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# Understanding distribution and abundance of metals with space and time in estuarine mudflat sedimentary environment

Anant Pande · G. N. Nayak

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**Abstract** Spatial and depth-wise distribution of sediment components, organic carbon and selected metals (Fe, Mn, Al, Ni, Cr, Co, Zn and Pb) is studied across upper and middle tidal flats from lower and middle estuarine regions of Kundalika Estuary, central west coast of India. Silt and clay form the major components in lower and middle estuary, respectively. Sand, silt, clay and organic carbon showed band-type distribution along the estuary. The sediment deposition over the years took place in varying hydrodynamic conditions in lower and middle estuarine regions. Upper flats of the lower estuary represent mud while middle flats of the lower estuary facilitated the deposition of sandy mud. Correlation results indicated the importance of clay and organic carbon in removal and trapping of metals at lower and middle estuary, respectively. Factor analysis indicated that the distribution of metals is largely controlled by Fe–Mn oxyhydroxides and organic carbon. The middle flats of the lower estuary showed an anthropogenic source for Ni, Cr and Co while middle flats of the middle estuary showed a mainly lithogenic source.

**Keywords** Mudflats · Kundalika Estuary · Sediment components · Metals distribution

## Introduction

The estuarine mudflats are large un-vegetated areas that are exposed during low tide and submerged during high

tide. Mudflats primarily consist of fine sediment deposits (<63  $\mu\text{m}$ ) originating from two main sources, namely land and sea (Lesueur et al. 2003). The intertidal mudflats can be divided into three distinct zones: the lower tidal flats lie between mean low water neap and mean low water spring tide levels and are often subjected to strong tidal currents; the middle flats, located between mean low water and mean high water neaps; the upper tidal flats lie between mean high water neap and mean high water springs (Dyer et al. 2000). The upper flats are characterized by fine grained sediments (coarse clays), middle flats by fine silts and the lower flats by sandy mud (Shi and Chen 1996). Amos (1995) classified tidal flats into supratidal, intertidal and subtidal zones. The important conditions for the formation of tidal flats are the dominance of tides and tidal currents and the accumulation of fine sediments. Tidal flats generally form in a mesotidal and macrotidal environment since tidal currents tend to be relatively large compared with those of a microtidal coast (Gao 2009b). The sediment deposition across tidal mudflats is controlled by various estuarine processes such as settling and scour lag, tidal pumping, density-driven circulation and flocculation (Pejrup and Andersen 2000). The estuarine mudflats are key habitats for a number of macrofaunal species and are consequently important areas for both avifaunal population (as roosting/feeding areas) and also act as important food sources or nursery grounds for fish communities (Boyes and Allen 2007). At tropical latitude, mangrove forests grow at the upper tidal flats (Harbison 1986; Wells and Coleman 1981) and are the least inundated parts of estuarine mudflats.

Estuarine mudflat sediments are an important sink for a wide range of contaminants, heavy metals in particular, showing a high affinity for fine grained sediments (Cundy and Croudace 1995; Spencer et al. 2003). Once they

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become part of suspended particulate matter, they are transported through the water column and finally incorporated into the intertidal mudflats. Due to their potential role as contaminant storage areas, mudflat environments may continue to release heavy metals into estuarine waters even after effluent discharge has ceased. A variety of physical, chemical and biological processes may facilitate mixing, remobilise and ultimately rework the metals into the water column through the processes of erosion, dredging, early diagenesis and bioturbation (Deng et al. 2010; Filho et al. 2011). The present study is carried out to understand sources and factors that control distribution of metals.

### Study area

The Kundalika is one of the major rivers along the central west coast of India that originates in the Sahyadris at an altitude of 820 m near Hirdewadi and meets the Arabian Sea near Revdanda in Raigad District of Maharashtra. The river is fed in the north by the Amba River catchment and in the south by the Savitri River basin. The total length of the river is 67.9 km. The basin perimeter of Kundalika River is approximately 166.96 km and basin area is 489.44 km<sup>2</sup> (Shindikar 2006). Geologically, the collision of Indian and Eurasian plate during early Tertiary initiated the tectonic upliftment and resulted in the evolution of the Western Ghats. The source region of Kundalika is characterized by basalt and intrusives. The catchment area is moderately dissected plateau of Deccan trap. The soil is of an alluvial type rich in phosphorous, manganese and copper (Shindikar 2006). The region experiences tropical warm, humid or maritime climate throughout the year and temperature range is between 25 and 35 °C. Kundalika is a tide-dominated estuary. The tides here are semi diurnal in nature (Inamdar 2010). The spring tidal range decreases considerably from lower to middle estuary (Dineshkumar et al. 2001). The highest tide near Revdanda is 4.12 m (Chauhan et al. 2004). However, neap tides show only a small decrease. The limit of tidal flooding is 30 km approximately. The middle region of Kundalika Estuary is an ideal sink for suspended sediment matter. The region receives an average annual rainfall of 3,750 mm. Mangrove species from genera like *Acanthus*, *Avicennia*, *Rhizophora* form the dominant vegetation in upper tidal flats of Kundalika River. Maharashtra industrial development corporation (MIDC) has set up several chemical industries at Dhatav-Roha located in the middle region of Kundalika Estuary. These industries discharge their effluents directly into the river without any treatment (Maharashtra Pollution Control Board 2004–2005).

### Materials and methods

#### Field studies and sampling

Sediment cores were collected from four selected locations in estuarine mudflats representing distinctive geomorphology and sedimentological settings within Kundalika River. Among the four cores, two cores were collected from lower estuary representing upper (S-47) and middle flats (S-62) and the remaining two cores (S-63 and S-45) were collected from middle flats along the middle estuary (Fig. 1). The two cores (S-63 and S-45) represent the middle estuary, one (S-63) from lower region and the other from upper region (S-45). Cores were recovered during low tide using a hand-driven PVC coring tube of 63 mm inner diameter. The core length varied between maximum of 82 cm in core S-47 and minimum of 66 cm in cores S-62, S-63 and S-45. Each core was sub-sampled into 2 cm intervals.

#### Laboratory analysis

The sediment samples were dried in Petri dish at 60 °C for 48 h. Each of the sub-samples was analyzed for sediment components (sand, silt, clay) using Pipette method detailed by Folk (1968) and organic carbon was estimated using the method detailed by Gaudette et al. (1974) in which exothermic heating and oxidation with K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> and concentrated H<sub>2</sub>SO<sub>4</sub> are followed by titration of excess dichromate with 0.5 N Fe(NH<sub>4</sub>)<sub>2</sub>(SO<sub>4</sub>)<sub>2</sub>·6H<sub>2</sub>O. Three cores (S-47, S-62 and S-45) were further analyzed for concentration of major (Fe, Mn and Al) and trace (Ni, Cr, Co, Zn and Pb) elements. For this samples were digested using hydrofluoric-perchloric-nitric acid mixtures in Teflon beakers. Complete digestion was ensured by repeating the digestion steps until

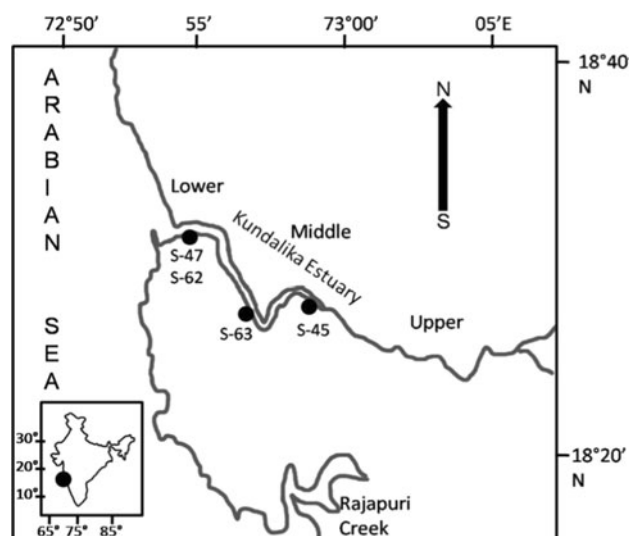


Fig. 1 Study area with core location

clear solutions were obtained (Jarvis and Jarvis 1985). The concentrations of major and trace elements were determined using Varian AA 240 FS flame Atomic Absorption Spectrometry (AAS) with an air/acetylene flame for all of the above elements except for Al for which nitrous oxide/acetylene flame was employed at specific wavelength. The instrument was calibrated by running blank and standard solutions prior to each element analysis. Recalibration check was performed at regular intervals. All chemicals used in the study were of analytical grade. Together with the samples, certified reference standard from the Canadian National Bureau of Standards (BCSS-1) was digested and run, to test the analytical and instrument accuracy of the method. The recoveries were between 86 and 91 % for Fe, Ni and Al; 87–92 % for Mn and Co; 80–85 % for Pb and Zn; 90–95 % for Cr, with a precision of +6 %.

