,038	821	18,213	10,577	16,914	26,150	0.136	0.06	0.014	<0.020	0.09	14.3	456	3.8	0.97	0.063	19.5	119
96KL	6–9 cm	36	62.75	4532	76,156	39,798	689	14,836	17,295	16,172	21,335	0.132	0.09	0.012	<0.020	0.08	8.0	406	3.0	0.49	0.047	16.1	99
16KH	TOP	36	58.67	4508	80,178	43,225	774	16,283	13,651	21,439	23,742	0.132	0.12	0.025	0.022	0.08	10.7	431	3.3	0.60	0.098	17.1	107
16KH	1–5 cm	26	53.07	5018	96,426	52,388	759	18,635	10,006	18,101	25,320	0.134	0.08	0.013	<0.020	0.11	15.0	456	3.9	0.94	0.068	19.6	126
31KH	0–4 cm	45	67.14	4173	67,265	35,671	596	12,665	16,724	16,469	20,007	0.140	0.05	0.013	0.051	0.05	8.3	383	3.2	0.36	0.048	14.3	86
34KH	TOP	49	68.32	3903	63,613	36,021	620	12,423	16,581	16,988	19,342	0.129	0.05	0.017	0.066	0.06	8.7	371	2.6	0.33	0.041	14.8	77
22KL	0–3 cm	23	54.61	4898	93,250	49,870	790	17,489	11,364	15,579	25,652	0.134	0.09	0.01	<0.020	0.13	12.8	455	3.4	0.96	0.071	18.2	111
22KL	3–6 cm	25	53.76	4844	93,462	50,080	759	17,670	11,006	16,692	26,399	0.129	0.07	0.012	0.067	0.12	11.4	450	2.6	0.97	0.074	16.3	94
30KL	0–3	26	53.21	4946	95,420	51,059	782	18,092	10,077	16,543	27,395	0.132	0.06	0.008	<0.020	0.10	12.3	464	3.0	0.99	0.070	18.5	106
30KL	3–6 cm	26	54.27	4844	95,579	49,730	682	17,670	9577	16,543	27,229	0.130	0.04	0.006	<0.020	0.09	11.6	457	3.2	0.91	0.072	18.6	108

Method	1	2	2	5	5	5	5	5	2	5	2	2	2	2	3	4	3	3	3	3	3	3
2 PZ	19	–	–	95,120	65,320	1580	17,850	6360	–	27,860	–	–	–	–	24.0	17.5	401	3.7	1.02	0.22	21.8	233
13 PZ	15	–	–	86,050	61,140	10,380	17,250	8436	–	24,880	–	–	–	–	5.91	17.9	383	3.6	1.14	0.27	22.1	237
18 PZ	13	–	–	86,890	74,290	7260	17,790	7353	–	25,240	–	–	–	–	2.76	26.1	427	3.5	1.21	0.13	21.1	248
20 PZ	9	–	–	37,400	26,330	437	7845	5170	–	11,300	–	–	–	–	5.85	10.1	195	1.8	0.43	1.44	10.2	201
29 PZ	39	–	–	64,600	45,010	1480	13,210	14,530	–	22,750	–	–	–	–	0.42	11.5	361	3.4	0.59	0.14	16.6	201
48 PZ	45	–	–	63,960	37,370	931	12,330	16,500	–	21,650	–	–	–	–	0.52	7.0	385	2.5	0.39	0.22	14.6	196
53 PZ	22	–	–	78,670	56,110	897	15,670	7640	–	26,050	–	–	–	–	0.29	13.9	421	3.4	0.91	0.14	18.9	146
54 PZ	23	54.53	5318	100,924	54,836	960	19,359	8290	8605	27,644	0.132	0.06	0.007	0.023	0.11	14	488	3.2	0.80	0.074	18	128
55 PZ	21	52.15	5306	101,771	56,375	914	18,997	6932	12,982	27,229	0.147	0.06	0.006	<0.020	0.12	15	459	3.2	1.14	0.11	19	121

