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ORIGINAL ARTICLE

Geochemical and sedimentological characteristics of surface sediments from Ashtamudi Estuary, Southern India: implications for provenance and modern sedimentary dynamics

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Abstract

Geochemical and end-member mixing analyses (EMMA) of the grain-size distribution were conducted on recent surface sediments from Ashtamudi Estuary, Southern India to investigate the hydrodynamic factors that influence the depositional processes in the region. The complex interplay between the natural (fluvial and marine) and anthropogenic influences on the Ashtamudi Estuary have been delineated based on the inter-relationships between geochemical elements and the end members (EM) derived from the grain-size parameters. Si, Al and Fe are the major elements in the sediment of Ashtamudi Estuary constituting 26–47%, 9–21%, and 8–14%, respectively. Four grain-size end members were identified with dominant modes of 7ϕ (EM1), 6ϕ (EM2), 4.5ϕ (EM3), and 3.3ϕ (EM4) corresponding to fine silt, medium silt, coarse silt, and fine sand, respectively. The high contributions of Al, Fe, Cr and Ni combined with the EM1 and EM2 in the upstream of the estuary indicated that the fine-grained sediments were derived from the fluvial inputs into the basin. The marine/tidal influenced lower estuary is characterised by high concentrations of Si, Ti, Ca, Sr and coarse fraction end member (EM3). The elemental concentration of Cu, Zn and Co along with EM4 shows higher values along the shore region suggesting human interventions in the basin. This integrated geochemical analysis and EMMA provide advancement in the knowledge of the transportation mechanisms and regional sediment dynamics from the estuarine system in Southern India.

Keywords End-member modelling · Grain size · Estuary · Geochemistry · Southern India

Introduction

The estuaries are dynamic environments characterised by variable energy conditions and multiple sediment sources (Meade 1972; Zwolsman and Van Eck 1999). The sediment particles in estuaries are comprised of allochthonous river-borne terrestrial detritus and tidally advected marine inputs and autochthonous materials derived from biogenic sources (Burton and Liss 1976; Ankit et al. 2017). The sediment transported across the estuarine system is mostly controlled by the interaction between fluvial discharge and tidal or oceanic currents (Wright 1977). In addition, human activities (e.g., basin dredging and port constructions) also alter the morphodynamics of the estuarine environments (Perillo et al. 2005; Williams et al. 2013). The surficial sediment composition and their spatial distribution characteristics are widely used to decipher the recent sediment dynamics and level of natural versus anthropogenic impacts in estuarine systems (Rubio et al. 2000; Ip et al. 2007; Liu et al. 2016).

The Ashtamudi Estuary, a RAMSAR site, is the second largest estuarine system in the southwest coast of India. This estuary is characterised by riverine dominant upper part, whereas the middle and lower ends have a marine/tidal influence (Jennerjahn et al. 2008; Anooja et al. 2013; Ankit et al. 2017). Most surveys and studies from Ashtamudi Estuary have dealt with understanding the hydrochemical

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characteristics (Divakaran et al. 1981; Sujatha et al. 2009; Babu et al. 2010; Antony and Ignatius 2016), estimation of organic matter sources (Nair et al. 1984; Jennerjahn et al. 2008; Reshmi et al. 2015; Ankit et al. 2017) and assessment of heavy metal concentrations in sediments for enrichment and contamination (Babu et al. 2010; Krishnakumar et al. 2015; Nagendra et al. 2017). However, until now, there has been a lack of information on the geochemical and sedimentological processes that control the behaviour and transport of the particles across the estuary.

The major objective of the study is to acquire a better understanding of the modern processes that control the supply and composition of the inorganic sediment components in the Ashtamudi Estuary. The surface sediment samples from Ashtamudi Estuary were analysed for their elemental composition and grain-size distribution. The geochemical distribution patterns of major, minor and trace elements constituents of Ashtamudi sediments have been described by multivariate analysis. The end-member mixing analysis (EMMA) was performed on the grain-size parameters to understand the sediment mixtures in a series of sedimentary components (end members) reflecting the transport mechanisms and sediment sources.