To distinguish between anthropogenic and natural input of trace metals, we use enrichment factor (EF) as an index, which is observed metal to aluminum ratio in the sample divided by the background metal/aluminum ratio (Seshan et al. 2012; Xia et al. 2012). It is expressed mathematically by

$$EF = [(M/Al)_{\text{sediment}} / (Me/Al)_{\text{shale}}]$$

where  $(M/Al)_{\text{sediment}}$  is the metal to Al ratio in the samples of interest and  $(Me/Al)_{\text{shale}}$  is the metal to Al ratio in average shale (Turekian and Wedepohl 1961).

### Statistical analysis

The Pearson correlation coefficient (PC) and the principal component analysis (PCA) were applied to the data using the software Statistica 6.0. Principal component analysis was carried out by applying factor analysis to 12 variables.

## Results and discussion

### Sand–silt–clay

The range and average percentage of sand, silt and clay for each of the cores are given in Table 1 and their distribution profile is shown in Figs. 2 and 3a, b.

The core S-47 collected from upper flats of lower estuary can be divided into three sections depending upon the distribution pattern of sediment components, namely bottom (82–50 cm), middle (50–28 cm) and top (28–0 cm). In the bottom section (Fig. 2a), silt percentage is higher while clay percentage is lower between 74 and 54 cm than the average. In the middle section, silt percentage decreases and clay percentage increases, with some variations. The sand percentage remains constant in both the sections but shows lower values in the middle section. In the top section, silt percentage increases while clay percentage decreases. Sand percentage increases between 28 and 18 cm and then decreases further up to 6 cm before showing an increasing trend toward the surface. When the average values of the different components are compared with the distribution profiles of respective sections, they also support the division into three sections described above.

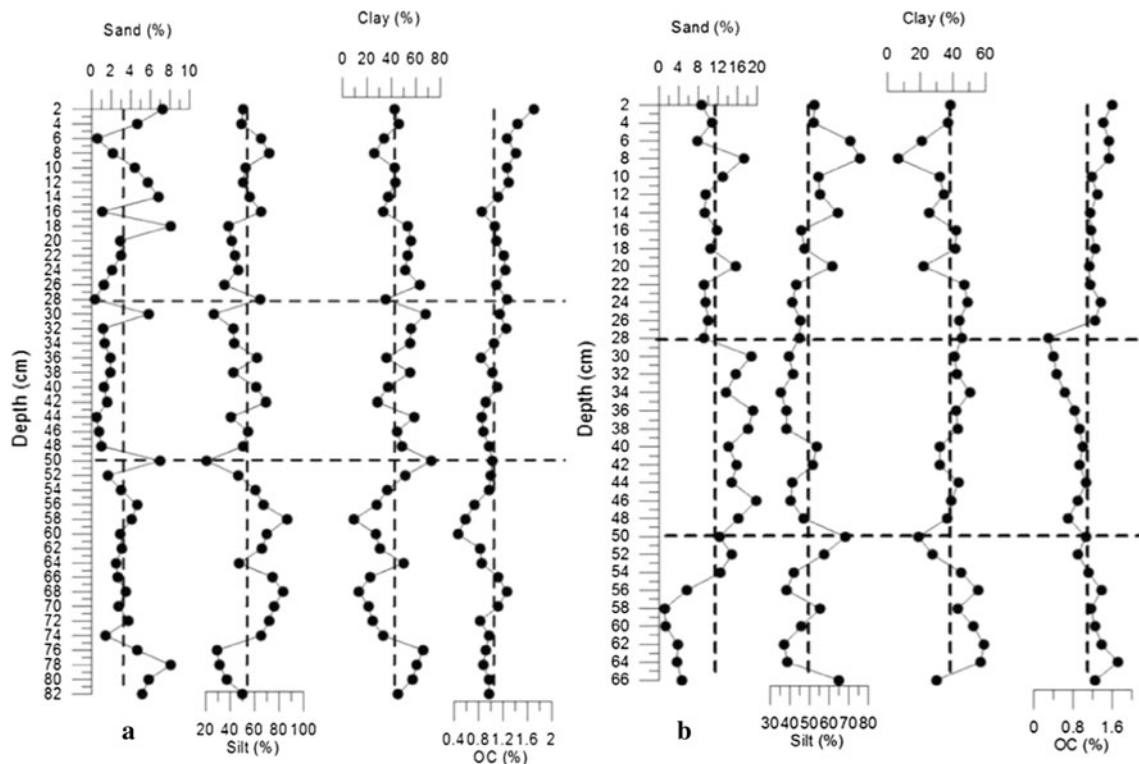
The core S-62, collected from middle flat of lower estuary is also divided into three sections, namely bottom (66–50 cm), middle (50–28 cm) and top (28–0 cm). In the bottom section (Fig. 2b), sand shows increase whereas silt and clay show fluctuating trends. In the middle section, clay percentage increases while silt percentage decreases, with some variations. Sand maintains higher values than average. In the top section, sand is nearly constant; silt percentage increases while clay percentage decreases.

The cores S-63 and S-45 collected from middle tidal flats in lower and upper regions of middle estuary are divided into three sections namely bottom (66–50 cm), middle (50–28 cm) and top (28–0 cm). In the bottom section of core S-63 (Fig. 3a), sand and silt percentage increases while clay percentage decreases. In the middle section, sand, silt and clay show large variations. In the top section they fluctuate around the average line. Similarly in core S-45, in bottom and middle section (Fig. 3b), sand remains constant while silt and clay show large fluctuations compensating each other. However in the top section, sand and silt increase and clay decreases. Also silt and clay compensate each other.

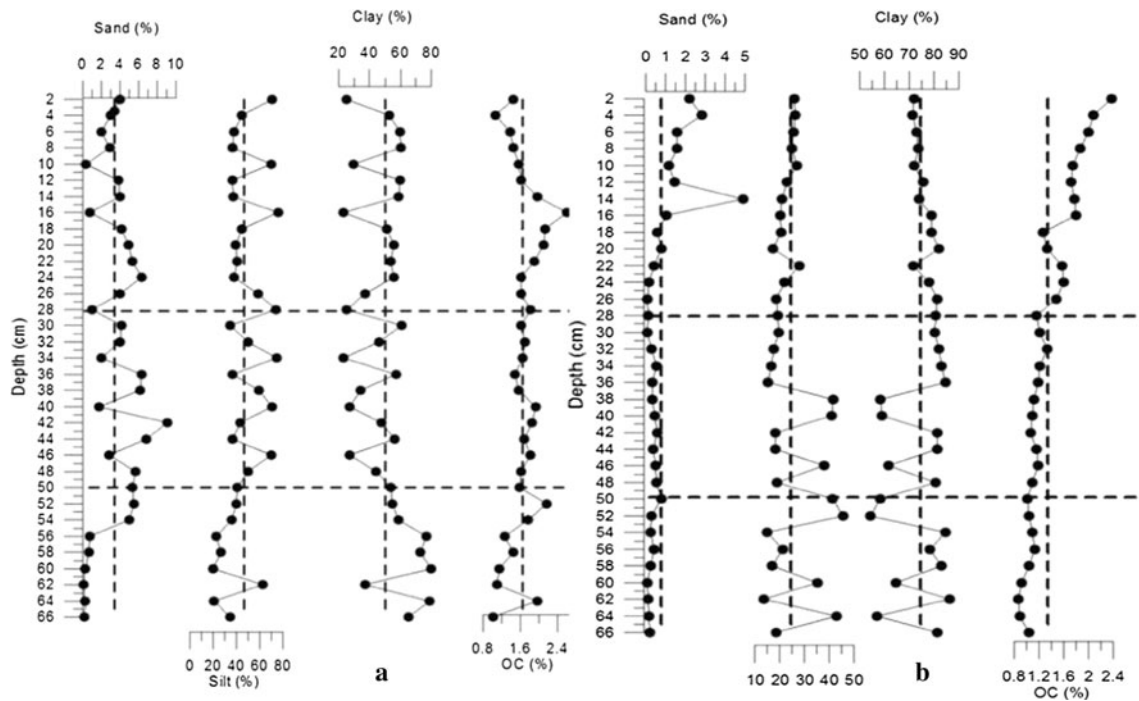
In order to understand the depositional environment, the sediment classification proposed by Flemming (2000) is

