

## **Control of source-to-sink processes on the dispersal, fractionation, and deposition of magnetic minerals in a tropical mesotidal estuarine system**

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### **Abstract**

This study sets out to elucidate the control of source-to-sink processes on the sediment dynamics of the complex mesotidal tropical estuarine system using rock magnetic, sedimentological, mineralogical, and geochemical methods. Integration of multiproxy data of catchment rocks, riverbank and hinterland soils, and bedload sediments from various depositional environment of Mandovi estuary revealed marked change in the magnetic mineral composition and accumulation pattern. The magnetic mineralogy mainly comprised of ferri- (magnetite, titanomagnetite) and antiferromagnetic (hematite, goethite) minerals. Higher accumulation of coarser magnetic particles in the catchment area followed by a systematic trend of progressive fining of magnetic grain size in the down reaches of the estuary can be attributed to the decrease in transport energy. Enhanced accumulation of coarser magnetic particles in the estuarine region can be reconciled with the strong tidal and wave-induced bottom currents, which preferentially favoured mineral-density selective fractionation resulting in higher deposition of heavy (magnetic) particles. A clear trend of loss of fluvial derived fine silt-size clastic sediments and selective retention of coarser and heavy magnetic particles within the estuary can be attributed to the hydrodynamic driven sediment partitioning regime which inhibited the permanent settling of fine sediment fraction and are therefore frequently flushed out to the sea. A marked drops in S-ratio in different zones of the estuary can be linked to the increased input of high coercive magnetic particles derived from pedogenic soils along eroding riverbanks. We further demonstrate that S-ratio is an effective parameter to track the riverbank erosion in an estuarine system.

### **Key Points:**

- Control of source-to-sink processes on the sediment magnetism of a mesotidal estuarine system.
- Magnetic signatures of mineral-density and grain size selective sorting and settling mechanism.
- Developed a sensitive magnetic proxy (S-ratio) that can be utilized for tracking riverbank erosion in an estuarine system.

## 1. Introduction

Fluvial systems deliver large amount of magnetic minerals to the estuary derived from catchment rocks, eroded soils (top and subsurface) from hinterland, riverbank, agricultural input and anthropogenic sources (Evans and Heller, 2003; Rey et al., 2005; Franke et al., 2009; Kissel et al., 2016; Nguyen et al. 2016; Kayvantash et al., 2017). Within the estuary, the magnetic minerals derived from different sources get mixed, sorted, enriched, and dispersed depending on the composition (density, grain size) and underlying hydro- and sediment dynamics (Sikora and Kjerfve, 1985; Slingerland and Smith, 1986; Tomkins et al., 2003; Liu et al., 2016 ). Following the differential hydrodynamic properties, heavy magnetic mineral grains get preferentially enriched locally on the bed due to gravitational sorting, while finer and lighter sediment fractions mostly remain as suspended load, gets bi-directionally entrained, and are frequently exported out from the estuary to the marine realm ( Slingerland, 1977; Komar and Wang, 1984; Frihy and Dewidar, 2003; Hatfield et al., 2010; Gallaway et al., 2012). Magnetite settling patterns, concentration, and grain size distributions in different sub-environments of the estuarine system provide vital insights on the processes controlling the accumulation, fractionation, and dispersal of magnetic particles during their transport from source-to-sink (Hatfield et al., 2010; Gallaway et al., 2012; Nguyen et al. 2016; Kulgemeyer et al., 2017; Wang et al., 2020). However, a better understanding of the source-to-sink processes is necessary to elucidate how magnetic minerals respond to changing hydro-and sediment dynamics in different sub-environments of the estuary.

Magnetic minerals are ubiquitous and indicative of sediment constituents in estuarine, coastal, and shelf sedimentary systems (Thompson and Oldfield, 1986; Evans and Heller, 2003; Roberts, 2015). Easy detection of geological and hydrodynamic processes by magnetic methods makes magnetic minerals, an exemplary proxy to gain vital insights into the dynamics of magnetic particles in sedimentary systems (Hatfield, 2014; Kulgemeyer et al., 2017). Magnetic methods have been increasingly used to study the estuarine and coastal systems for example, to decipher the origin, and dynamics of heavy (magnetic) minerals (Hatfield et al., 2010; Gallaway et al., 2012; Kulgemeyer et al., 2017), identify the sediment accretion and erosion sites (Hatfield et al., 2010; Wang et al., 2011; Kayvantash et al., 2017; Franke et al., 2020), track changes in sediment provenance, transport pathways, and depositional system (Booth et al., 2005; Maher et al., 2009; Hatfield, 2014), map the heavy (magnetic) mineral deposits (Badesab et al., 2012), reconstruct the pollution history (Blaha et al., 2011), characterize the sedimentary environment and littoral drift system (Hatfield and Maher,

2008, 2009; Kulgemeyer et al., 2016), and investigate the magnetic mineral diagenesis (Zhang et al., 2007).

Geological setting dominated by volcanic, metamorphic, metasedimentary rocks, and pedogenic soils (top and subsurface) from the riverbank and hinterlands assures abundant supply of detrital magnetic minerals to the Mandovi estuary and these minerals archive the crucial signatures of source-to-sink related sedimentary processes. Ferri- and antiferromagnetic mineral such as magnetite, titanomagnetite, hematite, and goethite are commonly found in source (rocks, soils) and sink (riverine bedload) region of the Mandovi estuary (Gokul et al., 1985; Dhoundial et al., 1987; Naqvi, 2005; Dessai et al., 2009; Mascarenhas and Kalavampara, 2009; Singh et al., 2014; Prajith et al., 2015; Kessarkar et al., 2015). Abundant and seasonally distributed rainfall causes flooding, high surface water runoff, and soil erosion which generate additional sediment load to the Mandovi estuary and their contribution varies over time (Rao et al., 2015). The problem of soil erosion in Mandovi estuary has received increased attention in recent years. Highly concentrated sediment plume in the catchment and other parts of the Mandovi estuary is mainly linked with heavy rainfall, increased land-use, grazing, and agricultural activities which have jointly enhanced the soil erosion (Ibrampurkar, 2012). Soil erosion is a complex process and interlinked by many causative factors (Eriksson and Sandgren, 1999). Considering the large size and geologically complex catchment of Mandovi River, a baseline information on changes in the sediment sources, composition, their relative contributions, and dispersal system is prerequisite to identify the erosional sites as well as to determine the relative contribution of soils from eroding riverbanks and catchment areas to the total estuarine sediment load.

Mandovi estuary is a tropical, transitional, mesotidal, and monsoon fed estuarine system located on the central west coast of India (Figure 1), where runoff, rainfall, tides, and salinity play a dominant role in determining its sedimentary environment compared to the other estuaries in different parts of the world (Shetye et al., 2007; Vijith et al., 2009; Rao et al., 2011). The large size of the catchment and drainage basinal area, rapid erosion of hinterland dominated by a complex geological terrains, and seasonal runoff provide a permanent source of magnetic minerals to the Mandovi estuary in varying proportion and wide range of grain sizes. Hence the fluvial sediments from the Mandovi estuary carry distinct magnetic signatures of their respective provenance and underlying hydro-and sediment dynamics. Hence the magnetic properties can be used as a potential tracer to investigate the fate of detrital sediments in the Mandovi estuary from a source-to-sink perspective. Sediments in the Mandovi estuary typically contain high concentration of iron (ferri-and antiferromagnetic) minerals and have been extensively studied using magnetic and geochemical methods to resolve range of scientific questions for example, to delineate the sediment provenance and diagenetic history (Rao et

al., 2015; Prajith et al., 2015, 2016), characterize the depositional environment (Singh et al., 2014; Nayak and D'Mello, 2018), and monitor anthropogenic activities (Fernandes et al., 2009; Kessarkar et al., 2015; Veerasingam et al., 2015). However, a dedicated magnetomineralogical based study elucidating the control of source-to-sink processes on the dynamics (sorting, dispersal, settling) of magnetic particles in the Mandovi estuary was so far lacking and is crucial for the development of magnetic-based environmental proxies. In addition, compilation of magnetic properties of source rocks, fluvial, estuarine, and nearshore sediments is prerequisite to gain fundamental information on the provenance changes and underlying hydro-and sediment dynamic regime in the Mandovi estuary, which remain poorly constrained. The research questions addressed in this study will enhance the interpretative value of existing magnetic mineralogy and granulometry based proxies which are sensitive to geological and environmental conditions.