Study area

Ashtamudi Estuary (8°53′–9°02′N, 76°31′–76°41′E) is the interface between the Kallada River in the north and the Arabian Sea in the south (Fig. 1a, b). The estuary is approximately 16 km long with an average width of ~ 3.2 km and covers an area of ~ 51 km^2 (Sajan et al. 1992).

Morphologically, the region around the Ashtamudi Estuary consists of gently sloping valleys, coastal plains, channel fill deposits and tidal flats (Nair et al. 2010; Padmalal et al. 2011). The beaches and inner shelf environment of the region composed of sandy sediments are dominated by quartz and heavy minerals (Prakash 2000; Prakash et al. 2007). The major lithological units in the Ashtamudi region comprise the Archaean crystalline basement, Tertiary and Quaternary sedimentary sequences (Nair et al. 1984; Varghese 2014). The Archaean crystalline basement is represented by a group of metamorphic rocks (e.g. garnet biotite gneisses, khondalites and charnockites) dominant in the eastern and southeastern parts of the Kallada basin (Mohan et al. 2016; Vijith et al. 2016). However, the sedimentary sequences of Tertiary and Quaternary deposits occupy the southeastern part of the estuary (Varghese 2014).

The Ashtamudi Estuary exhibits a funnel-shaped bathymetry with a maximum depth of ~ 9 m (Nair et al. 2010; Ankit et al. 2017). Climatically, the region is situated within the tropical climate zone with mean annual temperature ranging from 26 to 33 °C (Divakaran et al. 1981) and annual precipitation varies from ~ 1170 to ~ 2300 mm/year (source: metrological data from Indian Meteorological Department during 2012–2016). The summer monsoon contributes ca. 53% of rainfall, whereas the northeast (NE) monsoon contributes ~ 24% of annual rainfall in the region. The hydrographic characteristics of the Ashtamudi Estuary are controlled by the monsoon-fed Kallada River discharge and strong tidal influence in the lower estuary (Nair et al. 1984, 2001).

The human interventions in and around Ashtamudi Estuary have resulted in deteriorating water quality and massive land reclamation (Sitaram 2014). The industrial pollution, tourism development, contamination from fishing and its processing industries and urban sewage discharge have substantially modified the environmental equilibrium of the Ashtamudi Estuary (Jennerjahn et al. 2008). In addition, continuous dredging activity by the Port Department for navigation purposes has altered the natural characteristics of sediment deposition in downstream of the estuary (Nair et al. 1983; Krishnakumar et al. 2015).

Methodology

Sample collection

A total of 34 surface sediment samples was collected from the Ashtamudi Estuary using the Van Veen Grab sampler with a penetration depth of 5–7 cm. The samples were sealed in glass vials and sent to laboratory for analysis.

Laboratory analyses

Bulk elemental geochemistry

A total of 21 powdered sediment samples (~ 0.2 g) was digested by a mixture of acids (HNO_3, $HClO_4$, HF) (conventional method—open system) for total elemental concentration measurements (e.g., Balachandran et al. 2003; Papaefthymiou et al. 2010). The elemental analysis was performed using ICP-AAS (Varian AA-30 model). The National Research Council Canada (NRC Canada; BCSS-1 and MESS-1) standards were analysed to check the accuracy of the results. The precision of the major and minor elements is < 2% and 5% for trace elements.

Grain-size analysis

For grain-size analysis ($n = 34$), the pretreatment includes wet oxidation of organic matter at room temperature using 10 mL H_2O_2 (30%) in ~ 0.3 g of the sediments. Furthermore, the pretreated samples were centrifuged and washed several times to remove extra oxidizing agent. The solution was then