**Table 1** Range and average value of sand, silt, clay and OC in lower estuarine mudflats (S-62, S-47) and middle estuarine mudflats (S-63 and S-45)

Parameters (%)	Lower estuary mudflats		Middle estuary mudflats	
	Upper flats (S-47) (%)	Middle flats (S-62) (%)	Middle flats (S-63) (%)	Middle flats (S-45) (%)
Sand	0.3–8.1 (3.29)	1.2–20 (11.52)	0.14–9.1 (3.46)	0.06–4.9 (0.76)
Silt	20.6–86.7 (53.83)	35–76 (49.58)	19.93–75.85 (46.57)	13.47–45.58 (24.62)
Clay	9.2–72.4 (42.88)	6.5–59 (38.90)	23–80 (49.97)	54–86 (74.62)
OC	0.5–1.7 (1.05)	0.3–1.7 (1.10)	1.01–2.6 (1.65)	0.87–2.37 (1.35)



**Fig. 2** Downcore variations of sediment compounds and organic carbon with vertical lines of average value in lower estuary (upper flats) S-47 (a) and lower estuary (middle flats) S-62 (b)



**Fig. 3** Downcore variations of sediment components and organic carbon with vertical lines of average value in middle estuary S-63 (a) and S-45 (b)

used. On the basis of average sand/mud ratios, core S-45, S-63 and S-47 are classified as “Mud” while core S-62 as “Slightly sandy mud” (Fig. 4a). Further, an attempt has

been made to understand the hydrodynamic conditions of depositional environment using ternary diagram proposed by Pejrup (1988). Plot (Fig. 4b) reveals that the cores

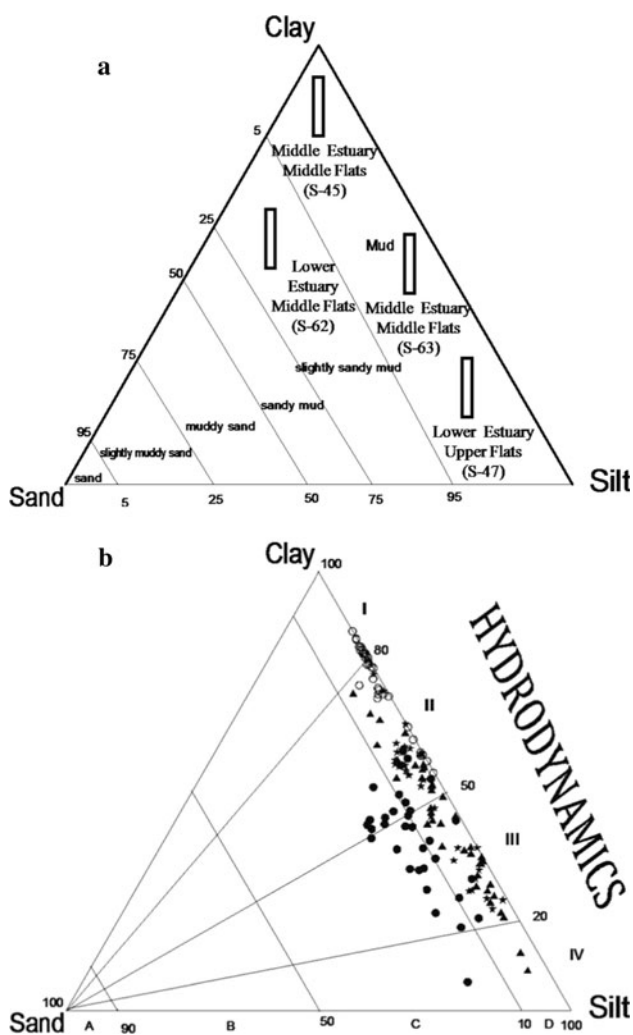
collected from middle (S-62) and upper (S-47) tidal flats of lower estuary fall largely within III and II sections indicating that sediment deposition took place under relatively violent to less violent conditions. However, the sediments in the upper flats (S-47) fall within D type, while the middle flats core (S-62) fall within C type. Sediment core (S-63) collected from lower middle estuary falls largely within sections III and II (Fig. 4b) while core (S-45) collected from upper middle estuary falls largely within sections II and I indicating that sediment deposition took place under less violent to calm conditions. Both the cores (S-63 and S-45) fall within D type, in the middle estuary.

The major difference among the cores is that cores collected from lower estuary are dominated by silt component whereas cores collected from middle estuary are

dominated by clay component (Table 1). In all the four cores, the variations of silt are compensated by those of clay.

In the lower estuarine mudflats (S-47 and S-62), average sand content is found to be more than threefold higher in middle flats (S-62) when compared to upper tidal flats (S-47) (Table 1). The sediments in upper flats (S-47) are typically muddy with mud content up to 97 %. Wave attenuation (Semeniuk 1981) must be responsible for landward migration of finer sediments and decreasing grain size from un-vegetated to vegetated surface (Yang et al. 2008; Shi and Chen 1996). The mangrove vegetation, abundant on upper flats, is associated with a unique horizontal root network (Kumaran et al. 2004) that enhances the deposition of fine grained suspended sediments and prevents re-suspension and erosion of fine sediments (Soto-Jime'nez and Pa'ez-Osuna 2001).

Likewise, when the distribution of sediment components along middle tidal flats (S-62, S-63, S-45) is compared (Table 1), it is observed that sand showed a “band-type” distribution, increasing from the minimum at the upper region of middle estuary (S-45) to the maximum at the lower estuary (S-62), indicative of increasing energy conditions from middle to lower estuary (Lorenzo et al. 2007). The average sand is less than 1 % in upper region of middle estuary (S-45) while more than 11 % in lower estuary (S-62) (Table 1). Klein (1985) reported that intertidal deposits typically exhibit coarsening in seaward direction. Chen (1992) discussed the role of hydrodynamics in sediment transport process on the intertidal mudflats. The relatively high tidal energy in lower estuary results in higher hydro dynamics near the mouth which facilitates the deposition of coarser sediments as finer sediments are more mobile and are carried to the middle estuary where tidal currents are weaker resulting in the deposition of mud (Manning et al. 2010). Thus, differences in average sand content are due to variations in tidal range at two places (Feng 1985; Cao et al. 1989). The high range (13.7–20 %) and average (~17 %) sand percentage are observed in middle section (28–50 cm) of middle tidal flats (S-62) in lower estuary (Fig. 2b). Sand is transported as bed load during strong flood and ebb currents during spring tides, high river discharge (Walsh and Nittrouer 2004) or during extreme events such as storm surges (Gao 2009a). One or more of these factors may be responsible for higher sand content in the middle section. Additionally, factors such as agricultural practices/anthropogenic activities, including sand mining, may also contribute to increased sand content (Dandekar 2010). A high range and average sand content in middle section (24–44 cm) are also reported from mudflats of nearby Ulhas Estuary (Fernandes and Nayak 2010). On the other hand middle tidal flats of upper region in middle estuary (S-45) consist primarily of clay component with an



**Fig. 4** **a** Ternary diagrams for a revised textural classification of sediments on the basis of sand/mud ratios after Flemming (2000). **b** Triangular diagram for the classification of hydrodynamics after Pejrup (1988) for lower estuary upper flat core S-47 (filled triangle), lower estuary upper flats S-62 (filled circle), middle estuary middle flats S-63 (filled star) and S-45 (open circle)

average of 75 %, deposited possibly from turbidity maxima (Dyer 1986) developed within this part of estuary. The clay content decreases from 75 % and becomes nearly half (39 %) in lower estuary (S-62) while the silt content increases from 25 % in middle estuary (S-45) and becomes double (50 %) in lower estuary (S-62) (Table 1). Pejrup et al. (1997) stated that the transportation and deposition of fine grained sediments in a tidal basin is governed mainly by physiographical and hydro dynamical parameters. The fine grained sediments generally move in suspension and to some extent saltation, with the tide and finally get deposited in the upper reaches of middle estuary.