The aims of this study was to address three key questions:

- (i) Elucidate how differential detrital input (source rocks, fluvial, riverbank) and hydrodynamics (runoff, tides, currents) control the mixing, sorting, dispersal, and settling of heavy (magnetic) particles during their transit from source-to-sink in a mesotidal tropical estuarine system.
- (ii) Which sub-environment of the estuarine system preferentially favour the accumulation of heavy (magnetic) minerals?
- (iii) Test the relationship between rock magnetic concentration and grain-size dependent proxies and clastic particle sizes in a different sub-environments of the estuary.

## **2. Study Area**

### **2.1. Geography and hydro-and sediment dynamics of the Mandovi estuary**

Mandovi estuary located on the central west coast of India is a tropical, monsoonal, and mesotidal estuarine system (Figure 1). The tidal range in the estuary vary between ~2.3 m and 1.5 m during the spring and neap tides, respectively (Shetye et al., 2007; Manoj and Unnikrishnan, 2009). The Mandovi River is the major river system and has a length of 75 km and covers a drainage basinal area of ~1580 km<sup>2</sup>. The width of the Mandovi River at the tidal inlet (Aguada Bay) is ~ 4 km, and the mean depth is 5 m. Mandovi estuary is well-connected to Arabian sea through a tidal inlet (Aguada Bay) and to the proximal Zuari estuary via Cumbharjua Canal. Mandovi River is joined by a large network of many tributaries including Khandepar, Nerul, Mapusa, Valvanti, Bicholim, Ragada, Kotrachi Nadi, Udnai, Assonora, St Inez Creek, and Rio de Ourém that delivers heterogenous sediments to the Mandovi estuary.

Mandovi River is dominantly monsoon rainfed and the rainfall over the drainage basins vary considerably in both time and space. Annual rainfall at the catchment area of the Mandovi River is about 660 cm and 290 cm respectively (Shetye et al., 2007). Mandovi estuary receives enormous amount of fluvial sediment load through large river runoff ( $\sim 258 \text{ m}^3\text{s}^{-1}$ ) generated by intense southwest monsoon (June to September) (Vijith et al., 2009). Fluvial sediments get efficiently transported through different parts of the Mandovi estuary and finally gets accumulated at the mouth of tidal inlet near Aguada Bay to form sandbars (Qasim and Sen Gupta, 1981). During non-monsoon season, sediment dynamics within the Mandovi estuary is mainly driven by tide and wind-induced currents. As a consequence, the salinity intrusion extend upto  $\sim 45$  km in the upper reaches from the mouth of tidal inlet (Aguada Bay) (Sundar and Shetye, 2005; Shetye et al., 2007; Manoj and Unnikrishnan, 2009). The sediment dynamics within the Mandovi estuary are mainly governed by tidal currents (till the commencement of the southwest monsoon), while wave action is limited to the tidal inlet i.e., at Aguada Bay.

## **2.2. Catchment geology**

The geology of the Mandovi catchment is diverse (Figure 1; Gokul et al., 1985). Mandovi River has a catchment area of about  $\sim 1580 \text{ km}^2$  and mainly drains through complex geological terrains of the Goa Group belonging to the Dharwar Super Group of the Archaean-Proterozoic age (Figure 1; Gokul et al., 1985). The Goa Group occupies a large area in the upstream of Mandovi River and mainly comprised of banded-iron and manganese formations, quartzite, meta-conglomerate/breccias, metagreywacke, quartz-chlorite-biotite-amphibole schist, and carbonate quartz-chlorite schist (Dessai et al., 2009). Banded iron formation (BIF) horizons are formed with upper (magnetite-rich) and lower (hematite-rich) quartz/chert. The catchment rocks are exposed to intense chemical weathering under humid, tropical climate conditions and overlain by thick lateritic profiles, and soils along the riverbank (Figure 1; Rao et al., 2015). Vegetation cover, land use pattern, hinterland and riverbank erosion, and monsoonal changes has a significant impact on the provenance and amount of sediments entering the Mandovi River system. The catchment area of Mandovi River is dominated by grazing lands, with few areas of the floodplain are extensively utilized for agriculture. Erosion and transport of surface and subsoils from the hinterland, floodplains, and adjacent river banks into the Mandovi River during monsoon results in the formation of thick plume of suspended sediments (Pathak et al., 1988; Kessarkar et al., 2010, 2013, 2015; Rao et al., 2011; Ibrampurkar, 2012; Reddy et al., 2016; Renjan et al., 2017; Fernandes et al., 2018). Contrasting catchment lithology, differential sediment input by the tributaries, intense monsoonal conditions, and erosional processes in the hinterland and riverbank

provide high compositional (concentration, grain size, mineralogy) diversity in the bulk sediment magnetic mineral assemblages, which gets well-recorded in the Mandovi estuary sediments.

### **3. Materials and Methods**

#### **3.1. Rock, soil, and sediment sampling survey**

As part of a larger sediment sampling program in the Mandovi estuary, the surficial (riverine, and nearshore yielding 177 nos) sediment samples were collected during normal weather conditions using a Van Veen grab sampler (uppermost 1-5 cm) on a motorized fishing trawler in March 2022 (Figure 1; Figure S1c and S1f). Representative rock samples (114 nos) were collected from the catchment areas of Mandovi River during two field campaigns in 2021 and 2022 (Figure 1; Figures S1a and S1b). Soil samples (vertical profile RB-01, at 5 cm interval yielding 39 nos) were collected along an eroding riverbank site in the middle reaches of Mandovi River (Figures S1d and S1g). In addition, we collected three subsurface soil profiles (SP-1, SP-2, and SP-3 yielding 103 nos) from the Mandovi River catchment area and surficial (16) sediment samples from the Ragada River (Figure 1). The sediment sampling design considered in this study has the limitation as the sample collection sites along the Mandovi estuary are irregularly spaced.

##### **3.1.1 Preparation of Maps, Data Interpolation, and Potential Limitations**

Spatial maps presenting the distribution of magnetic mineral (Figure 2) and total organic carbon (Figure 9) content were generated using the inverse distance weighted (IDW) interpolation method within the QGIS software. This approach utilizes known values associated with vector points to predict values at unknown locations, resulting in a continuous raster surface. In the IDW method, each sample point is assigned a weight determined by a coefficient, and this weight decreases as the distance from the unknown point increases. Higher coefficients diminish the influence of distant points, causing the estimated value at the unknown location to converge toward that of the nearest observed point. However, it is important to note that the IDW method does possess certain limitations. Particularly, when sample points are distributed in an extremely uneven manner, the quality of the interpolation can be compromised, resulting in less accurate outcomes. Additionally, the interpolated maximum and minimum values of surfaces are intrinsically linked to the positions of the sample data points. Consequently, this linkage often results in the formation of minor peaks and depressions in the vicinity of these sample points.

## 3.2. Laboratory methods

### 3.2.1. Rock Magnetism

For rock magnetic measurements, sediment, soil, and rock samples were dried at 40°C, weighed, and packed in 25-mm plastic cylindrical sample bottles. Frequency-dependent magnetic susceptibility measurements were conducted using the Bartington MS2B dual frequency ( $\chi_{lf} = 0.47$  kHz and  $\chi_{hf} = 4.7$  kHz) susceptibility meter. Raw volume specific susceptibility values  $k$  of the samples were converted to respective mass specific susceptibility by multiplying the volumetric susceptibility with the nominal value and then dividing the bulk signal by the weight of the sample. Anhyseretic remanent magnetization (ARM) was applied using 100 mT AF field superimposed with a fixed DC bias field of 50 mT. Isothermal remanent magnetization (IRM) of 700 mT, 1000 mT, 2000 mT, and 2500 mT in the forward direction and -20mT, -30mT, -100mT, and -300 mT in the backward direction was imparted using MMPM10 pulse magnetizer. All remanence were measured using AGICO automatic JR-6A spinner magnetometer housed at the paleomagnetic laboratory of CSIR-National Institute of Oceanography (CSIR-NIO), Goa, India. Mass-normalized IRM acquired with a peak field of 2500 mT is considered to be the saturation isothermal remanent magnetization (SIRM). S ratio is calculated as the ratio between the IRM at -300 mT and SIRM ( $IRM_{-300mT}/SIRM$ ; Thompson and Oldfield, 1986). The hard isothermal remanent magnetization (HIRM) is estimated as  $0.5 \times (SIRM + IRM_{-300mT})$ , where  $IRM_{-300mT}$  represents the remanent magnetization obtained by first saturating the sample in a high field (e.g., 2.5 T), and then applying a back-field of -300 mT to reverse the SIRM (Thompson and Oldfield, 1986). Specialized magnetic measurements including temperature-dependent magnetization curves and the hysteresis loop analyses were carried out on twenty six bulk sediment samples using Advanced Variable Field Translation Balance (AVFTB) at the paleomagnetic laboratory of CSIR-National Geophysical Research Institute (CSIR-NGRI), Hyderabad, India. The magnetite content in the bulk sediments was calculated by dividing bulk magnetic mass susceptibility ( $\chi$ ) with the mineral-specific value for pure multi-domain (MD) magnetite  $\chi_{Mag} \sim 660 \times 10^{-6} \text{ m}^3 \text{ kg}^{-1}$  following Maher (1988).