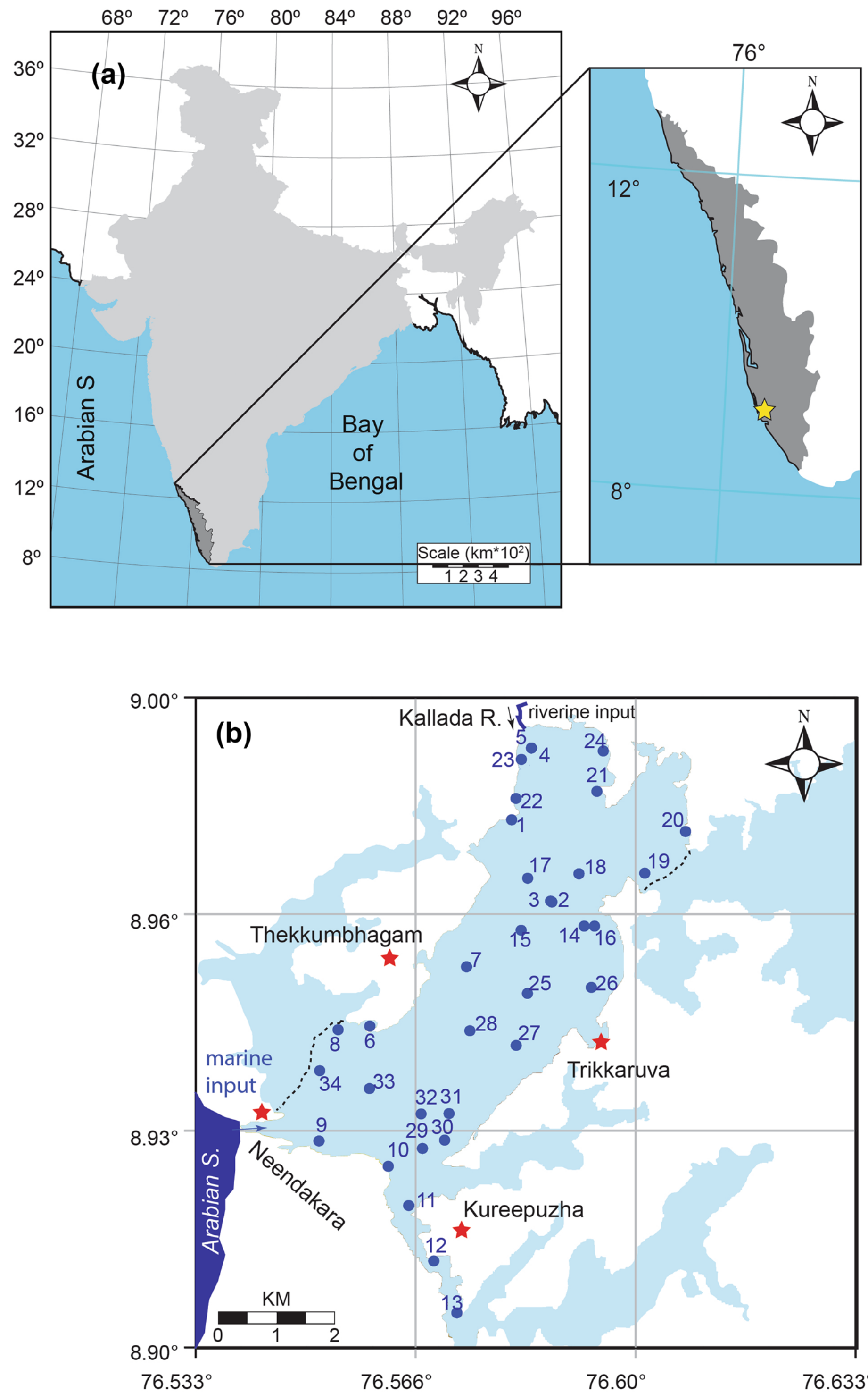

Fig. 1 **a** Location of the study area (indicated with a 'yellow star'); **b** map of Ashtamudi basin with sampling locations of the sediments

treated with an ultrasonic bath to prepare a homogenous solution for analytical purpose. The grain-size measurements were carried out using a Malvern Mastersizer 2000E laser grain size analyzer. The grain-size distribution was calculated for 100 grain-size classes (particle sizes between 0.02 and 2000 μm) and the analytical error was less than 1%.

Statistical analyses

Principal component analysis

The principal component analysis (PCA) was conducted on the geochemical parameters of Ashtamudi sediments to understand the factors controlling the spatial distribution of sediments in the basin. PCA was performed using *princomp*-package in R-language-based software (e.g., Venables and Ripley 2013).