Method	1	2	2	2	2	2	2	2	2	2	2	2	2	2	3	4	3	3	3	3	3	3
Estuary1	53	77.73	3633	51,441	23,921	542	6513	20,083	13,650	11,622	0.18	<0.01	0.005	0.024	0.02	2.1	266	1.8	0.14	0.063	7.0	64
Estuary2	47	71.96	6427	56,469	39,309	914	10,373	25,872	12,908	12,203	0.282	<0.01	0.008	<0.02	<0.01	4.1	255	2.0	0.25	0.11	10.9	109
Estuary3	49	75.38	3705	56,681	28,467	596	7659	16,438	14,021	16,188	0.147	<0.01	0.005	<0.02	0.01	4.7	344	2.0	0.19	0.068	8.9	70
Estuary4	49	76.38	2602	57,792	24,690	465	6513	13,079	14,540	18,014	0.092	<0.01	0.006	<0.02	0.02	5.0	382	2.0	0.13	0.069	9.5	54
Estuary5	54	74.67	4077	56,998	30,076	620	8021	17,653	13,724	15,939	0.151	<0.01	0.004	0.061	0.01	4.2	340	2.2	0.16	0.063	10.2	73
Estuary6	51	73.92	6151	53,664	38,399	929	8443	23,370	12,908	12,037	0.234	<0.01	0.015	0.021	<0.01	2.3	262	1.7	0.19	0.11	9.0	110

Separated light and heavy minerals from Meghna estuary samples

Method	SiO_2 %	TiO_2 %	Al_2O_3 %	Fe_2O_3 %	MnO %	MgO %	CaO %	Na_2O %	K_2O %	P_2O_5 %	Ag mg/kg	As mg/kg	Ba mg/kg	Be mg/kg	Bi mg/kg	Cd mg/kg	Co mg/kg	Cr mg/kg
	2	2	2	2	2	2	2	2	2	2	3	4	3	3	3	3	3	3
4 200–112 μm heavy minerals	41.59	2.307	15.66	16.62	0.375	7.28	11.926	1.07	1.018	0.664	0.22	4.6	66	−0.28	0.18	0.28	19	168
6 200–112 μm heavy minerals	40.62	2.639	15.59	16.63	0.4	6.73	13.106	0.99	0.755	0.974	0.49	5.8	48	−0.42	1.29	0.36	20.6	174
6 112–63 μm heavy minerals	31	6.98	13.77	29.21	0.735	4.05	10.29	0.45	0.315	1.124	0.22	7.6	26	−0.3	0.69	3.13	17.6	468
4 200–112 μm light minerals	86.28	0.031	7.98	0.14	0.005	0	1.023	2.13	1.963	0.02	0.09	6.7	338	1.56	0.05	0.19	0.7	5
6 200–112 μm light minerals	87.54	0.055	7.05	0.15	0.005	0.02	1.133	1.83	1.547	0.15	<0.01	8.8	307	1.9	0	0.27	0.8	9

(continued on next page)

Appendix A (*continued*)