Hysteresis loops were measured for a set of samples (n=26) at a constant ambient temperature (33°C) on AVFTB. Magnetization was measured in the applied magnetic field which ranges from -800 mT to +800 mT. The hysteresis shape parameter ( $\sigma_{hyst}$ ) was estimated to identify the distinct mixtures of magnetic particle sizes (Fabian, 2003). Strong hysteresis shape anomalies were produced by the mixtures of magnetic minerals with contrasting coercivities that are a function of magnetic domain sizes (Fabian, 2003). A superparamagnetic (SP) particle admixture generates wasp-waisted shaped hysteresis loops ( $\sigma_{hyst}>0$ ), whereas a single domain (SD) - multidomain (MD) mixing produces the

loops exhibiting pot-bellied shape ( $\sigma_{\text{hyst}} < 0$ ) (Fabian, 2003). The hysteresis shape parameter ( $\sigma_{\text{hyst}}$ ) was calculated using following equation:

$$\sigma_{\text{hyst}} = \log \left( \frac{E_{\text{hyst}}}{4M_s B_c} \right)$$

$E_{\text{hyst}}$  : total hysteresis area calculated from the difference between upper and lower branch areas of the hysteresis loop

$M_s$ : represent the saturation magnetization

$B_c$  : coercive force (coercivity)

The IRM curves serve as a diagnostic tool to identify the magnetic mineralogies in the marine sediments (Kruiver et al., 2001). Magnetic fields were imparted on selected samples representing different reaches of the Mandovi River in a stepwise manner (total fields applied: 45) starting from 10 mT to 2500 mT, and subsequently remanent magnetization was measured. We utilized Max UnMix program (Maxbauer et al., 2016) for the identification of suitable magnetic components. This program utilizes skew-normal density functions to fit the identified components into original IRM curve (Kruiver et al., 2001; Maxbauer et al., 2016). The magnetic components are characterized by 1) their contribution to the total magnetic remanence in percentage (C%), 2) the spread of the distribution measured by the dispersion parameter (DP) (one standard deviation on log scale), and 3) the mean coercivity ( $B_{1/2}$ ).

### 3.2.2. Magnetogranulometry

Representative estuarine sediment samples (upper, middle, lower, and nearshore zone) were selected for magnetogranulometry. Bulk samples of 120 g were dried, weighed, suspended and wet-sieved for 25 min with an automated sieve shaker (Fritsch Analysette 3) creating the six sieve fractions <20  $\mu\text{m}$ , 20-32  $\mu\text{m}$ , 32-40  $\mu\text{m}$ , 40-45  $\mu\text{m}$ , 45-63  $\mu\text{m}$ , and >63  $\mu\text{m}$ . The particle assemblages obtained in each sieved fraction were then subjected to the volume specific magnetic susceptibility and magnetic remanence measurements following the standard protocol mentioned in section 3.2.1.

### 3.2.3. Grain Size Analyses

Clastic grain-size distributions of all surficial riverine sediments samples were measured with the laser diffraction particle sizer analyzer (Beckman Coulter LS 13320) housed at National Centre for Polar and Ocean Research (NCPOR), Goa, India. Sediment samples were first desalinated prior to analysis, and were further decarbonated using dilute HCl (1 N). Sediment suspensions were treated with 10%  $\text{H}_2\text{O}_2$  to remove organic carbon, and 300 mg of dispersing agent (sodium



hexametaphosphate) was added to the suspension, which was then ultrasonicated before the analysis. Grain size values are represented as volume %.

### 3.2.4. Sediment Geochemistry

The total carbon (TC) content was determined on bulk powdered sediment samples following a flash dynamic combustion method using the Thermo Scientific FLASH 2000 Series Nitrogen and Carbon (CN) analyzer housed at CSIR-NIO, Goa, India. The total inorganic carbon (TIC) content was measured using UIC CM 5014 Coulometer. The total organic carbon (TOC) content was calculated by subtracting the TIC from TC.

### 3.2.5. Electron Microscopy

The magnetic particles were separated from the bulk sediment samples using the extraction method described in Petersen et al. (1986). Images of the magnetic particles were captured using a scanning electron microscope (JEOL JSM-5800 LV) in a secondary electron (SE) imaging mode (Energy levels: 5 and 20 keV). Elemental compositions was determined by the energy dispersive X-ray spectroscopy (EDS) probe attached to the microscope.

## 4. Results

### 4.1. Rock Magnetic and Grain size Properties of Surficial Sediments and Soil Profiles

We demarcate the magnetic profile of the Mandovi estuary into four different sedimentary zones (upper, middle, lower, and nearshore; Figure 1 and Figure 3) mainly based on the variations in the salinity and estuarine turbidity maximum (Shetye and Murty, 1987; Manoj and Unnikrishnan, 2007; Shetye et al., 2007; Kessarkar et al., 2010; Rao et al., 2011; Shynu et al., 2015; Prajith et al., 2016). Environmental magnetism data of surficial sediments within Mandovi estuary show a high variability in terms of magnetite concentration ( $\chi_{lf}$ ), grain size (ARM/IRM), and mineralogy (S-ratio) (Figure 2 and Figure 3). Several discrete zones of magnetite accumulation are identified throughout the estuary based on the magnetic susceptibility ( $\chi_{lf}$ ) data of the surficial sediments (Figure 2). Overall, the  $\chi_{lf}$  vary over three orders of magnitude from 0.0957 to  $26.317 \times 10^{-6} \text{ m}^3 \text{ kg}^{-1}$  (~0.0145 to 3.987 wt % equivalent magnetite content; Figure 3a; Table 1), with highest values found within upper zone of the estuary (Figure 3a). Magnetic grain size (ARM/IRM) ratio vary from 0.0019 to 0.062 (Figure 3b; Table 1). Higher magnetic susceptibility values are exhibited by the coarser magnetic grain sizes and are found to be concentrated in the upper and lower zone of the Mandovi estuary (Figures 3a and 3b). A general trend of increase in magnetic susceptibility (Figure 3a) manifested by fining of magnetic grain size (as indicated by increasing ARM/SIRM ratio; Figure 3b) is clearly seen and extend upto the Nerul River of the Mandovi estuary. In contrast, the samples from lower zone (Coco Bay to Reis

Magos) are dominated by high and relatively stable concentration of coarser ferrimagnetic particles (Figure 3a and Figure 3b). Magnetic mineralogy diagnostic proxy (S-ratio) show a large variation in the values ranging between 0.12 to 0.99 and indicate the presence of ferri- and antiferromagnetic minerals in the studied samples (Figure 3c; Table 1). A distinct drop in S-ratio within each zone suggest the increase input of antiferromagnetic relative to ferrimagnetic minerals (Figure 3c). HIRM (Figure 3d) profile closely mimic S-ratio (Figure 3c) trend mostly in all samples except Nerul River. Highest values of HIRM are found in the Cumbharjua Canal samples indicative of increase in the concentration of high coercive (hematite, goethite) minerals (Figure 3d). A magnetic proxy ( $SIRM/\chi_{lf}$ ; Figure 3e) for ferrimagnetic iron sulfides show higher values only in the upper zone (Khandepar River) of the Mandovi estuary (Snowball and Thompson, 1990; Peters and Dekkers, 2003). The median bulk sediment grain size data exhibit broad distribution (Figure 3f) varying from very fine silt (median = 6.20  $\mu\text{m}$ ) to coarse sand (median = 382.35  $\mu\text{m}$ ), and did not exhibit any distinct trends throughout the Mandovi estuary (Table 1). The sediments from the upper and nearshore zone show close resemblance interm of grain size distribution and are mainly dominated by very fine to medium silt (Figure 3e).