### Organic carbon

The range and average percentage of organic carbon for each core is given in Table 1.

In the core S-47, collected from upper flats of lower estuary, organic carbon is nearly constant with values falling near the average line except between 64–56 cm and 16–0 cm where organic carbon values fall below and above the average line, respectively (Fig. 2a). In core S-62, collected from middle flats, organic carbon values are more than average in bottom and top sections while less than average in middle section (Fig. 2b). In middle estuary (S-63), in bottom section organic carbon is less than average, in middle section organic carbon is constant with values falling on the average line. In top section between 28 and 10 cm values are more than average while between 10 and 0 cm values are less than average (Fig. 3a). In S-45 in bottom and middle sections, organic carbon values fall below the average line while in the top section, values fall around the average line (Fig. 3b) but overall show an increasing trend from bottom to top of the core.

Like sediment component, when the distribution of organic carbon along middle tidal flats (S-62, S-63 and S-45) are compared, it is observed that organic carbon also showed “band-type” distribution which decreases with increasing coarser fraction from maximum in middle to minimum in lower estuary. High organic carbon content is found in the middle estuary (S-63 av. 1.65 %; S-45 av. 1.35 %) where the sediments are muddy (>95 % mud) and relatively low content (S-62 av. 1.1 %) near the mouth where sediments are slightly sandy mud (75–95 % mud) (Flemming 2000) (Table 1). According to Muzuka and Shaghude (2000), muddier sites contain higher content of organic carbon relative to sandier sites which is mainly attributed by them to factors such as surface area/volume ratio of sediment grain. However, in addition, the high terrestrial input from the adjacent land masses (Jonathan et al. 2004) must be responsible for high organic carbon concentration in the middle estuary. Further, the high organic carbon content along with high percentage of finer

fraction (>95 %) in the middle estuary suggests calm environment of deposition (Kumar and Edward 2009).

The depth-wise distribution of organic carbon in cores showed high and increased percentage (S-47; S-62; S-45) (Figs. 2a, b, 3b) in the top section (0–28 cm) and relatively low percentage (S-47; S-63; S-45) (Figs. 2a, 3a, b) in the bottom section indicating degradation of organic matter with depth. The high and increased organic carbon (S-45 1.2–2.4 %; S-63 1.07–2.6 %; S-62 0.3–1.6 %; S-47 0.9–1.1 %) in the top section (0–28 cm) is mainly attributed to extensive use of fertilizers, population growth and increased inputs of particulate sedimentary matter and organic carbon associated with urban waste (Fernandez et al. 2003; Zourarah et al. 2009). However in lower estuary (S-62), low concentration (0.4–1.1 %) is found in the middle section (28–50 cm) which reflects the dilution by addition of coarse grained (>63  $\mu\text{m}$ , 14–20 %) sediments (Pichaimani et al. 2008) because of poor absorbability of organics on negatively charged quartz (Chatterjee et al. 2007). Relatively high concentration (0.9–1.7 %) in the bottom section (54–66 cm; 1.12–1.71 %) is due to the presence of high percentage of finer sediments (<63  $\mu\text{m}$  av. 93 %).

When the two cores (S-63 and S-45) (Table 1), one from lower region (S-63) and other from upper region (S-45) of middle estuary, are compared, relatively high average organic carbon concentration is found in lower region of middle estuary (S-63). Although organic carbon generally depends upon the grain size and is enriched in fine grained sediments (Falco et al. 2004), factors such as balance between accumulation and degradation rate of organic matter and the source and type of organic material are keys which control the concentration of organic carbon in sediments (Schorer 1997).

### Major elements (Fe, Mn, Al)

In the core S-47 collected from upper flats of lower estuary (Fig. 5a), in the bottom section (82–50 cm), Fe and Mn generally fluctuate around average line while Al is lower than their average values. In the middle (50–28 cm), Al increases with values more than average while Fe and Mn concentration decreases with values less than their respective averages. In the top section (28–0 cm), Al content decreases while Fe and Mn concentration increases with values mostly more than their respective averages. In the core S-62, collected from middle flat of lower estuary (Fig. 5b), in the bottom section (66–50 cm), Fe and Mn fluctuate with overall increasing trend while Al concentration increases up to 56 cm. Negative peak of Fe at 54 cm depth coincides with that of Al indicating their common terrigenous source. In the middle (50–28 cm) Al shows increasing trend while Fe and Mn show decreasing



trend. In the top section (28–0 cm), Fe, Mn and Al show large variations with values falling around the average line. In the core S-45 collected from middle tidal flats of middle estuary (S-45) (Fig. 5c), in the bottom section (66–50 cm), Al is nearly constant with values falling on the average line, Fe and Mn values are less than their respective averages. In the middle (50–28 cm), Fe, Mn and Al values fluctuate around the average line. In the top section (28–0 cm), Al, Fe and Mn values are more than their respective averages.

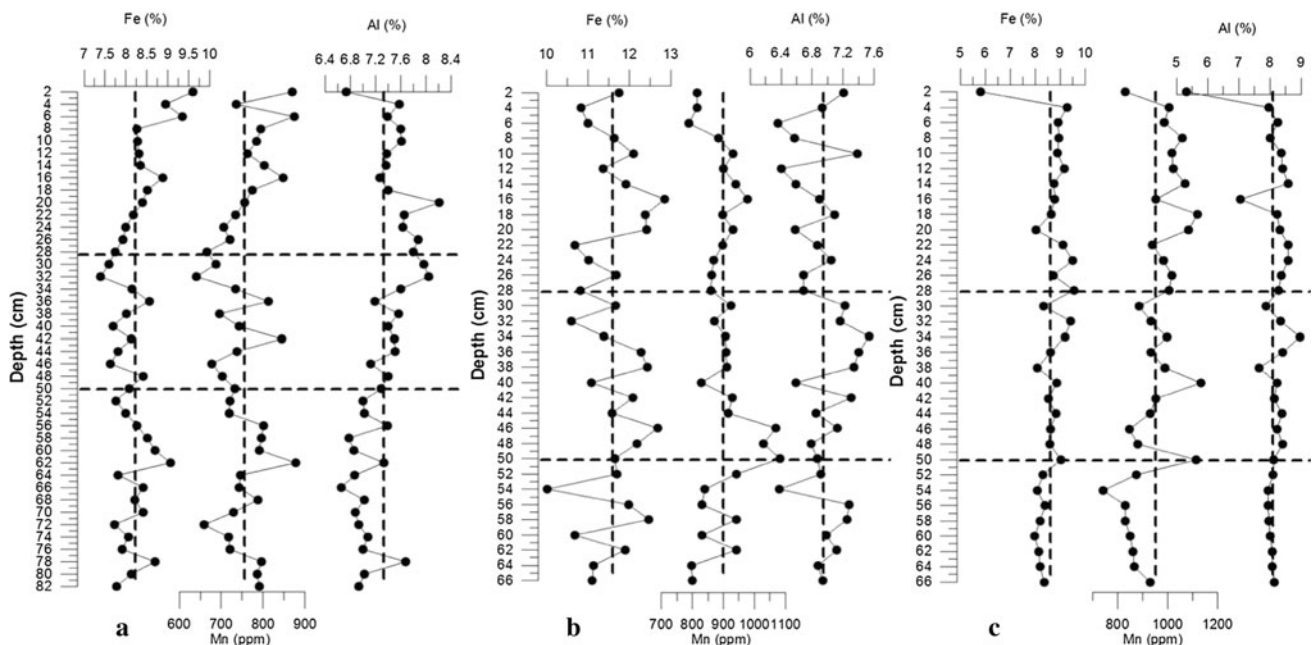
In lower estuarine mudflats (S-47, S-62), average Fe and Mn content is found to be higher in middle flats (S-62) whereas average Al content is higher in upper flats (S-47) (Table 2). Fe and Mn are diagenetically mobile in the aquatic systems. The vertical profile of Fe and Mn in middle flats (S-62) showed reduced concentration at surface (0–8 cm). This indicates that Fe and Mn ions with greater mobility are removed from the surface sediments to the upper water column through active diffusion and advective processes (Janaki-Ramann et al. 2007; Badr et al. 2009), whereas in upper flats (S-47), the vertical profiles of Fe and Mn show enrichment in the top section (20–0 cm) due to early diagenetic process. Klinkhammer et al. (1982) stated that  $\text{Fe}^{+2}$  and  $\text{Mn}^{+2}$  species get precipitated in the top layers in mangrove sediments as these elements diffuse upwards. The synchronous decrease of Fe and Mn profile below 20 cm, i.e. between 32 and 20 cm (Fig. 5b), suggests oxic/sub-oxic interface (Santschi et al. 1990).