Analytical methods: Bulk samples Samples		Cs mg/kg	Cu mg/kg	Ga mg/kg	Ge mg/kg	Hf mg/kg	In mg/kg	Li mg/kg	Mo mg/kg	Nb mg/kg	Ni mg/kg	Pb mg/kg	Rb mg/kg	Sb mg/kg	Sc mg/kg	Sn mg/kg	Sr mg/kg	Ta mg/kg	Th mg/kg	Tl mg/kg	U mg/kg	V mg/kg	W mg/kg	Y mg/kg	Zn mg/kg
Method		3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
3SL	0–5 cm	9.8	47.9	21.3	0.09	2.9	0.081	73.0	0.50	15.2	68.7	28.3	72.9	0.59	14.0	4.6	109	1.41	10.0	0.85	2.26	128	10	18.7	102
3SL	5–10 cm	10.7	50.2	22.0	0.12	3.0	0.083	72.2	0.50	15.3	67.8	28.7	86.7	0.63	14.7	4.8	110	1.40	11.9	0.86	2.18	128	4.0	21.1	104
4SL	TOP	8.4	32.3	18.3	0.10	2.6	0.068	64.5	0.35	15.9	50.2	24.9	96.6	0.50	13.6	4.4	111	1.38	13.1	0.76	2.27	107	14	20.0	86
50KH	0–10 cm	4.9	14.7	13.4	0.09	3.4	0.049	27.1	0.19	13.9	31.5	17.3	57.9	0.39	10.1	3.5	150	1.31	17.7	0.51	2.74	72	8.0	24.7	55
49KH	0–10 cm	8.0	16.1	15.1	0.07	1.8	0.056	31.5	0.22	13.0	35.4	22.3	123	0.33	11.2	4.2	127	1.29	13.3	0.64	2.08	72	2.2	20.8	68
57KH	0–5 cm	9.8	45.2	21.0	0.06	3.2	0.088	67.2	0.54	16.4	68.8	28.2	61.4	0.67	10.7	5.3	77	1.50	8.8	0.82	1.54	116	7.0	18.5	102
57KH	5–10 cm	9.3	40.8	20.2	0.07	3.5	0.085	73.4	0.57	16.4	69.1	27.8	58.0	0.62	9.9	4.9	69	1.48	8.0	0.81	1.44	116	3.2	17.0	95
56KH	TOP	10.6	47.3	21.7	0.06	2.9	0.086	61.7	0.52	16.7	63.4	27.4	69.4	0.69	11.8	5.6	75	1.59	9.6	0.87	1.74	112	12	19.0	116
56KH	0–10 cm	11.0	50.3	22.0	0.05	3.0	0.087	65.1	0.45	16.8	65.7	28.7	73.4	0.71	12.0	5.6	76	1.61	9.5	0.89	1.84	114	7.0	19.0	105
96KL	0–3 cm	11.0	52.4	21.1	0.08	2.8	0.085	60.9	2.53	16.1	58.4	30.7	81.0	0.70	12.0	5.4	83	1.51	11.0	0.86	1.52	117	10	19.2	97
96KL	6–9 cm	7.7	28.8	16.8	0.08	2.6	0.063	41.8	0.81	15.7	42.1	22.3	90.2	0.52	11.3	4.6	116	1.43	14.4	0.67	2.40	92	14	22.4	74
16KH	TOP	8.4	36.5	17.9	0.08	2.8	0.072	46.5	0.48	14.7	49.6	26.5	65.6	0.63	10.3	4.6	102	1.36	11.7	0.73	2.06	99	3.0	19.6	241
16KH	1–5 cm	10.2	52.0	20.6	0.12	2.8	0.076	60.9	0.51	16.0	60.2	27.1	67.9	0.67	11.0	5.2	87	1.46	10.7	0.85	1.67	118	3.4	18.8	107
31KH	0–4 cm	6.8	21.2	15.5	0.08	2.4	0.054	34.5	0.28	13.5	35.9	20.4	78.2	0.41	10.8	3.9	131	1.28	14.5	0.59	2.28	80	8.0	21.8	71
34KH	TOP	6.8	17.9	14.4	0.06	2.0	0.054	32.3	0.23	13.0	35.4	21.4	83.4	0.37	10.1	3.8	134	1.23	12.9	0.59	2.10	74	2.1	19.9	79
22KL	0–3 cm	10.8	46.9	20.2	0.03	2.7	0.086	54.5	0.47	16.1	52.9	26.9	81.7	0.69	11.6	5.5	80	1.52	12.3	0.84	1.69	117	14	20.3	95
22KL	3–6 cm	11.3	41.1	19.7	0.16	3.0	0.085	44.4	0.48	15.0	47.1	27.8	119	0.70	12.4	5.5	78	1.58	15.1	0.86	2.41	90	14	23.9	102
30KL	0–3	11.4	47.0	21.4	0.08	3.1	0.091	53.7	0.51	16.0	55.7	28.7	113	0.76	13.6	5.7	84	1.59	14.0	0.90	2.38	105	12	24.0	94
30KL	3–6 cm	11.3	48.9	21.5	0.10	3.2	0.089	56.1	0.52	16.0	58.3	27.2	102	0.73	13.0	5.7	86	1.56	12.6	0.89	2.21	107	12	23.2	95