The rock magnetic record of riverbank soil profile (RB-01) is presented in Figure 4a-e. A gradual downcore rise in  $\chi_{lf}$  and SIRM suggest increase in magnetic mineral concentration (Figures 4a and 4c). Magnetic grain size diagnostic parameters ( $\chi_{fd\%}$ , ARM/SIRM) mimic each other and exhibit trend of downward decline which indicate the dominance of coarser magnetic particles in the uppermost 80 cm (Figures 4b and 4d). Higher  $\chi_{fd\%}$ , ARM/SIRM, and lowest  $\chi_{lf}$  values from 80 to 140 cm indicate the presence of fine-grained magnetic particles in low concentration (Figures 4a-4c). S-ratio profile (Figure 4e) reveal a general trend of slight downward decline indicating the increase in the concentration of high coercive minerals. The rock magnetic records of the soil profiles (SP-1, SP-2, SP-3; Figures 4f-4t) covering abundant rocks of the Mandovi River catchment show relatively different characteristics interm of magnetic mineral concentration and grain size (Figure 4f-4t). Magnetic susceptibility is relatively low in SP-1, SP-2, SP-3 samples compared to RB-01, and show large variations in magnetic grain size (Figures 4g, 4i, 4l, 4n, 4q, and 4s).

## **4.2. Magnetic mineralogical and granulometric proxies**

### **4.2.1. Temperature-dependent magnetic susceptibility ( $\chi$ -T)**

$\chi$ -T curves of representative samples from all sedimentary zones of the estuary are presented in Figure 5 and Figure S6. A noticeable drop in  $\chi$  values between 540 °C and 650 °C in majority of the samples suggest that the bulk magnetic mineralogy is dominated by ferri-(magnetite, titanomagnetite) and antiferromagnetic (titanohematite) particles (Figure 5 and Figure S6; Dunlop et al., 1997; Dunlop

and Özdemir, 1997; Liu et al., 2012). A minor  $\chi$  rise between 352 °C to 452 °C in few samples (Figure 5 and Figure S6) is attributed to the dominance of titanomagnetite exhibiting wide range of Ti-contents (Lattard et al., 2006), transformation of maghemite into magnetite (Gehring et al., 2009), or due to conversion of paramagnetic minerals and iron-containing silicates to magnetite during heating process (Hirt and Gehring, 1991; Hirt et al., 1993; Passier et al., 2001; Liu et al., 2005; Philips, 2018), or transformation of iron-containing silicates to magnetite.

#### **4.2.2. Magnetogranulometric Parameters**

Magnetic susceptibility and remanence data of sieved fractions provide an opportunity to elucidate the possible link between estuarine sedimentary processes and the rockmagnetic proxies. A general trend of increase in magnetic grain size with increasing physical grain size is observed in most of the samples representing different estuarine zones (Figure S3, Figure S4, and Figure S5). A systematic pattern of S-ratio variation in different sieved fraction representing different estuarine zones is noticed. Highest magnetic susceptibility values followed by lowest ARM/SIRM in the mid-range sieve fraction (45 - 63  $\mu\text{m}$ ) suggests that isolated monocrystalline magnetite (coarse-silt sized) particles mainly exist and get enrich within this grain-size range. Lowest values of S-ratio are found in the <20  $\mu\text{m}$  (upper zone), 32-40  $\mu\text{m}$  (middle zone), and >63  $\mu\text{m}$  (lower zone) fractions (Figure S3r, Figure S4r, and Figure S5r).

#### **4.2.3. Hysteresis loops and IRM Acquisition Curves**

The concentration of magnetic particles in the sediments varied significantly throughout the Mandovi River (Ms: 0.85 to 72.2  $\text{Am}^2/\text{kg}$ ) (Figure 6 and Figure S7). The magnetic mineral content is highest in the upper zone of the Mandovi River (Ms: 0.9 to 72.2  $\text{Am}^2/\text{kg}$ ) (Figure 6 and Figure S7) and decrease further from the middle (Ms: 0.89-36.2  $\text{Am}^2/\text{kg}$ ) to lower (Ms: 0.85-13.7  $\text{Am}^2/\text{kg}$ ) zone. A noticeable rise in magnetic mineral concentration (Ms: 31.8-34.4  $\text{Am}^2/\text{kg}$ ) is observed in the nearshore samples (Figure 6 and Figure S7). Majority of the hysteresis curves remain unsaturated up to 400 mT, indicating the presence of high coercivity magnetic mineral phases (background) (Figure 6 and Figure S7). For instance, the samples from the upper zone (Figure S7e) show the saturation magnetization of 67.5  $\text{Am}^2/\text{kg}$  indicating the dominance of ferrimagnetic mineral phase. However, the curve did not show saturation upto 800 mT (Figure S7e). The samples from middle zone of the Mandovi River exhibited high coercivities (upto 22.74 mT), in contrast with lower values in the nearshore sample (upto 15.21 mT) (Figure 6 and Figure S7).

Variations in  $\sigma_{\text{hyst}}$  and the wide and narrow shape distribution of the hysteresis curves provide compelling evidence on the presence of mixed size magnetic grains in all sedimentary zones of the

Mandovi estuary (Figure 6). The wasp-waisted curves in the sediment samples manifested by large drops in S-ratio ( $< 0.6$ ; Figure 3c) suggest increased contribution from the high coercivity antiferromagnetic minerals (Figures 6c-6e; Roberts et al., 1995; Tauxe et al., 1996). For instance, the sample from the middle zone reveal  $\sigma_{\text{hyst}} = 0.96$ , a relatively high value that indicate the presence of SP grains in substantial proportion (Figure 6c). Comparatively, the pot-bellied curves suggest the presence of single domain (SD), pseudosingle domain (PSD), and multi-domain (MD) type magnetic grains (Figures 6a, Figure 6b, and Figure 6f; Figure S7). We examine a Neel plot of squareness ( $M_{\text{rs}}/M_{\text{s}}$ ) versus coercive field ( $B_{\text{c}}$ ) to further clarify the domain states (Figure S8; Néel, 1955; Tauxe et al., 2002). The ratio  $M_{\text{rs}}/M_{\text{s}}$ ,  $B_{\text{c}}$ , and  $B_{\text{cr}}$  are all sensitive to the magnetic grain (domain) sizes. Majority of the analyzed samples clustered in a triangular zone indicative of mixing of uniaxial single domain (USD) and superparamagnetic (SP) grains. The presence of MD grains lowered the coercivity and remanence ratio as seen through lower  $M_{\text{rs}}/M_{\text{s}}$  and  $B_{\text{c}}$  values (Figure S8; Tauxe et al., 2002). Nearshore samples show more affinity with the samples falling in the MD group indicating the predominance of coarse magnetic grain sizes (Figure S8; Tauxe et al., 2002). Samples with low S-ratio (Figure 3c) exhibit high values of remanence ratio (Figure S8) suggesting the presence of SP (antiferromagnetic) and SD (ferrimagnetic) (Dunlop, 2002; Tauxe et al., 2002). Overall, SD-PSD-MD mixing is observed in majority of the samples (Figure S8), while SP magnetic particles re visible in the hysteresis curves (Figures 6c-6e) and Neel plot (Figure S8) only in few samples which contain relatively higher proportion of SP particles (Tauxe et al., 2002).

Three coercivity components are identified to be optimum for fitting the shape of coercivity distribution of IRM acquisition curves (Figure 7; Table 2). The summary of the data from the analysis of IRM curves is provided in the Table 2. The coercivity of Component 1 varied between 17 - 26 mT and its contribution to the remanence remained significant (29 - 55 %) in the samples from the upper and middle zone of the Mandovi estuary (Table 2). Drastic reduction in the contribution (Average = 11 %) of Component 1 is clearly visible in the samples representing the lower zone of the Mandovi estuary (Figure 7; Table 2). The dispersion parameter of Component 1 show large variations throughout the samples (0.23 - 0.47) indicating broad grain size distribution (Figure 7; Table 2; Maxbauer et al., 2016). Hence, Component 1 could be related to the fine-grained magnetite with grain sizes ranging between the SP and SD grains (Egli, 2004; Just et al., 2012).