Likewise, when the distribution of major elements (Fe, Mn and Al) along middle tidal flats (S-62 and S-45) is

compared (Table 2), it is observed that average Al and Mn content is greater in the middle estuary (S-45), whereas average Fe content is more in lower estuary (S-62). The Al content is considered as a good indicator of the amount of finer clay material (Ram et al. 2003) of terrestrial origin. The result of sand–silt–clay analysis clearly indicated twofold increase in clay content in middle estuary (S-45) when compared to lower estuary (S-62). Therefore, the observed higher concentration of Al here is attributed to high clay content in middle estuary (S-45). The high Al and Mn content in middle estuary (S-45) also reflects greater input by mechanical and chemical weathering of rocks (De-Carlo and Anthony 2002). On the other hand, elevated Fe content in lower estuary (S-62) could be attributed to terrigenous supply of ferromagnesian minerals, industrial and municipal discharges (Sagheer 2004; Bhagure and Mirgane 2011) and greater recycling of Fe by resuspension of bottom sediments due to tidal mixing in lower estuarine region (Kumar and Edward 2009). The depth-wise distribution of Fe and Mn in middle estuary (S-45) indicates enrichment in top section (0–28 cm) due to the precipitation of these redox sensitive elements as hydroxides and oxides, whereas the low Fe and Mn concentrations in the bottom section (66–50 cm) reflect their dissolution.

Trace metals (Ni, Cr, Co, Zn, Pb)

In the core S-47 collected from upper flats of the lower estuary, in the bottom section (82–50 cm) (Fig. 6a), Ni, Cr, Co and Zn to some extent is higher while Pb shows lower



**Fig. 5** Downcore variations of major elements with vertical lines of average value in **a** lower estuary (upper flats) S-47, **b** lower estuary (middle flats) S-62 and **c** middle estuary (middle flats) S-45

**Table 2** Range and average value of major (Al, Fe and Mn) and trace (Ni, Cr, Co, Zn and Pb) elements in lower estuarine upper flats (S-47), lower estuarine middle flats (S-62) and middle estuarine middle flats (S-45)

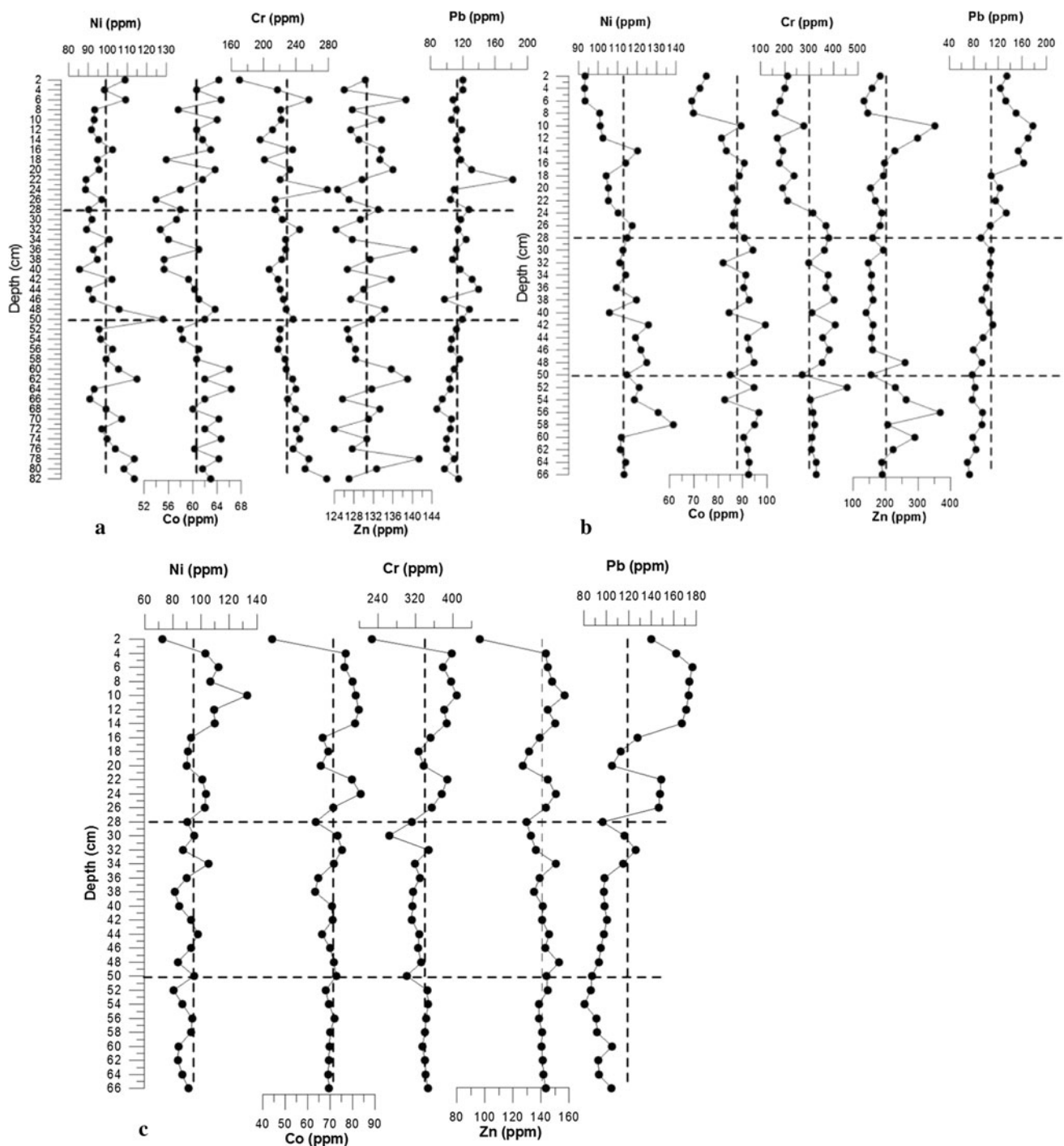
Metals	Lower estuary		Middle estuary
	Upper flats (S-47)	Middle flats (S-62)	Middle flats (S-45)
Fe (%)	7.4–9.6 (8.23)	10–12.8 (11.59)	5.83–9.57 (8.61)
Mn (ppm)	642–878 (757)	790–1083 (900)	742–1134 (954)
Al (%)	6.7–8.2 (7.33)	6.4–7.5 (6.95)	5.31–8.97 (8.10)
Ni (ppm)	86–128 (99)	93–139 (113)	73–133 (95)
Cr (ppm)	170–279 (229)	160–457 (299)	227–408 (341)
Co (ppm)	54–66 (61)	69–99 (88)	44–84 (71)
Zn (ppm)	124–140 (131)	134–368 (203)	96–157 (141)
Pb (ppm)	95–182 (114)	70–178 (109)	80–177 (119)