Method	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
2 PZ	10.3	312	22.3	0.13	3.0	0.077	73.8	24.6	14.3	121	46.9	76.2	0.86	17.1	37.0	145	1.53	15.9	0.90	2.54	132	3.51	19.7	261
13 PZ	10.0	1880	21.2	0.13	2.8	0.030	63.4	20.8	12.8	136	63.0	75.3	0.93	16.0	257	204	1.20	13.9	0.83	2.54	114	3.44	19.3	523
18 PZ	10.6	988	21.2	0.13	2.9	0.042	69.4	18.2	14.4	139	63.4	89.3	0.98	16.7	202	156	1.32	16.0	0.86	2.77	125	3.58	20.4	437
20 PZ	5.4	1040	9.6	0.07	1.1	0.008	30.0	25.8	5.2	117	38.4	61.4	0.44	8.2	141	158	0.47	6.2	0.41	1.32	51	1.69	9.22	471
29 PZ	8.7	156	18.1	0.07	2.2	0.063	46.0	9.45	13.4	105	28.8	79.4	0.66	12.3	23.0	125	1.27	12.9	0.73	2.09	91	3.01	18.7	161
48 PZ	7.6	170	16.4	0.07	2.4	0.053	35.8	8.04	12.7	108	31.0	106	0.51	11.3	17.4	161	1.25	13.4	0.68	2.23	75	2.70	20.9	209
53 PZ	11.2	98.8	22.9	0.24	3.3	0.087	68.3	5.29	16.4	83.3	33.2	120	0.75	14.6	12.0	102	1.57	14.4	0.92	2.43	116	3.51	23.0	155
54 PZ	10	60	28	0.06	3.1	0.090	63.1	1.04	16	68	29	176	0.71	22	6	98	1.65	23	0.92	1.84	130	3.82	34	120
55 PZ	10	64	23	0.07	3.2	0.095	70.2	0.74	16	64	31	179	0.82	14.0	6.1	96	1.66	22	0.94	2.12	134	3.73	35	112

Method	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
Estuary1	1.9	3.9	10.8	0.06	2.7	0.043	11.8	0.20	11.7	17.0	15.3	49.0	0.23	9.6	2.6	122	1.21	19.8	0.29	2.80	62	17	32.9	38
Estuary2	3.0	7.7	13.6	0.11	6.5	0.067	15.9	0.30	20.3	23.7	16.7	50.4	0.32	15.1	3.9	133	2.27	46.6	0.32	6.55	96	6	61.3	52
Estuary3	3.8	6.9	12.9	0.05	3.3	0.047	17.7	0.15	12.4	21.5	19.3	67.0	0.27	9.5	3.0	117	1.49	22.6	0.41	3.31	67	9	29.3	46
Estuary4	4.9	6.6	12.8	0.03	2.2	0.038	18.7	0.22	9.5	25.5	20.3	77.2	0.25	7.4	2.7	123	1.03	14.6	0.49	2.29	55	2	18.8	71
Estuary5	4.8	6.8	13.1	0.05	3.3	0.047	19.4	0.21	13.6	23.9	19.1	72.3	0.29	8.9	3.4	112	1.53	22.2	0.46	3.36	69	2.2	29.4	52
Estuary6	2.4	4.9	12.9	0.14	6.2	0.058	12.8	0.30	20.3	18.4	16.2	54.1	0.33	13.4	3.7	128	2.41	55.5	0.31	7.95	89	4	59.4	47