Component 2 is a major contributor to the remanence in all samples of the Mandovi River and display higher coercivities (47 - 64 mT) as compared to Component 1 (Figure 7). Noticeably, the contribution of Component 2 is 2/3<sup>rd</sup> in the nearshore sample (Figure 7; Table 2). The dispersion parameter for Component 2 show less variability compared to other components (Average = 0.27)

suggesting the limited variation in grain size (Figure 7; Table 2). These properties represent typical characteristics of finer titanomagnetite grains (Egli, 2004; Garming et al., 2005; Just et al., 2012). Component 3 exhibit very high coercivity (515-838 mT) and reveal broad dispersion (0.24 - 0.36) (Figure 7; Table 2). The SIRM contribution of Component 3 in the upper, middle, and nearshore zone samples remained minimal (Average = 14.4%) compared to the lower zone samples of the Mandovi River (Average = 53%) (Figure 7; Table 2). The coercivity of high orders in Component 3 is a characteristic feature of highly coercive (titano-) hematite or goethite (from stoichiometric to Al-substituted) (Peters and Dekkers, 2003; Egli, 2004; Just et al., 2012).

### **4.3. Grain Size Distributions and Organic Carbon Content**

Variation in the sediment input and hydrodynamic regime within different parts of the Mandovi estuary could create a complex sediment grain size distribution pattern. We analyze the grain size distribution curves of samples representing all sedimentary zones of the Mandovi estuary (Figures 8a-8d). A clear trend of loss of fluvially supplied very fine silt size sediment fraction from source-to-sink (upper to lower zone) is evident (Figures 8a - 8c). Upper zone samples exhibit large variation in mean grain size ranging from 10.85  $\mu\text{m}$  to 390.89  $\mu\text{m}$  (Table 1). Two distinct patterns in the grain size distribution are noticed (Figures 8a - 8c). The finer grain size window is dominated by silt (mode 7.0  $\mu\text{m}$  to 19.76  $\mu\text{m}$ ; Figure 8a) and the coarser one by sand size (mode 168.87  $\mu\text{m}$  to 471.14  $\mu\text{m}$ ; Figure 8a) grains. Within the middle zone, we observe well-sorted sediments exhibiting broad and unimodal type grain size distributions with mean grain size ranging between 54.77  $\mu\text{m}$  to 382.35  $\mu\text{m}$  (Figure 8b and Table 1). Samples from the lower zone show remarkably well-sorted unimodal fine (mode 87.90  $\mu\text{m}$  to 517.20  $\mu\text{m}$ ; Figure 8c; Table 1) and coarse (mode 269.15  $\mu\text{m}$ ; Figure 8c) sand fraction. Nearshore samples show the dominance of silt size grains (mode 14.4  $\mu\text{m}$  to 31.51  $\mu\text{m}$ ; Figure 8d; Table 1) and exhibit the distribution pattern (Figure 8d) which is similar with the finer grain size window of the upper zone (Figure 8a).

The surficial TOC distribution map of Mandovi estuarine sediment is presented in Figure 9. Overall, the TOC content vary between 0.1 % to 3.0 % (Figure 9). We did not observe any specific trends in the TOC distribution throughout the estuary. High TOC values are noticed in the Cumbharjua Canal (2.65 %), Mapusa River (3.0 %), Coco Bay (2.2%), Nerul River (1.8 %), and nearshore region ( 1.72).

### **4.5. Mineralogical Analyses of Magnetic Particles**

SEM-EDS analyses of magnetic particles augment our magnetic observations and further confirm that magnetite, titanomagnetite, and hematite are the major magnetic minerals present in the

analyzed sediments (Figure 10; Figures S9 and S10). Detrital origin ferrimagnetic (magnetite, titanomagnetite) and antiferromagnetic (hematite, goethite) minerals of various sizes and shapes are identified in all sediment magnetic zones (Figure 10; Figures S9 and S10). Octahedral magnetite grains exhibiting well-preserved morphology are abundant and occur throughout all zones (Figures 10a and 10g; Figures S9c and S9h, Figures S10c and S10f). Few grains exhibit the signature of pervasive low-temperature oxidation of detrital magnetic particles including surficial etching, pits, skeleton lamellae, and distinct shrinkage cracks (Figures 10b, 10e, 10g, and 10h; Figure S9b; Figures S10f – S10h). Hematite (amorphous and crystalline; Figure 10c, 10e, and 10h; Figure S9b, Figure S10g) and goethite (Figures 10d and 10f) occur as individual fine-grained particles and found predominantly in the upper and middle zone of the Mandovi estuary.

#### **4.5. Relationship Between Magnetic Properties of Catchment Rocks, Catchment Soil, Riverbank Soil, and Estuarine Sediments**

Bivariate plots are used to evaluate the relationship between magnetic mineral concentration, grain size, and mineralogy (Figure 11). Overall, the magnetic susceptibility in the studied samples vary over three orders of magnitude from  $0.0957$  to  $26.317 \times 10^{-6} \text{m}^3 \text{kg}^{-1}$  ( $\sim 0.0145$  to  $3.987$  wt % equivalent magnetite content; Figure 3a; Table 1). The catchment rock samples show large scattering interm of concentration, grain size, and mineralogy of magnetic particles, while estuarine sediments, riverbank and catchment soil samples show better grouping (Figures 11a-11e). A good covariation between  $\chi_{lf}$ , ARM, and SIRM is observed in most of the samples (except catchment rocks), but show wide range in their values due to the presence of different types and occurrences of magnetic minerals (Figures 11a and 11b). A good relationship exist between magnetic susceptibility and magnetic grain size in majority of the estuarine, catchment, and riverbank soil samples, with highest values being dominated by coarse-grained magnetic particles and vice versa (Figure 11c). Poor correlation ( $R^2 = 0.007$ ; Figure 11f) between TOC content and magnetic susceptibility is observed.

### **5. Discussion**

Magnetomineralogical results complemented by grain size, mineralogy, and electron microscopy data revealed the spatial distribution and composition of magnetic minerals within and off Mandovi estuary is controlled by the complex interplay between detrital sedimentation, differential (mineral density and grain size) selective settling mechanisms and dispersal system driven by underlying hydro and sediment dynamics. We now discuss how magnetic minerals respond to changing hydro-and sediment dynamics in the different subsystems of the estuary.

## **5.1. Control of Source-to-Sink Processes on the Dispersal, Sorting, and Depositional Dynamics of Magnetic Particles**

### **5.1.1. Sources of Magnetic Minerals**

Geological terrains dominated by igneous, sedimentary, and metamorphic rocks in the catchment area provided a high supply of low and high coercivity magnetic minerals (wide range in concentration and grain sizes; Figure 2, Figure 3, Figure 10, Figure 11, Figure S9, Figure S10) to the Mandovi estuary (Gokul et al., 1985; Dhoundial et al., 1987; Dessai et al., 2009; Mascarenhas and Kalavampara, 2009; Rao et al., 2015). Rock magnetic, temperature-dependent magnetization curves, and SEM-EDS data revealed that magnetite, titanomagnetite, (titanio) hematite, and goethite are the major magnetic minerals in the studied samples and largely contribute to the bulk sediment magnetism (Figure 3, Figure 5, Figure 6, Figure 7, Figure 10, Figure S6, Figure S7, Figure S9, Figure S10).

Rock magnetic data display considerable variation in terms of magnetite content, mineralogy, and grain size (Figure 2, Figure 3; Table 1) which can be mainly attributed to the differential sediment inputs by inflowing rivers, and changing hydrodynamics conditions in different sedimentary zones. A clear trend of increase in magnetite concentration followed by the progressive fining of magnetic grain size in upper and middle and lack of such pattern in lower zone of the Mandovi estuary can be very well linked with the differential sorting and transport mechanism prevailing in each sub-environment. Such processes mainly constrained the dynamics of magnetic particles during their transit from source-to-sink (Figures 3a and 3b). Few surface sediment samples from each sediment magnetic zone show remarkably high ARM/SIRM values (Figure 3b) and plot mostly within USD region (Figure S8) indicating the presence of fine magnetic particles.

### **5.1.2. Mineral Density versus Grain Size Selective Sorting and Settling of Magnetic Particles in an Estuarine System**

Comparison of rock magnetic, grain size, and mineralogical data of catchment rocks against the pedogenic soils, estuarine, and nearshore bedload sediments help to quantify the magnetite concentration by grain size and mineral-selective processes (Figure 11). Magnetic particles in the studied sediments exhibit wide range in grain sizes (ARM/SIRM = 0.0019 to 0.062; Figure 3b) and the equivalent magnetite content range between 0.014 to 3.987 wt % (Table 1). This suggest that enhancement and depletion of magnetic particles occurred during their transport from source-to-sink.