than their respective averages. Ni and Cr show decreasing trend while Pb increases, Co and Zn show large fluctuations. In the middle section (50–28 cm), Ni, Co and Zn display an overall decreasing pattern while Cr and Pb show constant trend. In the top section (28–0 cm), Ni, Co, Zn and to some extent Pb show an increasing trend while Cr decreases with large variations. In the core S-62, collected from middle flat of lower estuary, in the bottom section (66–50 cm) (Fig. 6b), Ni, Cr and Co decreases while Pb concentration increases with some variations. Zn shows an overall increase till 56 cm then shows gradual decrease. In the middle section (50–28 cm), Ni, Cr and Co are higher while Zn and Pb concentration is lower than their respective average with Ni, Cr, Co showing decreasing trend, Zn constant and Pb increasing trend. In the top section (28–0 cm), Ni, Co and Cr show decreasing trend from 28 to 6 cm whereas Zn and Pb display an increase in concentration reaching the highest value at 10 cm. All the metals show increase in concentration at surface (0–6 cm). In the core S-45 collected from middle tidal flats middle estuary (S-45) (Fig. 6c), in the bottom section (66–50 cm) and middle section (50–28 cm) almost all the metals except Pb display constant trend with values below or around the average line while in the top section (28–0 cm), between 28 and 4 cm all the metals show values above the average line followed by drastic decrease at surface. Pb values, however, were much less than average in bottom and middle sections and show an overall increasing trend in middle and top sections.

In lower estuarine mudflats (S-47 and S-62), average Ni, Cr, Co and Zn content is found to be more along middle flats (S-62) when compared to upper flats (S-47) (Table 2). The intertidal sediment is redistributed by tidal waters which carry suspended sediment from sub tidal to intertidal areas. Semeniuk (1981) stated that the middle flats are inundated by flowing tidal waters for 80 % of their time and for about 50 % of the time in upper flats. Wave attenuation along with frequency of tidal inundation rate must be responsible for landward decrease in trace metals

content. The vertical profile of trace elements indicated a decreasing trend in middle flats (S-62) with almost all metals exceeding their average values in the bottom (66–50 cm) and middle section (28–50 cm). Sand and organic carbon also exceeded their average value in middle (50–28 cm) and bottom (66–50 cm) sections, respectively. The elevated metal concentration may therefore be due to their association with detrital sand (i.e. mining and smelt products) particles and organic matter rich in heavy metals in middle and bottom section, respectively. Similar enrichment of trace metals in coarse particles has been reported in estuaries subjected to mining (Cundy et al. 2003). The reduced concentration of trace elements in surface sediments (0–12 cm) is similar to that of Fe–Mn and is attributed to diagenetic removal or diffusion process. In the upper flats (S-47), a fluctuating trend with metal enrichment in upper layers of the top section (0–28 cm) is observed. Zorana et al. (2009) stated that metal enrichment in the upper layer is not necessarily due to anthropogenic input only. This enrichment also may be explained by the redox sensitive elements (Fe and Mn) which show similar increase toward the surface (20–0 cm). The fluctuating metal profile may be attributed to varying hydrodynamic energy conditions prevailing in upper flats as mentioned earlier as well as bioturbation by organisms associated with mangroves (Clark et al. 1998; Gao 2009b).

Likewise, when the distribution of trace elements (Ni, Cr, Co, Zn and Pb) along middle tidal flats (S-62 and S-45) is compared (Table 2), it is observed that average content of Ni, Co and Zn is more in the lower estuary (S-62) whereas average Cr and Pb content is more in middle estuary (S-45). The distribution of metals depends upon factors such as the distance of element sources to estuary, hydrodynamics, the chemical characteristics (e.g. sorption-adsorption capacity of trace metals, flocculation, etc.), and the chemical and biochemical condition of sedimentary environment (Williams et al. 1994). The depth-wise distribution showed different trends in metal profiles in lower (S-62) and middle



**Fig. 6** Downcore variations of trace elements with vertical lines of average value is **a** lower estuary (upper flats) S-47, **b** lower estuary (middle flats) S-6. **c** Downcore variation of trace elements with vertical lines of average value in middle estuary (middle flats) S-45

(S-45) estuaries. In middle estuary (S-45) the metals analyzed are enriched in the top section (28–4 cm) with a constant trend in middle (50–28 cm) and bottom (66–50 cm) sections. Clay, organic carbon and major elements (Al, Fe and Mn) also show enrichment in top section similar to trace elements thus indicating a common source. However, at surface all the metals including Fe, Mn and Al

show drastic decrease which may be due to erosion of surface sediment or dredging activities.

#### Correlation

The good association of metals with finer particles in S-62 (Table 3b) and with organic carbon in S-45 (Table 3c)

**Table 3** Pearson correlation between sediment components and metals in lower estuarine upper flats (S-47), lower estuarine middle flats (S-62) and middle estuarine middle flats (S-45)

	Sand	Silt	Clay	OC	Fe	Mn	Al	Ni	Cr	Co	Zn	Pb
(a) Lower Estuary Upper Flats (S-47) ( $n = 41$ )												
Sand	1.00											
Silt	-0.34	1.00										
Clay	0.21	-0.99	1.00									
OC	0.12	-0.15	0.14	1.00								
Fe	0.27	0.25	-0.30	0.16	1.00							
Mn	0.26	0.30	-0.35	-0.03	<b>0.77</b>	1.00						
Al	-0.14	-0.45	<b>0.48</b>	0.29	-0.20	-0.24	1.00					
Ni	<b>0.39</b>	-0.12	0.07	-0.19	<b>0.44</b>	<b>0.44</b>	-0.28	1.00				
Cr	-0.16	-0.01	0.03	-0.26	-0.18	-0.11	-0.05	0.28	1.00			
Co	0.19	0.21	-0.25	-0.20	<b>0.48</b>	<b>0.41</b>	-0.45	<b>0.45</b>	0.24	1.00		
Zn	0.07	0.04	-0.05	-0.22	<b>0.53</b>	<b>0.57</b>	0.10	<b>0.41</b>	0.08	<b>0.44</b>	1.00	
Pb	-0.22	0.07	-0.04	0.17	-0.16	-0.25	0.26	-0.18	-0.15	-0.14	0.09	1.00
(b) Lower Estuary Middle Flats (S-62) ( $n = 33$ )												
Sand	1.00											
Silt	-0.06	1.00										
Clay	-0.38	-0.90	1.00									
OC	-0.53	0.33	-0.07	1.00								
Fe	0.29	0.01	-0.14	-0.03	1.00							
Mn	<b>0.44</b>	0.01	-0.21	-0.39	<b>0.61</b>	1.00						
Al	0.11	-0.51	<b>0.43</b>	-0.26	<b>0.39</b>	0.18	1.00					
Ni	-0.05	-0.30	0.30	-0.35	0.31	<b>0.39</b>	0.25	1.00				
Cr	0.22	-0.51	<b>0.38</b>	-0.54	0.07	0.16	<b>0.47</b>	<b>0.62</b>	1.00			
Co	0.00	-0.53	<b>0.50</b>	-0.43	<b>0.38</b>	<b>0.37</b>	<b>0.53</b>	<b>0.76</b>	<b>0.72</b>	1.00		
Zn	-0.34	-0.13	0.27	0.18	0.05	-0.01	0.07	0.23	-0.01	0.28	1.00	
Pb	0.13	<b>0.34</b>	-0.37	0.23	0.18	-0.02	-0.15	-0.49	-0.66	-0.47	0.14	1.00
(c) Middle Estuary Middle Flats (S-45) ( $n = 33$ )												
Sand	1.00											
Silt	-0.03	1.00										
Clay	-0.07	-0.99	1.00									
OC	<b>0.68</b>	-0.13	0.05	1.00								
Fe	-0.04	-0.10	0.10	-0.04	1.00							
Mn	0.32	0.10	-0.14	0.25	<b>0.48</b>	1.00						
Al	-0.17	-0.12	0.13	-0.39	<b>0.78</b>	0.33	1.00					
Ni	<b>0.37</b>	-0.18	0.14	<b>0.44</b>	<b>0.56</b>	<b>0.41</b>	<b>0.47</b>	1.00				
Cr	0.29	-0.07	0.04	0.28	<b>0.59</b>	0.23	<b>0.53</b>	<b>0.68</b>	1.00			
Co	0.18	-0.04	0.02	0.10	<b>0.75</b>	0.32	<b>0.71</b>	<b>0.73</b>	<b>0.79</b>	1.00		
Zn	-0.01	0.05	-0.05	-0.20	<b>0.70</b>	0.18	<b>0.79</b>	<b>0.60</b>	<b>0.71</b>	<b>0.81</b>	1.00	
Pb	<b>0.62</b>	-0.12	0.06	<b>0.85</b>	0.26	<b>0.40</b>	0.01	<b>0.72</b>	<b>0.57</b>	<b>0.51</b>	0.17	1.00

