



Geochemical characterization of mangrove sediments of the Zuari estuarine system, West coast of India



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ABSTRACT

The grain size, clay mineralogy and geochemistry were studied in the sediment cores collected from the mangrove environments of the Zuari estuary to understand sources and factors affecting accumulation, mobility, bioavailability and toxicity of metals. Finer sediments, organic matter and metals were higher in the middle estuary and canal sediments while coarser sediments with fewer metal concentrations were seen in the lower estuary. Kaolinite, smectite, illite and traces of chlorite constituted the clay mineral assemblage and had a minor influence on metal distributions. In the study area, the hydrodynamic conditions changed from lower estuary towards the upstream regions owing to mixing of riverine and sea water that led to finer sediment deposition in the middle estuary. The variations in metal abundance were attributed to a difference in hydrodynamic conditions regulated by the tide, freshwater flow and geomorphology of the Zuari estuary. The results revealed that the estuary received material from natural weathering of rocks as well as from anthropogenic sources such as mining and industrial/domestic discharges. Enrichment factor and Geo-accumulation index showed that Fe, Mn and Cr were enriched in the mangrove sediments whereas fractionation of metals revealed that concentrations of bioavailable Mn pose a considerable risk to biota. Increased accumulation of Fe and Mn in the upper middle estuary and canal sediments, trap trace metals that may considerably affect sediment quality and dredging of these sediments can cause re-suspension and mobilize metals from loosely bound sedimentary forms to the water column.

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1. Introduction

The coastal zone is characterized by a variety of landforms out of which estuaries have received considerable attention due to large land–sea interaction mechanisms (Bianchi, 2006; Buddemeier et al., 2008). Estuaries receive reciprocating water flow, comprised of terrestrial and marine particulates (Yuan et al., 2014) which on mixing of seawater and fresh-water, facilitate deposition of a large amount of the suspended load to the sediments. Many metals associated with particulates are either adsorbed or co-precipitated with carbonates, oxy-hydroxides, sulfides and clay minerals (Spencer and MacLeod, 2002). The mangrove sub-environment is a distinct intertidal habitat, where fine sediments with high organic content accumulate in areas protected from high-energy wave action. Mangrove sediments act as long-term sinks for

various metals as they constitute fine grain-size sediments that favor sorption of metals and the metals get trapped in the dense root network of the mangrove vegetation (Huerta-Diaz and Morse, 1992). Further, mangrove sediments being anaerobic and reduced with high sulfide and organic matter content have a high capacity to retain heavy metals received from natural and anthropogenic sources (Fernandes et al., 2012). The metals are also present in various forms such as dissolved species, free ions or forming organic complexes with humic and fulvic acids within estuaries. The metals, however, cannot be degraded; they either may accumulate locally (Marchand et al., 2006) or are re-mobilized within an estuary. It is therefore essential to identify and quantify the forms in which a metal is present in sediments to gain a precise understanding of transportation, deposition and release processes under changing environmental conditions (Li et al., 2000). Several studies have been carried out on sediment distribution, transport and geochemistry within the estuarine channel and mudflat sediments of the Zuari estuary (Dessai et al., 2009; Dessai and Nayak, 2009; Rao et al., 2011, 2015; Kessarkar et al., 2013; Shynu et al., 2013;

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Singh et al., 2013). In the present study, metal distribution patterns in sediment cores collected from mangroves of the Zuari Estuary were investigated to identify sources and factors that have facilitated metal accumulation in the sediments. An attempt was also made to understand mobility, bioavailability and potential toxicity of metals to the estuarine environment.

2. Study area

The Zuari estuary is classified as a tide-dominated, coastal plain estuary and geomorphologically as drowned river valley estuary (Murty et al., 1976). The Zuari River is about 70 km long with catchment area of around 550 km² that drains through rocks of the Goa Group belonging to the Dharwar Super Group of the Archaean Proterozoic age (Gokul et al., 1985). The Dharwarian rocks are represented by metamorphosed basic and acidic volcanic rocks (Gokulam, 1972). The rocks at the base are overlain by greywacke suite of rocks, followed by pyroclasts and tuffs with the associated chaemogenic precipitation of lime, manganese and iron, and then again by greywackes. The hinterland rocks of the Zuari estuary are subjected to intense chemical weathering under humid tropical climate conditions. The catchment areas of both the Zuari and adjacent Mandovi rivers are known for open cast iron and ferromanganese mines and are used to transport iron ores to the Mormugao harbor located at the mouth of the Zuari estuary. Iron ore mined in the hinterland of Zuari and Mandovi rivers is stored on riverbanks in the upstream regions for loading onto the barges. During the monsoons, the stacked ore gets washed to the estuary carrying them in suspension (Singh et al., 2013). Ore transport and handling at the Mormugao port can also contribute to the suspended load in the estuary (Panchang et al., 2005). Thus, the estuarine waters contain a high concentration of suspended matter that mainly comprises of iron and manganese due to mining activities. Several jetties, barge building and repairing industries, constructions and domestic waste discharges along the estuary also release a large amount of organic and inorganic contaminants to the estuary. The Cumbharjua canal links the Zuari estuary to the adjacent Mandovi estuary and adds much of the mining material to the Zuari during the monsoons and ebb tide (Dessai and Nayak, 2009).