Separated light and heavy minerals from Meghna estuary samples

Method	Cs mg/kg 3	Cu mg/kg 3	Ga mg/kg 3	Ge mg/kg 3	Hf mg/kg 3	In mg/kg 3	Mo mg/kg 3	Nb mg/kg 3	Ni mg/kg 3	Pb mg/kg 3	Rb mg/kg 3	Sb mg/kg 3	Sc mg/kg 3	Sn mg/kg 3	Sr mg/kg 3	Ta mg/kg 3	Th mg/kg 3	Tl mg/kg 3	U mg/kg 3	V mg/kg 3	W mg/kg 3	Y mg/kg 3	Zn mg/kg 3
4 200–112 μm heavy mineral	1.86	111	15.7	1.9	4.9	0.13	2.1	23.1	41.8	9.73	38.6	0.41	68.9	6.3	165	2.09	36.5	<0.005	4.68	229	1.6	91.7	138
6 200–112 μm heavy mineral	1.33	15.4	14.9	1.96	5.7	0.24	2.37	28.9	36.9	9.8	23.7	0.48	68.8	7	190	2.77	45.1	0.29	5.77	225	1.9	104	80
6 112–63 μm heavy mineral	0.36	12.9	14.1	2.78	157	0.19	2.65	71.1	24.2	12.7	6.5	0.68	61.1	18.2	137	7.85	275	0.18	45.5	159	8	279	89
4 200–112 μm light mineral	1.18	14.5	6.4	0.98	1.3	0.007	1.67	0.8	7.1	19.2	68.7	0.17	69.4	1.2	125	0.13	2.4	0.057	0.61	<10	0.3	3.2	8
6 200–112 μm light mineral	1.04	12.1	6	1.19	2.4	<0.005	1.73	1.3	5.6	15.6	60.1	0.19	87.8	0.7	111	0.15	2.5	0.062	0.69	<10	0.4	7.2	11

(continued on next page)

Appendix A (*continued*)

Analytical methods: Bulk samples Samples / Method		Zr mg/kg 3	La mg/kg 3	Ce mg/kg 3	Pr mg/kg 3	Nd mg/kg 3	Sm mg/kg 3	Eu mg/kg 3	Gd mg/kg 3	Tb mg/kg 3	Dy mg/kg 3	Ho mg/kg 3	Er mg/kg 3	Tm mg/kg 3	Yb mg/kg 3	Lu mg/kg 3
3SL	0 cm	133	20.4	45.7	5.05	19.4	3.99	0.79	3.76	0.60	3.71	0.74	2.25	0.33	2.28	0.36
3SL	5–10 cm	132	24.8	46.4	6.06	23.3	4.61	0.93	4.36	0.68	4.06	0.81	2.43	0.35	2.37	0.39
4SL	TOP	182	28.5	54.9	6.59	25.3	5.04	0.98	4.82	0.72	4.23	0.82	2.41	0.34	2.27	0.37
50 KH	0–10 cm	359	43.2	79.8	10.1	37.9	7.25	1.20	6.38	0.90	4.95	0.91	2.61	0.36	2.35	0.36
49 KH	0–10 cm	211	28.3	43.4	7.48	28.0	5.47	1.03	4.96	0.73	4.04	0.76	2.16	0.31	1.97	0.31
57 KH	0–5 cm	142	17.3	22.2	4.89	18.7	3.91	0.80	3.75	0.60	3.67	0.76	2.26	0.33	2.33	0.37
57 KH	5–10 cm	137	15.7	19.3	4.45	17.1	3.52	0.73	3.42	0.56	3.41	0.70	2.14	0.33	2.17	0.35
56 KH	TOP	134	17.8	24.4	5.17	19.9	4.07	0.80	3.86	0.61	3.64	0.74	2.26	0.33	2.24	0.36
56 KH	0–10 cm	133	17.8	24.6	5.12	19.6	4.03	0.80	3.77	0.60	3.74	0.75	2.22	0.34	2.23	0.36
96 KL	0–3 cm	134	21.4	29.7	5.49	21.1	4.29	0.82	3.96	0.61	3.71	0.73	2.15	0.32	2.16	0.35
96 KL	6–9 cm	205	32.2	53.2	7.76	29.2	5.75	1.06	5.20	0.78	4.38	0.84	2.43	0.35	2.26	0.34
16 KH	TOP	169	23.5	43.1	5.81	22.2	4.61	0.87	4.17	0.64	3.69	0.71	2.12	0.31	2.03	0.32
16 KH	1–5 cm	131	21.0	27.1	5.25	19.9	4.05	0.78	3.80	0.59	3.60	0.72	2.13	0.32	2.10	0.33
31 KH	0–4 cm	267	33.1	59.4	8.08	30.4	5.96	1.07	5.27	0.77	4.25	0.78	2.25	0.32	2.09	0.32
34 KH	TOP	222	31.1	53.8	7.40	28.0	5.46	1.00	4.84	0.70	3.99	0.74	2.12	0.29	1.89	0.30
22 KL	0–3 cm	137	23.4	32.6	6.14	23.5	4.62	0.90	4.36	0.67	3.95	0.78	2.33	0.34	2.26	0.36
22 KL	3–6 cm	136	26.3	36.9	7.68	29.2	5.85	1.11	5.41	0.82	4.86	0.95	2.76	0.40	2.65	0.41
30 KL	0–3	132	25.7	36.7	7.15	27.0	5.43	1.06	5.12	0.78	4.72	0.93	2.73	0.40	2.69	0.42
30 KL	3–6 cm	129	23.8	34.2	6.55	24.7	4.99	0.96	4.75	0.73	4.42	0.87	2.59	0.38	2.56	0.40
Method		3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
2 PZ		103	25.8	55.6	6.65	25.8	5.15	1.04	4.71	0.72	4.30	0.86	2.55	0.38	2.45	0.40
13 PZ		97	23.0	46.5	5.81	22.5	4.48	0.92	4.29	0.66	3.93	0.78	2.33	0.34	2.20	0.36
18 PZ		102	25.9	49.0	6.54	25.4	5.07	1.02	4.88	0.76	4.38	0.87	2.59	0.37	2.48	0.40
20 PZ		40	12.0	22.6	2.90	11.0	2.10	0.42	1.94	0.29	1.65	0.33	0.96	0.14	0.98	0.16
29 PZ		78	25.5	43.0	6.41	24.2	4.68	0.89	4.28	0.65	3.69	0.72	2.07	0.30	1.94	0.30