Relationship between magnetite content and magnetic grain size with clastic grain size is poor (Figure S11). Lack of good correlation between mineral-density ( $\chi_{lf}$ ) and grain size (ARM/SIRM) selective sediment sorting mechanism suggest that these processes are largely disjunct (Figures S11a

and S11b). Poor covariation between magnetic susceptibility and clastic sediment grain size (Figure S11a) reveal relative density differences between magnetic and non-magnetic fraction and further suggest that most of the magnetic particles are discrete. Biplot of magnetic grain size versus median grain size showed narrow range in magnetic grain size, but large scattering in the clastic sediment grain size distribution (Figure S11b). This could be explained by the fact that magnetic particles that are transported along with quartz grains showed relatively large variation in the grain size due to the density-driven hydraulic differences. Successive mixing, and sorting of magnetic particles by the river runoff, tidal, and wave induced bottom currents is clearly reflected by the elongated and relatively narrow grouping of the estuarine bedload samples from all sedimentary zones of the estuary (Figures 11a-11c). A better grouping and consistent trend, but large scattering in terms of magnetite content and grain size in all estuarine bedload samples (Figures 11a-11c) is attributed to the heterogeneity of the catchment lithologies (source rocks, soils), and changing hydrodynamics which produce compositional diversity in the studied sediments.

Majority of the samples from the upper zone of the Mandovi estuary possess higher values of  $\chi_{lf}$  manifested by coarser magnetic grains (Figure 3a and Figure 3b). It is highly possible that low river runoff prior to commencement of monsoon in the upper zone of the estuary might have favoured mineral-density selective entrainment, which resulted in the rapid burial of magnetic particles. Such mechanism most likely led to the accumulation of coarser magnetic particles close to the source area. Laboratory experiments by Gallaway et al. (2012) demonstrated that the burial of magnetic grain was most effective beneath coarser rather than finer non-magnetic sand under less energetic conditions. We hypothesize that most of the coarser magnetic particles from the upper zone gets readily flushed out only during heavy rainfall. Under such condition, high river runoff enhance the erosion and transport of large amount of the bedload and suspended sediments from the catchment to the other downstream areas of the Mandovi estuary. The observed trend of increase in  $\chi_{lf}$  (Figure 3a) followed by the progressive fining of magnetic grain size (Figure 3b), and subsequently selective loss of coarse ferrimagnetic grains in the upper and middle zone could also be attributed to the decrease in energy of fluvial transport. A similar trend of fining trend of ferrimagnetic minerals from source-to-sink region was observed in the Yangtze River sediments (Li et al., 2012), Tauranga Harbour, New Zealand (Badesab et al., 2017), South China Sea (Wang et al., 2014), and Red River (Nguyen et al., 2016). Majority of the samples from the lower zone of the estuary are marked by higher concentration of coarser ferrimagnetic magnetic particles as evident in  $\chi_{lf}$ , ARM/SIRM, and S-ratio profile (Figures 3a - 3c). Absence of such fining trend in the lower zone (Coco Bay, Aguada-Miramar transect ; Figure 3b) of the Mandovi estuary could be attributed to the deeper water depth, strong tidal exchange, and



bottom currents which efficiently facilitated mixing and sorting resulting in preferential enhancement of coarser (heavy) ferrimagnetic particles as seen through elevated values of magnetic susceptibility (Figures 3a - 3c). Such findings demonstrate the sensitivity of the magnetic methods to detect even subtle variations in the magnetic grain size.

Relative loss of finer magnetite particles (Figure 3b) and clastic sediments (mode 7.08  $\mu\text{m}$  to 19.76  $\mu\text{m}$ , Figures 8a - 8c; Table 1) followed by subsequent enhancement of coarser magnetic grains (Figure 3b) especially in the lower zone is attributed to the high-energy driven sediment partitioning regime which inhibit the permanent settling of  $< 19.76 \mu\text{m}$  fluvial sediment fraction and are therefore frequently flushed out to the sea. The tidal channel network within the lower zone provided wide-range of bottom current velocities which is clearly evident in a broad range of clastic grain size distribution (mode 7.08 to 517.2  $\mu\text{m}$ ; Table 1) and magnetite content (equivalent magnetite content range between 0.014 to 0.64 wt %; Table 1). Nearshore samples exhibit high concentration of coarser magnetic particles and finer clastic grain size (mode size  $< 31.50 \mu\text{m}$ ; Figure 3a and Figure 3e; Table 1). Close resemblance between lower and nearshore samples in terms of magnetite content (Figure 3a) and grain size (Figure 3b) can be directly linked to the higher sediment exchange between these zones. It is quite possible that fluvially derived fine-grained magnetic particles occurring as suspended load in the lower zone of the Mandovi estuary gets frequently exported out and accumulate in the nearshore region where relatively low-energy environment persist.

Previous studies have highlighted that the sediment cycling between ebb and flood tide is the major mechanism controlling the enhancement of magnetic particles within the estuarine environment (Badesab et al., 2012; Kulgemeyer et al., 2016, 2017). The samples from the lower zone exhibit wide range in magnetite content (Figure 3a; Table 1), mean grain size (Figure 3e), and unimodal type clastic grain size distribution (Figure 8c). This imply that mineral-density selective sorting is the major mechanism in the lower zone, where well-sorted sediments are entrained by bi-directionally cyclic reversing tidal currents for example, in the tidal channel network and other parts of the lower zone of the Mandovi estuary. Such process over time probably resulted in concentrating heavy (magnetic) particles and frequent washout of the lighter sediment fractions from the lower zone. The samples from upper and middle zone showed better grouping and exhibit relatively less variation in terms of their magnetite content and magnetic grain size (Figures 11a - 11c), but showed large scattering in the clastic grain size and mixed type distribution (mode 7.08  $\mu\text{m}$  to 471.14  $\mu\text{m}$ ; Figures 7a and 7b; Table 1). This imply that grain size selective sorting mechanism is the dominant in these zones, where one-directional flow (river runoff) of decreasing intensity first erode and then suspend bedload sediments, and later allow the magnetic particles to settle as the energy of the flow decreases. The trend of fining of

magnetic particles in the upper and middle zones (Figure 3b) support the above interpretation. Near-uniform and higher values of  $\chi_{lf}$  (Figure 3a) and S-ratio (Figure 3c) accompanied by the lowest values of ARM/SIRM (Figure 3b) suggest the dominance of coarse-grained ferrimagnetic particles in the lower zone.

## 5.2. A Magnetic Proxy for Tracing Riverbank Erosion

Soil losses linked with erosion of catchment and riverbank causes large increase in sediment fluxes in fluvial and estuarine systems, and are currently major environmental issues. Magnetic methods has been successfully utilized for tracing the soil erosion in the fluvial systems. For example, Franke et al (2020) established a linkage between the enhanced runoff input of high-coercivity particles in suspended sediment load and flood events in the Canche River watershed (Northern France). A mineral magnetic study on catchment soils successfully identified the highly eroding sites in Shibanzhao Catchment, Guizhou Plateau, southwest China (Wang et al., 2011). However, a direct relationship between soil erosion and the magnetic proxies for precise tracking of erosional sites in the geologically and hydrodynamically complex tropical estuarine system has not been established so far mainly due to lack of information on relative magnetic contribution derived from different sources to the total estuarine sediment load. For instance, geological terrains dominated by igneous, sedimentary, and metamorphic rocks in the catchment assure a high supply of ferrimagnetic minerals to the Mandovi estuary. Furthermore, the detrital magnetic particles derived from catchment areas gets sorted, settled, and dispersed during their transit from source-to-sink in response to the changing hydrodynamics in different sub-environments of the Mandovi estuary. The flood plains and riverbanks along the tropical Mandovi estuary are dominated by lateritic soils (top and sub), which are rich in antiferromagnetic minerals. Elevated soil contribution from eroding (laterite-rich) riverbanks would increase the concentration of high coercive minerals in the bedload sediments of the Mandovi estuary. Hence the erosional processes would remarkably hinder the magnetic signature of ferrimagnetic particles derived from catchment (magnetite-rich) rocks and such mineralogical changes can be easily detected by the sensitive magnetic (S-ratio) proxy.