3. Methodology

Four sediment cores were collected, one each from the upper middle-S1 (Borim), lower middle-S3 (Cortalim), lower estuarine-S4 (Chicalim) regions of the Zuari estuary and adjoining Cumbharjua canal-S2 representing mangrove environment of lengths 34, 40, 24 and 54 cm respectively (Fig. 1). The cores were collected at low tide using a PVC tube of 6.5 cm diameter and sectioned at 2 cm intervals. The subsamples were stored in numbered polythene bags and were transferred to the lab in ice boxes. The subsamples were later oven dried at 60 °C and part of the sample was used for sediment grain size analysis (Folk, 1974). Clay minerals in sediments were analyzed by the procedure given by Rao and Rao (1995). Another part of the sample was ground and used to determine Total Organic Carbon (TOC) by the method proposed by Walkley and Black (1934) and bulk metal analysis after digesting in a mixture of HF–HClO₄–HNO₃ (Jarvis and Jarvis, 1985). Separated clay fractions from grain size analysis, of selected samples were also digested following the same procedure. The digested extracts were analyzed for metals Al, Fe, Mn, Cr, Co, Ni, Cu and Zn on Varian AA240FS flame atomic absorption spectrometry (AAS) with an air/acetylene flame for all metals except Al for which nitrous oxide/acetylene flame was employed at specific wavelengths. A modified sequential extraction procedure proposed by Tessier et al. (1979) was applied to selected

subsamples to determine metal speciation. The procedure involves extraction of five geochemical fractions in sequence viz. exchangeable phase, carbonate phase, Fe–Mn oxide (reducible) phase, organic matter/sulfide (oxidizable) bound phase, and the residual phase. The fractions were analyzed on AAS. The accuracy of the analytical method was analyzed by the standard reference 2702 obtained from National Institute of Standards and Technology (NIST). The average recoveries and \pm standard deviations found for metals Al, Fe, Mn, Cr, Co, Ni, Cu and Zn, were 96 ± 12 , 89 ± 12 , 91 ± 15 , ± 14 , 74 ± 12 , 86 ± 16 , 82 ± 15 , and 84 ± 16 respectively. Precision was monitored by running triplicates after every 10 samples and the relative standard deviation was generally <6% for major and trace elements. Analytical grade reagents and standards were used in all the analysis. To establish geochemical associations and ascertain the factors and processes that influence the behavior of metals in the sediments, two-tailed Pearson's correlation test was computed using the software package SPSS 17. Enrichment factor was computed to evaluate metal enrichment in sediments using the equation " $EF = (M_{sed}/R_{sed})/(M_{bk}/R_{bk})$ " where (M_{sed}/R_{sed}) is the ratio of heavy metal concentration (M_{sed}) to that of the reference element (R_{sed}) in sediment and (M_{bk}/R_{bk}) is the same ratio in the natural background. Aluminum was used as the reference element to counter-balance lithogenic influences of granulometric and mineralogical variations of sediments (Qi et al., 2010). Geo-accumulation index-Igeo (Muller, 1979) was computed to assess the level of metal contamination in sediments using the equation " $I_{geo} = \text{Log}_2 (C_n/1.5B_n)$ " where C_n is the concentration of the metal in the sample and B_n is the background metal concentration in average shale (Turekian and Wedepohl, 1961). Concentration of metals in the bioavailable fractions and bulk sediments was compared with the sediment quality values (SQV) following screening quick reference table (SQUIRT) developed by NOAA (Buchman, 1999) and Risk Assessment Code-RAC (Perin et al., 1985) to explain the toxicity level of the metals.

4. Results and discussion

4.1. Sediment grain size

The range and average values of sand, silt and clay of the four cores are presented in Table 1. Sediments exhibited wide variations in grain size at different stations of the estuary. The percentage of the coarser sediment fraction decreased towards the upper middle estuary with a corresponding increase in finer sediment. High sand content in the lower estuary sediments was due to the higher hydrodynamic conditions prevailing owing to high energy waves that prevented the accumulation of finer sediments. High silt was observed in the canal sediments whereas clay was notably higher in the lower middle estuarine sediments. Overall, clay and silt dominated the sediments collected from the upper middle, lower middle estuary and Cumbharjua canal. Thus, the high-energy conditions in the lower estuary caused the finer sediments to be carried in suspension and deposited at places where hydrodynamic conditions were quiet.

The vertical profiles of sand, silt and clay of cores S1, S2, S3 and S4 are presented in Fig. 2. Sand distribution in core S1 showed percentages above average up to 26 cm followed by constant percentages below average towards the surface (Fig. 2a). Silt and clay distribution patterns compensated each other with fluctuations. Core S2 showed a decrease in sand percentage from the core base to 22 cm followed by an increase towards the surface (Fig. 2b). Clay percentage decreased towards the surface and was compensated by an increase in silt in this core. In core S3, the distribution of sand, silt and clay percentages showed little variation with depth (Fig. 2c). However, silt and clay compensated each other. In core S4

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