48 PZ	86	28.6	44.2	7.59	28.6	5.45	1.04	4.97	0.72	4.14	0.78	2.26	0.31	2.04	0.32
53 PZ	120	26.1	38.7	7.40	28.0	5.49	1.09	5.14	0.78	4.71	0.92	2.74	0.40	2.64	0.43
54 PZ	122	17.1	23.5	5.00	19.1	3.91	0.79	3.79	0.61	3.77	0.77	2.31	0.34	2.33	0.37
55 PZ	125	21.2	27.5	6.16	23.7	4.76	0.98	4.55	0.72	4.44	0.87	2.63	0.39	2.59	0.42
Method	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
Estuary 1	287	41.1	53.8	11.0	41.2	7.96	1.32	7.21	1.06	6.08	1.17	3.33	0.48	2.97	0.47
Estuary 2	890	89.3	117	24.1	89.5	17.3	2.33	15.2	2.17	12.1	2.25	6.46	0.91	5.86	0.91
Estuary 3	429	41.4	62.5	11.5	42.4	7.99	1.25	7.13	1.02	5.63	1.07	3.07	0.43	2.80	0.43
Estuary 4	238	25.2	38.3	6.83	25.4	4.85	0.86	4.33	0.63	3.54	0.67	1.95	0.28	1.82	0.28
Estuary 5	442	42.6	62.0	11.6	43.0	8.15	1.24	7.13	1.02	5.73	1.07	3.09	0.44	2.84	0.43
Estuary 6	1058	92.0	120	25.4	93.4	17.6	2.18	15.3	2.14	11.5	2.15	6.20	0.87	5.62	0.88