Correlation highlighted in bold are significant at  $p < 0.05$

along middle flats indicates that their interaction with clay particles and organic matter is an important process for removal and fixation of Cr, Co and Pb in lower estuary (S-62) and Ni and Pb in middle estuary (S-45), respectively. On the other hand, some elements do not show good

association with clay and organic carbon as the interaction among metals, clay and organic carbon depends upon many factors including charge on metal ions (Bartoli et al. 2012). Further, organic carbon and clay content alone cannot be a decisive factor in the mechanism of metal accumulation in

sediments. The good association between sand and Mn in middle flats of lower estuary (S-62) is attributed to Mn oxide coatings on the sand grains (Badr et al. 2009). The mudflat sediments are dominated by finer components which consist of larger surface-active fractions and Fe–Mn oxide coating. The Fe–Mn oxides provide metal adsorbing sites and are known to co-precipitate trace elements (Carman et al. 2007). Strong relationship of trace metals with Fe–Mn therefore indicates that they are associated with Fe–Mn oxyhydroxides (Chatterjee et al. 2007; Jonathan et al. 2010) (Table 3a–c). Inter-relationships among metals in lower and middle estuary indicate a common source or a similar enrichment mechanism at their respective places. Further, good association of metals with Al in middle tidal flats (S-45 and S-62) indicates that these metals have terrigenous origin whereas the negative correlations found between Al and most of the metals analyzed in upper flats (S-47) of lower estuary indicated a distinct non-terrigenous source. Thus, in addition to Fe–Mn oxyhydroxides and aluminosilicates, nature/texture of the sediments, i.e. clay, silt or sand, organic carbon are important factors controlling the distribution and fate of metals along lower and middle estuaries.

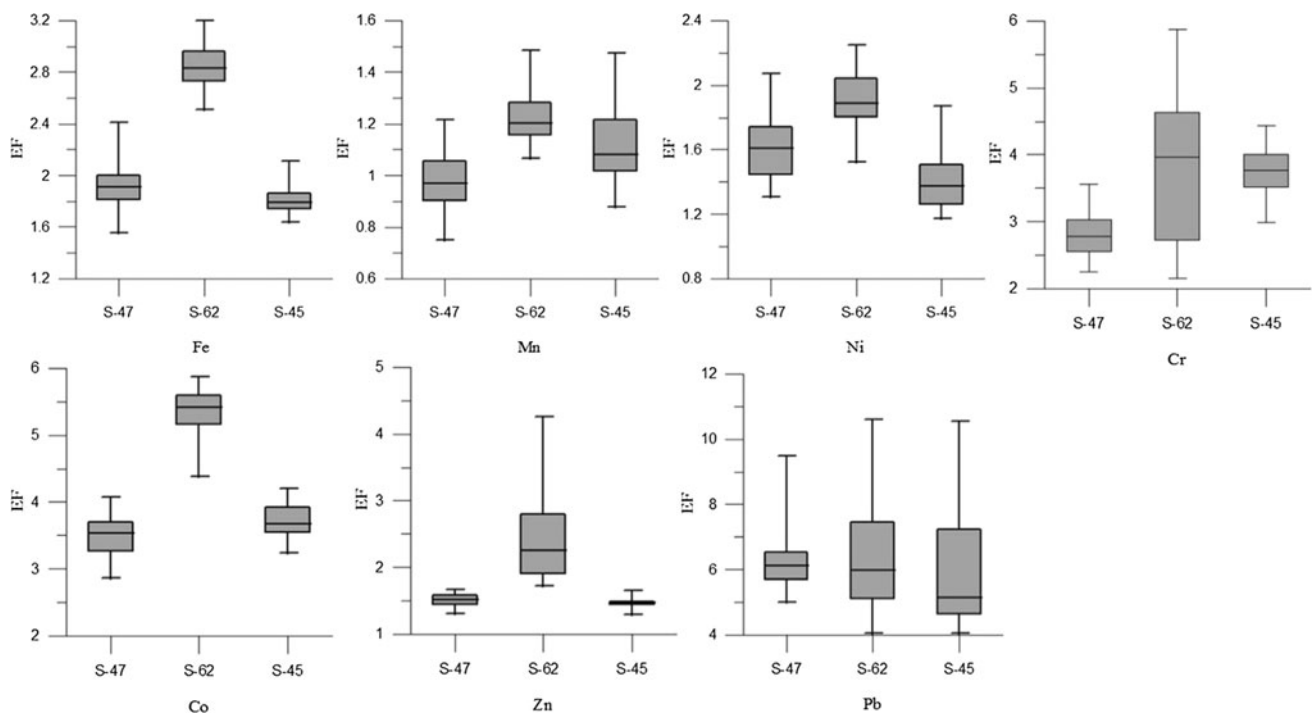
#### Enrichment factor (EF)

Figure 7 shows EF values of metals. If an EF value is between 0.5 and 1.5, it suggests natural input and if an EF

value is greater than 1.5, it suggest anthropogenic input of metals (Sarkar et al. 2011; Zhang and Liu 2002). In the present study, EF value of nearly all elements (except Mn) is greater than 1.5 suggesting anthropogenic input. The possible sources of anthropogenic input of metals into river are: (1) domestic waste directly discharged into the river without any treatment; (2) accidental discharges of effluents directly or indirectly into the river through nallas, from industrial areas; (3) unauthorized disposal of hazardous waste and (4) washing of chemical tankers in the river (Maharashtra Pollution Control Board 2004–2005; Maharashtra Pollution Control Board 2002–2003; Dandekar 2010).

#### Factor analysis

Factor analysis was employed on the dataset after removing critically enriched samples (Tables 4, 5, 6). This method was applied to understand the factors responsible for enrichment of 'background' metals. In the middle flats of lower (S-62) and middle (S-45) estuaries, the four factors (F1, F2, F3 and F4) correspond to 81.45 and 83.34 %, respectively. F1, F2, F3 and F4 accounted for 24.43, 21.72, 21.20 and 14.10 %, respectively, in the lower estuary (S-62) and 33.20, 19.13, 18.02 and 12.99 %, respectively, in the middle estuary (S-45) (Tables 4, 5, 6). In upper flats of the lower estuary (S-47), the five factors correspond to 84.04 % of the total variance while F1, F2, F3, F4 and F5



**Fig. 7** Box and Whisker plot of enrichment factor (EF) for lower estuary upper flats (S-47), lower estuary middle flats (S-62) and middle estuary middle flats (S-45)

**Table 4** Factor analysis after varimax rotation for lower estuary upper flats (S-47) after removing seriously polluted samples