Comparison of magnetic signals ( $\chi_{lf}$ , ARM/SIRM, S-ratio) of the source material, catchment soil, riverbank soil, and estuarine (upper, middle, lower, nearshore) sediments of the Mandovi estuary reveal large variation in terms of magnetic mineral content and mineralogy (Figures 11a-11e). Magnetic parameters ( $\chi_{lf}$ , ARM, SIRM and ARM/SIRM) of catchment soils and rocks show close grouping with the majority of estuarine samples suggesting that bulk sediment load of the Mandovi estuary is largely derived from hinterland (Figures 11a-11c). In contrast, riverbank samples exhibit distinct magnetic characteristics (highest  $\chi_{lf}$ , ARM, SIRM and lowest ARM/SIRM; Figures 11a-11c).

Elevated input of high coercive magnetic particles (Figure 3c) at few sites throughout all sedimentary zones of the Mandovi estuary can be linked with larger magnetic contribution from the eroding (lateritic) riverbanks. Frank and Nowaczyk (2008) demonstrated that hematite presence in sediments comprised of mixed mineral assemblage will be apparent in the magnetic remanence data only if the hematite content surpasses 80 wt% of total magnetic fraction. Uniform and higher values of  $\chi_{lf}$ , S-ratio, and low ARM/SIRM (Figures 3a-3c) in the lower zone (Coco Bay, Aquada, Reis Magos) indicate the dominance of coarser ferrimagnetic particles relative to antiferromagnetic which can be attributed either to the elevated flux of magnetic particles from catchment, reduced riverbank erosion in the bay area, increased magnetic contribution from the nearshore region or shift in the hydrodynamic regime (higher current velocities). Highest magnetic susceptibility and S-ratio values followed by lowest ARM/SIRM in the mid-range sieve fractions (40 – 45  $\mu\text{m}$ , 45 - 63  $\mu\text{m}$ ) suggests that isolated monocrystalline magnetite (coarse-silt sized) particles mainly exist and got enrich within this grain-size range which can be reconciled with the higher tidal action which controlled both mineral density and grain size selective entrainment and deposition (Figure S5p-r). This observation further confirm that higher energies in the lower zone favoured the accumulation of coarser magnetic particles which was clearly evident in the magnetic proxies (Figures 3a-3c).

Reductive diagenesis of magnetic minerals in an estuarine system could result in preferential dissolution of ferrimagnetic minerals compared to highly coercive antiferromagnetic minerals (Karlin and Levi, 1983; Robinson et al., 2000; Rey et al., 2005; Mohamed et al., 2011; Ge et al., 2015; Roberts, 2015; Nguyen et al., 2016). Antiferromagnetic minerals, such as hematite and goethite, are more stable and offers strong resistance to reductive dissolution compared to ferrimagnetic magnetite. Hence, a relative increase in the proportion of high coercive antiferromagnetic minerals in sediments will be recorded by changes in the S-ratio (Channell and Hawthorne, 1990; Poulton et al., 2004; Rey et al., 2005; Nowaczyk, 2011; Mohamed et al., 2011; Roberts, 2015). TOC content in the studied sediments vary between 0.1 to 3.0 % (Figure 3g). The bulk sediment magnetic properties are mainly dominated by coarse silt size ferri-and antiferromagnetic particles (Figure 3, Figure 10). In well-ventilated estuarine sediments such as Mandovi with permanent source of detrital magnetic minerals, secondary diagenetic minerals may still be present, but should have minor influence on the bulk sediment signal. Poor correlation between TOC content and magnetic susceptibility (Figure 11f) confirmed that reductive diagenesis had minimal effect on the magnetic properties of Mandovi River sediments and is not vital factor for the observed loss of finer magnetic particles in the different zones of the estuary. The observed decrease in S-ratio in different estuarine zones could be very well linked to the increased input of high coercive magnetic particles which are mainly derived from eroding riverbank soils (top

and subsurface), rather than significant reductive dissolution of primary ferrimagnetic minerals. This observation further established the direct link between S-ratio and the increased supply of high coercivity minerals from eroding riverbank deposits. The present study also highlights that S-ratio values of a sample from particular site are sensitive to the concentration of high coercivity magnetic fraction derived from various sources (catchment, riverbank erosion) and local hydrodynamic regime. As previously highlighted by Kayvantash et al. (2017) and Franke et al. (2020), the dependence of grain size between bulk sediment and magnetic fraction and concentration of magnetic particles are the crucial factors which needs to be considered while deconvolving magnetic variability caused by sediments originating from catchment (different lithology, topography), eroded soils from riverbank, and the prevailing hydrodynamics regime.

In estuarine environments, sedimentation rate plays an important role in controlling the magnetic mineral diagenesis in addition to other factors including organic matter supply, availability of sulfate, and reactive iron estuarine sediments (Canfield and Berner, 1987; Zhang et al., 2001; Rey et al., 2005; Robinson et al., 2000; Mohamed et al., 2011, 2017; Liu et al., 2012; Roberts, 2015; Ge et al., 2015; Nguyen et al., 2016). Increased sedimentation significantly influence the contact time between reactive iron, organic carbon, and sulfidic fluids which affect the reductive diagenesis of detrital magnetic particles (Canfield et al., 1992; Kasten et al., 2003; Reidinger et al., 2005; Dewangan et al., 2013; Roberts, 2015; Gaikwad et al., 2021). Higher SIRM/ $\chi_{lf}$  values (Figure 3e) only in the upper zone (Khandepar River) of the Mandovi estuary suggest the presence of diagenetically formed ferrimagnetic iron sulfides (Snowball and Thompson, 1990; Peters and Dekkers, 2003). In contrast, relatively lower values of SIRM/ $\chi_{lf}$  (Figure 3e) followed by a consistent trend in the majority of the samples (except in Khandepar River) suggest that early reductive diagenesis has minimal impact on the Mandovi estuarine sediments. This can be explained by the fact that enormous supply of detrital iron oxides from fluvial sources to Mandovi estuary might have hindered the diagenetic reactions and hence the detrital magnetic particles were less affected by dissolution and remained preserved (Kao et al., 2004; Amiel et al., 2020; Reilly et al., 2020; Gaikwad et al., 2021).

Biogenic magnetic particle synthesized by magnetotactic bacteria are ubiquitously found in estuarine environments (Kirschvink and Chang, 1984; Petersen et al., 1986; Stolz et al., 1986; Hounslow and Maher, 1996; Hilgenfeldt, 2000; Kopp and Kirschvink, 2008; Roberts et al., 2015). Magnetic grain size proxy (ARM/SIRM) sensitive to biogenic particles did not exhibit any anomalous values typical for bacterial mineralization (Figure 3b). IRM unmixing revealed that the significant magnetic contribution is mainly derived from detrital magnetic minerals and did not exhibit any magnetic components reflecting typical characteristics (narrow grain size distribution, low coercivity;

Figure 7) of biogenic magnetic minerals. In Neel's plot (Figure S8; Tauxe et al., 2002), the samples plot mostly within USD region and may not necessarily correspond to a grain size range of biogenic magnetite. A dedicated study focussing on bacterial mineralization aspect of the estuarine sediments is further warranted to elucidate the influence of biogenic magnetic minerals on the bulk sediment magnetism. Our findings are summarized in a generic conceptual model (Figure S2), which can probably be generalized to the other estuarine regions.

Bulk magnetic properties of estuarine sediments are sensitive proxies for environmental conditions, but the robustness of their interpretation require precise information on the different sources of magnetic minerals that determine the sediment magnetic signal. A suite of sediment magnetic properties presented in this study are highly sensitive to the iron bearing minerals, which are ubiquitous in estuarine system and offer to rapidly and non-destructively characterize the magnetic minerals concentration, grain size, and mineralogy of the natural samples rapidly compared to geochemical, sedimentological, and mineralogical analyses, which are time consuming and require specialized training and high end instrumentation.

## **6. Conclusion**

(1) Based on the careful evaluation of the rock magnetic, mineralogical, and sedimentological data of the catchment rocks, soils, riverbank soils, estuarine, and nearshore surface sediments of the Mandovi estuary, we provide new data and interpretations on the dynamics (mixing, dispersal, fractionation, settling) of the magnetic particles and the underlying constraints during their transit from source-to-sink.

(2) Multi-proxy data helped in evaluating the relative magnetic contribution derived from different sources to the bulk estuarine sediment load. We developed a sediment magnetic mineralogy based proxy (S-ratio), which is sensitive and can be utilized for tracking riverbank erosion in fluvial and estuarine system.