End-member mixing analysis

End-member mixing analysis is a statistical technique to provide a meaningful solution in various modes of transportation of the sediments by unmixing polymodal grain-size distribution (Weltje 1997; Prins et al. 1999; Dietze et al. 2012). The statistical analysis has been done using *EMMAgeo*-package, based on R-language (Weltje 1997; Dietze et al. 2012; Heinecke et al. 2017). To decipher the best-fit model for grain-size distribution, the relation between the coefficient of determination (r^2) and a probable number of end member (q) has been used (Dietze et al. 2013). This relation defines the stability of the model and explains the probable number of end members used to define the various modes of transportation in the basin (Zhou et al. 2004; Dietze et al. 2013; Borchers et al. 2016).

Results

Spatial distribution of elemental concentration

The geochemical concentration of 21 samples is listed in Table 1. The sediments are mainly dominant in Si, Al and Fe with average values of 32.6, 14.9 and 10.8 wt%, respectively, except in the samples (EL28–30 and EL32–34), where Ca has a significant concentration (average = 10.5 wt%). The spatial distribution of the major elemental composition in the surface sediments shows large variability along the Ashtamudi basin. The distribution of major elements indicated that the upstream of the estuary is dominant in Mg, Fe, Na and Al, whereas Si, Ti and Ca are dominant in the downstream of the estuary (Fig. 2).

The PCA analysis of geochemical parameters extracted three end members (PC1, PC2 and PC3) which together explain ~75% of the variance in the data set (Fig. 3). The first principal component (PC1) shows a strong positive correlation with Ti, Sr, Ca, Co, Si and K, whereas a negative correlation is observed for Ni, Al, Fe, Mg, Cu and Cr. The negative axis of PC2 suggests its close association with Mg, Cr, Na and K. In contrast, the PC3 shows a strong affinity with Cu and Zn indicating point source distribution in the basin near the southeastern end of the estuary (Trikkaruva) (Figs. 1b, 2).

Spatial distribution of grain-size variability

The grain-size distribution of the surface sediments from the Ashtamudi Estuary is largely characterised by silt with minor variation in the samples from the north (dominant in clayey silt) and south (comprises sand-sized particles) (Fig. 4a). The mean grain size of the Ashtamudi estuarine sediments ranges between 7.1 and 11.3ϕ (average = 8.3ϕ) and shows the lowest value towards the downstream of the estuary (Fig. 4b).

The end-member mixing analysis of grain-size distribution in Ashtamudi estuarine sediments yielded an optimal model with four end members (Fig. 5a) and explains 86% of the variability in the total dataset. EM1 (mode = 7.0ϕ) and EM2 (mode = 6.0ϕ) are the highest near the confluence of Kallada river and southeastern end of the estuary (Fig. 5b, c). Similarly, EM3 with a mode of 4.5ϕ that lies in the range of coarse silt is the highest towards the seaward side of the estuary near Neendakara (Fig. 5d). The coarsest end member (mode = 3.3ϕ), i.e., EM4 is highest near the shore of Thekkumbhagam and around the small islands in the southern end of the estuary (Fig. 5e).

Discussion

Spatial distribution of geochemical and sedimentological parameters

The distribution pattern of geochemical elements in the estuarine environment is mostly governed by variability in riverine inputs, redistribution of sediments by coastal processes, biological activities as well as anthropogenic influence (Zwolsman and Van Eck 1999; Liaghati et al. 2004; Zhou et al. 2004). PCA loadings are used in the present study to elucidate the factors that are primarily responsible for influencing spatial distribution of geochemical concentrations in Ashtamudi surface sediments. The loading plot from the PCA analysis revealed four groups of elements.