Factor	F1	F2	F3	F4	F5
Variance (%)	26.81	22.34	13.23	10.89	10.77
Sand	0.221	0.213	-0.142	<b>0.868</b>	-0.009
Silt	0.146	-0.957	0.001	-0.176	-0.003
Clay	-0.182	<b>0.956</b>	0.019	0.058	0.004
OC	-0.123	0.071	0.021	0.022	<b>0.962</b>
Fe	<b>0.877</b>	-0.203	0.046	0.131	0.134
Mn	<b>0.839</b>	-0.256	-0.157	0.131	-0.025
Al	0.052	<b>0.685</b>	-0.244	-0.328	0.453
Ni	<b>0.698</b>	0.041	0.395	0.169	-0.326
Cr	0.154	0.053	<b>0.913</b>	-0.144	0.017
Co	<b>0.609</b>	-0.237	0.400	0.049	-0.162
Zn	<b>0.849</b>	0.152	0.039	-0.132	-0.103
Pb	0.134	0.375	-0.574	-0.554	-0.001

Number of samples = 35

Loadings highlighted in bold are >0.5 significant at  $p < 0.05$ **Table 5** Factor analysis after varimax rotation for lower estuary middle flats (S-62) after removing seriously polluted samples

Factor	F1	F2	F3	F4
Variance (%)	24.43	21.72	21.20	14.1
Sand	0.104	-0.040	<b>0.551</b>	-0.731
Silt	-0.379	-0.853	-0.023	0.069
Clay	0.316	<b>0.833</b>	-0.224	0.260
OC	-0.478	-0.250	-0.186	0.562
Fe	0.030	0.049	<b>0.868</b>	0.003
Mn	0.332	0.086	<b>0.836</b>	-0.069
Al	-0.029	<b>0.741</b>	<b>0.503</b>	-0.120
Ni	<b>0.937</b>	0.043	0.201	0.001
Cr	<b>0.733</b>	0.463	0.008	-0.263
Co	<b>0.735</b>	0.474	0.306	0.137
Zn	0.061	0.090	0.186	<b>0.807</b>
Pb	-0.614	-0.335	<b>0.530</b>	0.101

Number of samples = 26

Loadings highlighted in bold are >0.5 significant at  $p < 0.05$ 

accounted for 26.81, 22.34, 13.23, 10.89 and 10.77 %, respectively (Table 4).

Factor analysis was used to identify the factors that control trace metal distribution (Selvaraj et al. 2004) and fate. In the middle flats of the lower estuary (S-62), F1 showed significant loadings on Ni, Cr and Co and in F2, Al and clay showed significant loadings. Fe, Mn, Al and sand showed significant loadings in F3 and in F4, organic carbon and Zn showed significant loadings (Table 5). In upper flats of the lower estuary (S-47) in F1, Fe, Mn, Ni, Co and Zn showed significant loadings; in F2, Clay and Al showed

**Table 6** Factor analysis after varimax rotation for middle estuary middle flats (S-45) after removing seriously polluted samples

Factor	F1	F2	F3	F4
Variance (%)	33.20	19.13	18.02	12.99
Sand	<b>0.773</b>	0.060	-0.088	-0.038
Silt	-0.101	-0.072	-0.983	0.030
Clay	0.030	0.066	<b>0.992</b>	-0.027
OC	<b>0.884</b>	0.318	0.119	0.006
Fe	0.205	<b>0.869</b>	0.075	0.135
Mn	0.309	<b>0.665</b>	-0.240	-0.521
Al	0.273	<b>0.755</b>	0.240	0.345
Ni	<b>0.738</b>	0.425	0.212	0.162
Cr	<b>0.712</b>	-0.037	0.070	0.455
Co	<b>0.702</b>	0.270	0.002	0.437
Zn	0.235	0.244	-0.142	<b>0.846</b>
Pb	<b>0.884</b>	0.314	0.089	0.086

Number of samples = 28

Loadings highlighted in bold are >0.5 significant at  $p < 0.05$ 

significant loadings while in F3 Cr showed significant loadings. F4 showed significant loadings on sand and in F5, organic carbon showed significant loadings (Table 4). In the middle flats of middle estuary (S-45), F1 showed significant loadings on sand, organic carbon, Ni, Cr, Co and Pb; F2 showed significant loadings on Fe, Mn and Al; F3 showed significant loadings on clay and F4 showed significant loadings on Zn (Table 6).

In upper flats of the lower estuary (S-47), Fe, Mn and Al played an important role in the distribution of Ni, Co and Zn. F3 in middle flats (S-62), and F1 in upper flats (S-47) can be called Fe–Mn controlled factors. In middle flats of middle estuary (S-45), Al together with Fe and Mn in F2 can be called Fe–Mn–Al controlled factor. Fe, Mn and Al are the major components of silica minerals that are the products of rock and soil weathering on land (Zhou et al. 2004). This group can be called a lithogenic group, as the variability of these elements appears to be controlled by terrestrial and natural sources. However in the middle flats of lower estuary (S-62), Ni, Cr and Co are grouped together in Factor 1 and showed no significant association with Fe, Mn or Al. The weak correlation with Fe, Mn and Al suggests a different source. The significant association of these metals among each other suggests a common source, possibly anthropogenic, such as industrial and agricultural activities. In the lower estuary (S-62 and S-47), organic carbon seems to play a minor role in distribution of metals, except Zn in middle flats, where it shows good association. However in middle estuary (S-45), organic carbon played a major role in the distribution of Ni, Cr, Co and Pb. Factor 1 in middle estuary (S-45) and Factor F4 in lower estuary (S-62) can be called organic carbon-controlled factors.

**Table 7** Concentration of metals in core sediments of Kundalika Estuary and comparison with metal data of estuaries along west coast of India

Location	Fe (%)	Mn (ppm)	Al (%)	Ni (ppm)	Cr (ppm)	Co (ppm)	Zn (ppm)	Pb (ppm)	References
Kundalika Estuary	5.83–12.8	642–1,134	5.31–8.97	73–139	160–408	44–99	96–368	70–182	Present Study
Dharamtar Creek	7–9.32	646–923	6.71–10.06	81–127	117–322	46–81	112–218	6.67–133	Pande and Nayak (2012)
Thane Creek	10	1,000–2,000	11.4	–	150–250	–	100–200	90–100	Fernandes (2011)
Ulhas Creek	4.15–5.04	636–1320	4.45–7.90	61–154	221–7163	30–107	104–147	29–104	Fernandes and Nayak (2010)
Manori Creek	8.9–11.94 (10.26)	744–1274 (1026)	4.23–8.28 (7.25)	20–67 (42)	117–164 (143)	2142 (36)	117–188 (166)	50–75 (62)	Fernandes et al. (2011)
Mandovi Estuary	1.39–1.72 (1.53)	260–349 (296)	0.52–0.74	21–23	37–55	7–13	32–39	37–103	Sirsawar and Nayak (2011)
Kalinadi Estuary	3–5	0–1,000	–	–	80–280	0–80	30–80	–	Singh and Nayak (2009)
Manakudy Estuary	0.45	358.21	–	289.1	482.13	4.004	–	72.62	Kumar and Edward (2009)

## Comparison with other studies

Metal concentrations in core sediments of Kundalika Estuary were compared with other studies carried out along the west coast of India (Table 7). Generally, concentrations of metals in Kundalika Estuary were similar to metal concentrations reported for Dharamtar, Thane, Ulhas and Manori Creek but higher than those reported for Mandovi, Kalinadi and Manakudy Estuary.

## Conclusion

The grain size distribution across the upper and middle flats of lower estuary as well as lower and middle regions of middle estuary is controlled by factors such as wave and tide-controlled hydrodynamics and anthropogenic activities including sand mining. Depth-wise distribution of organic carbon indicated enrichment in top section (0–28 cm) at all the locations studied, possibly due to extensive use of fertilizers, population growth and urban waste. Almost all the elements showed high average content in middle flats when compared to upper flats of lower estuary which is attributed to wave attenuation along with frequency of tidal inundation rate. When the EF values are considered, nearly all elements (except Mn) show anthropogenic enrichment in the Kundalika Estuary. In middle flats of middle estuary, organic carbon played a major role in the distribution of trace metals. In upper flats of the lower estuary, Fe and Mn played a major role in the distribution of Ni, Cr and Zn.

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