Separated light -and heavy minerals from Meghna estuary samples

Method	Zr mg/kg 3	La mg/kg 3	Ce mg/kg 3	Pr mg/kg 3	Nd mg/kg 3	Sm mg/kg 3	Eu mg/kg 3	Gd mg/kg 3	Tb mg/kg 3	Dy mg/kg 3	Ho mg/kg 3	Er mg/kg 3	Tm mg/kg 3	Yb mg/kg 3	Lu mg/kg 3
4 200–112 μm heavy mineral	108	118	246	24.8	90.3	17.6	2.73	15.4	2.46	14.8	2.97	8.62	1.22	7.52	1.17
6 200–122 μm heavy mineral	120	137	296	29.6	107	21.1	3.17	18.7	2.83	17.8	3.42	9.87	1.41	8.75	1.35
6 112–63 μm heavy mineral	3470	560	1130	113	404	71.3	7.11	61.5	8.86	50.8	9.74	28.1	4.14	27.8	4.67
4 200–122 μm light mineral	26	5.2	10	1.07	3.53	0.68	0.33	0.78	0.1	0.61	0.12	0.33	0.043	0.31	0.061
6 200–122 μm light mineral	51	9	12.6	1.39	5.22	1.09	0.38	1.18	0.18	1.08	0.22	0.68	0.089	0.58	0.098

oxyhydroxide, it was not observed on surface of Fe-oxyhydroxide in marine environment (De Carlo and Wen, 1998).

REE spectra of shelf sediments are very similar to those of suspended matter. The REE concentrations of the estuary sediments are significantly higher and have well-developed negative Eu anomalies. The carriers of REE are commonly heavy minerals and/or accessory minerals. This can be seen in Fig. 6b in which the sediments from the estuary are clustered due to their estimated heavy mineral content. The group with highest heavy mineral content consequently shows the highest REE concentrations (i.e., the absolute amount of heavy minerals parallels the enrichment of REE). Yttrium and REE are enriched due to monazite, sphene, allanite, apatite or garnet (Cullers et al., 1988), whereas zircon was found to be carrier of Zr and Hf enrichment.

In Fig. 6c, the REE patterns are shown from selected fractions of the two, previously mentioned, extreme samples from the Meghna estuary (i.e., samples 4 and 6).The heavy minerals are, as expected, carriers of REE, with enrichments of one to two orders of magnitude. They show significant negative Eu anomalies. The light minerals contain low contents of REE but with positive Eu anomalies characteristic for plagioclase (Drake and Weill, 1975). In the mixture (Fig. 6b), only negative Eu-anomalies from REE-rich heavy minerals are conserved.

5. Conclusions

The suspended matter load of the Ganges–Brahmaputra drainage area reaches the Meghna Estuary where separation takes place. The fine sand fraction, enriched in quartz, feldspars, muscovite, and heavy minerals, settles down in estuary channels. The predominant part of the fine fraction, a suspended matter enriched in clay minerals, is transported to the shelf.

The residual fine sand settled in the estuary is a product of mechanical erosion of metamorphic rocks and intermediate to acid magmatic rocks. The transport to the estuary was relatively gentle, as concluded from poorly rounded grains and scarce physical weathering marks on the sediment grains.

The fraction enriched in clay minerals forms the suspended matter of the shelf area; its composition in the vicinity of the river mouth corresponds to the world average of river suspended matter load.

Changes in the suspended matter composition that occur during sediment transport are partly caused by grain size separation, because residual heavy minerals settle with distance from the coast, leaving the suspended matter depleted in REE, Zr, Hf, Nb, Ta, and W. The other reason for changes in composition is the formation of manganese oxide coatings on suspended grains, thus enriching the coast-distant suspended matter with Mn and adsorbed metals as transition metals, Pb and Cd.

The described suspended matter is representative for the low sediment transport season. During times of high sediment transport, the river is transporting a 10-fold load of suspended matter, which is expected to be coarser. This is evident from the composition of the shelf sediment.

The shelf sediment was deposited in oxygen-free bottom-near seawater, and this results in dissolution and recycling of manganese oxide stains with other elements incorporated. It does not remobilise iron oxides with incorporated arsenic.

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