(3) A magnetomineralogical and granulometry based approach presented in this study can be very well generalized to other types (micro and macro tidal) of an estuarine system to track the control of source-to-sink processes on the dynamics and fate of magnetite-size fractions. Furthermore, additional information on the suspended and beload sediment fluxes during fair and extreme weather conditions is prerequisite to model the magnetite distribution map and understand the dynamics of magnetic particles in different subsystems of the estuary.

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## **Conflict of Interest**

The authors declare no conflicts of interest relevant to this study.

## **Data Availability Statement**

All data obtained in this study are available and can be downloaded in free access at CSIR-NIO repository (Badesab et al., 2023).

## Figure Captions

Figure 1. (a) Geological map of the study area showing the distribution of main lithologies across Goa, India (Source: *Geological and mineral map of Goa (1:125,000)*, Geological Survey of India, 1996). (b) Locations of sampling sites (black dots) including rock, riverbank soil profile (RB-01), and surface and subsurface soil profiles (SP-1, SP-2, SP-3) from Mandovi River catchment, Goa, west coast of India. The entire study area has been demarcated into four sedimentary magnetic zones (upper, middle, lower, nearshore) mainly based on the variations in the salinity and estuarine turbidity maximum.

Figure 2. (a-g): Spatial distribution of magnetic minerals as indicated by concentration dependent magnetic susceptibility ( $\chi_{lf}$ ) within and off Mandovi estuary, Goa, west coast of India. The white (filled) circles indicate the location of sampling sites. The magnetic susceptibility map of surface sediments was generated using IDW interpolation in QGIS software.

Figure 3. Plot showing variation in magnetic (a) mass specific magnetic susceptibility ( $\chi_{lf}$ ) represents the abundance of ferrimagnetic minerals (magnetite), (b) magnetic grain size indicator (ARM/SIRM), (c) magnetic mineralogy indicators (S-ratio) and (d) hard isothermal remanent magnetization (HIRM), (e) magnetic iron sulfide proxy (SIRM/ $\chi_{lf}$ ), (f) median grain size, and (g) total organic carbon (TOC) content measured on surficial sediments within and off Mandovi estuary, Goa, west coast of India. The entire study area has been demarcated into four sedimentary magnetic zones (upper, middle, lower, nearshore) mainly based on the variations in the salinity and estuarine turbidity maximum. Background colors are used only to highlight different zones of the estuary. Each data point marked in blue crossed circle proximal to the sampling sites (Coco Bay and Mandovi River transects) indicates the geo-mean value of magnetic parameters.

Figure 4. Depth variations of magnetic parameters (a,f,k,p) magnetic susceptibility, (b,g,l,q) magnetic grain size indicator ( $\chi_{fd}\%$ ), (c,h,m,r) saturation isothermal remanent magnetization, (d,i,n,s) magnetic grain size indicator (ARM/SIRM), and (e,j,o,t) magnetic mineralogy proxy (S-ratio) of the soil profiles from riverbank (a-e) and catchment SP-1 (f-j), SP-2 (k-o), and SP-3 (p-t) within the Mandovi estuary, Goa, west coast of India.

Figure 5. Temperature dependence of magnetic susceptibility ( $\chi-T$ ) for selected representative samples from the four (upper, middle, lower, nearshore) sedimentary zones of the Mandovi estuary, Goa, west coast of India. Solid red lines indicate heating curves, and blue lines indicate cooling curves.

Figure 6. (a-f): The hysteresis loops for selected representative samples from the four (upper, middle, lower, nearshore) sedimentary zones of the Mandovi estuary, Goa, west coast of India. Hysteresis loops are corrected for non-paramagnetic contribution.

Figure 7. (a-g): The magnetic coercivity distribution of seven surficial sediment samples representing the four (upper, middle, lower, nearshore) sedimentary zones of the Mandovi estuary, Goa, west coast of India. Magnetic components of the samples were identified from deconvolution of IRM acquisition gradient curves on MAXUnMix program (Maxbauer et al., 2016).

Figure 8. Grain size distribution of surficial sediments from the four different sedimentary zones (a) upper (b) middle (c) lower, and (d) nearshore of the Mandovi estuary, Goa, west coast of India. Background arrow is used only to highlight the trend of loss of finer sediment fraction from upper to lower zone of the Mandovi estuary.

Figure 9: (a-f): Total organic carbon (TOC) distribution map of the surface sediments of the Mandovi estuary, Goa, west coast of India. The map was generated using IDW interpolation in QGIS software. The white (filled) circles indicate the location of sampling sites within and off the Mandovi estuary.

Figure 10. (a-h): Scanning electron microscope images (secondary electron) on magnetic extracts from different sedimentary zones of the Mandovi estuary, Goa, west coast of India. Energy dispersive X-ray spectroscopy spectra are placed adjacent to the respective images (a–h). Iron (Fe), titanium (Ti), oxygen (O), silicon (Si), aluminum (Al), and gold (Au) peaks are indicated. Please note that Mt: Magnetite, TM: Titanomagnetite, TH: Titanohematite, and Gt: Goethite.

Figure 11. (a-e): Scatter plots comparing the magnetic parameters of catchment rocks, catchment soil profiles, riverbank soil profile, and surficial sediments from the four (upper, middle, lower, nearshore) different sedimentary zones of the Mandovi estuary, Goa, west coast of India. (f) total organic carbon content (TOC) is plotted against the magnetic susceptibility of the surface estuarine sediments only. Arrows (gray color) are used only to highlight the trends.



## Tables

**Table 1.** Magnetic and grain size parameters of surficial sediments from the four (upper, middle, lower, nearshore) different sedimentary zones of the Mandovi estuary, Goa, west coast of India.

Parameters	Sediment magnetic zones	Minimum	Maximum
Magnetite (Wt. %)	Overall	0.0145	3.9874
	Upper	0.0610	3.9874
	Middle	0.0404	1.0987
	Lower	0.0145	0.6491
	Nearshore	0.559	0.667
Magnetic susceptibility $\chi_{lf}$ ( $10^{-6}\text{m}^3\text{kg}^{-1}$ )	Overall	0.0957	26.317
	Upper	0.4030	26.317
	Middle	0.266	7.251
	Lower	0.0957	4.2846
	Nearshore	3.6956	4.4030
Magnetic grain size indicator proxy (ARM/SIRM)	Overall	0.00197	0.0624
	Upper	0.00364	0.0624
	Middle	0.00197	0.0347
	Lower	0.00450	0.0291
	Nearshore	0.00667	0.00815
Magnetic mineralogy (S-ratio)	Overall	0.124	0.999
	Upper	0.124	0.999
	Middle	0.329	0.995
	Lower	0.179	0.970
	Nearshore	0.964	0.983
Clastic sediment mean grain size ( $\mu\text{m}$ )	Overall	10.8542	458.69
	Upper	10.8542	390.894
	Middle	54.7708	382.353
	Lower	85.4967	458.69
	Nearshore	29.0455	38.821
Clastic sediment median grain size ( $\mu\text{m}$ )	Overall	6.20667	382.353
	Upper	6.20667	322.564
	Middle	27.8101	382.353
	Lower	84.2691	372.892
	Nearshore	16.7492	24.6979
Clastic sediment mode grain size ( $\mu\text{m}$ )	Overall	7.08392	517.2
	Upper	7.08392	471.14
	Middle	168.872	429.182
	Lower	87.9022	517.2
	Nearshore	14.9396	31.5071

**Table 2.** Magnetic properties of the surficial sediment samples from the Mandovi estuary, Goa, India estimated by using unmixed IRM acquisition curves (Maxbauer et al., 2016). (Dp = dispersion parameter; B<sub>1/2</sub>=mean coercivity (mT); C = percentage of remanence contribution).

Sr. No	Sediment Magnetic Zones	Component 1			Component 2			Component 3		
		B <sub>1/2</sub> (mT)	DP	C (%)	B <sub>1/2</sub> (mT)	DP	C (%)	B <sub>1/2</sub> (mT)	DP	C (%)
01	Upper Zone	17	0.28	29	48	0.25	50	747	0.33	21
02	Upper Zone	26	0.47	55	57	0.28	33	587	0.29	12
03	Middle Zone	18	0.33	36	60	0.24	47	615	0.29	17
04	Middle Zone	26	0.43	58	64	0.28	34	525	0.23	8
05	Lower Zone	18	0.28	6	47	0.33	33	731	0.36	61
06	Lower Zone	20	0.27	16	47	0.26	39	838	0.29	45
07	Nearshore Zone	21	0.23	21	62	0.26	66	518	0.24	14

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