The first group comprises Si, Ti, Ca, Sr and Co with higher values towards the downstream of the estuary (Figs. 2, 3a). The high concentration of Si and Ti in the southern end of the estuary can be attributed to tidal advection of inner shelf sediments dominated by quartz and heavy minerals (e.g., TiO_2). The calcium content increased from 0.8 to 14.9% from the fluvial to the marine end. The Ca in sediments is contributed by biogenic ($CaCO_3$) sources and other calcium-bearing detrital minerals (as plagioclase feldspar). The negative relationship of Ca and Sr with Al indicates a dominant non-detrital (biogenic) source for these elements (Basavaiah et al. 2014; Cui et al. 2015; Mishra et al. 2015). Interestingly, the downstream of the estuary is characterised by the presence of bio-detritus which is

Table 1 Geochemical parameters of Ashtamudi estuarine sediments

Sample name	Latitude	Longi-tude	Si (%)	Al (%)	Fe (%)	Ti (%)	Ca (%)	Mg (%)	Na (%)	K (%)	P (%)	Cu (ppm)	Pb (ppm)	Zn (ppm)	Ni (ppm)	Co (ppm)	Cr (ppm)	Mn (ppm)	Sr (ppm)
EL 14	8.965	76.592	31.17	16.29	12.09	1.09	3.28	2.32	4.82	1.77	0.36	40	5	100	60	10	185	320	200
EL 15	8.964	76.582	30.9	16.35	11.49	1.05	3.82	2.32	5.53	1.82	0.32	35	5	95	60	10	175	290	200
EL 16	8.965	76.594	28.67	15.35	10.97	0.95	2.73	2.72	6.64	1.71	0.24	35	5	90	55	5	155	310	170
EL 17	8.972	76.583	29.94	15.55	10.61	0.92	3.16	2.63	6.91	1.74	0.32	35	25	100	60	5	195	490	220
EL 18	8.973	76.591	26.32	14.66	8.9	0.77	2.4	2.87	9.32	1.51	0.2	45	5	90	50	5	135	390	145
EL 19	8.973	76.601	34.11	15.8	14.33	0.94	3.61	2.12	3.78	1.07	0.1	45	5	90	45	5	100	550	80
EL 20	8.979	76.607	34.03	18.24	11.49	0.9	2.11	1.79	3.59	1.69	0.09	45	5	100	55	5	155	420	70
EL 21	8.986	76.594	27.32	18.75	10.47	0.8	1.84	2.51	6.03	1.51	0.24	60	5	90	65	5	200	580	100
EL 22	8.984	76.582	28.3	20.08	11.98	0.84	2.08	1.97	3.74	1.44	0.14	50	10	95	50	5	110	480	70
EL 23	8.990	76.582	33.1	21.41	12.94	1.09	1.08	0.99	2.43	1.74	0.18	55	10	100	55	5	120	430	85
EL 24	8.992	76.595	27.85	21.09	12.75	0.84	0.8	1.98	3.35	1.51	0.12	55	10	95	65	5	160	540	45
EL 25	8.954	76.583	32.09	14.24	12.24	1.14	5.31	2.23	4.94	1.85	0.29	35	5	90	55	2.5	170	250	135
EL 26	8.955	76.593	28.76	15.36	10.63	0.99	2.39	2.58	7.04	1.76	0.32	140	5	150	60	2.5	160	310	170
EL 27	8.946	76.582	31.95	14.12	10.66	1.14	3.48	2.54	6.61	1.86	0.36	35	5	100	65	2.5	200	260	210
EL 28	8.949	76.575	33.03	12.98	10.66	1.16	6.31	2.36	5.77	1.83	0.32	30	5	115	50	2.5	160	240	310
EL 29	8.931	76.567	34.65	9.09	8.61	2.35	12.15	1.67	4.93	1.58	0.28	35	5	85	35	10	130	350	500
EL 30	8.932	76.571	34.14	11.83	9.46	1.39	9.41	1.83	5.22	1.83	0.24	30	5	95	45	10	145	270	440
EL 31	8.936	76.572	33.76	13.88	10.97	1.21	4.29	2.08	5.97	1.83	0.24	35	5	95	50	10	165	220	235
EL 32	8.936	76.567	47.19	8.53	8.01	3.45	8.5	1.32	4.35	2.08	0.22	20	5	70	25	20	115	390	480
EL 33	8.940	76.559	43.01	8.81	7.92	3.3	11.54	1.23	4.26	1.98	0.2	30	5	85	25	20	120	390	540
EL 34	8.942	76.552	34.74	9.57	9.46	1.65	14.85	1.43	3.41	1.81	0.25	35	5	105	35	20	140	290	620

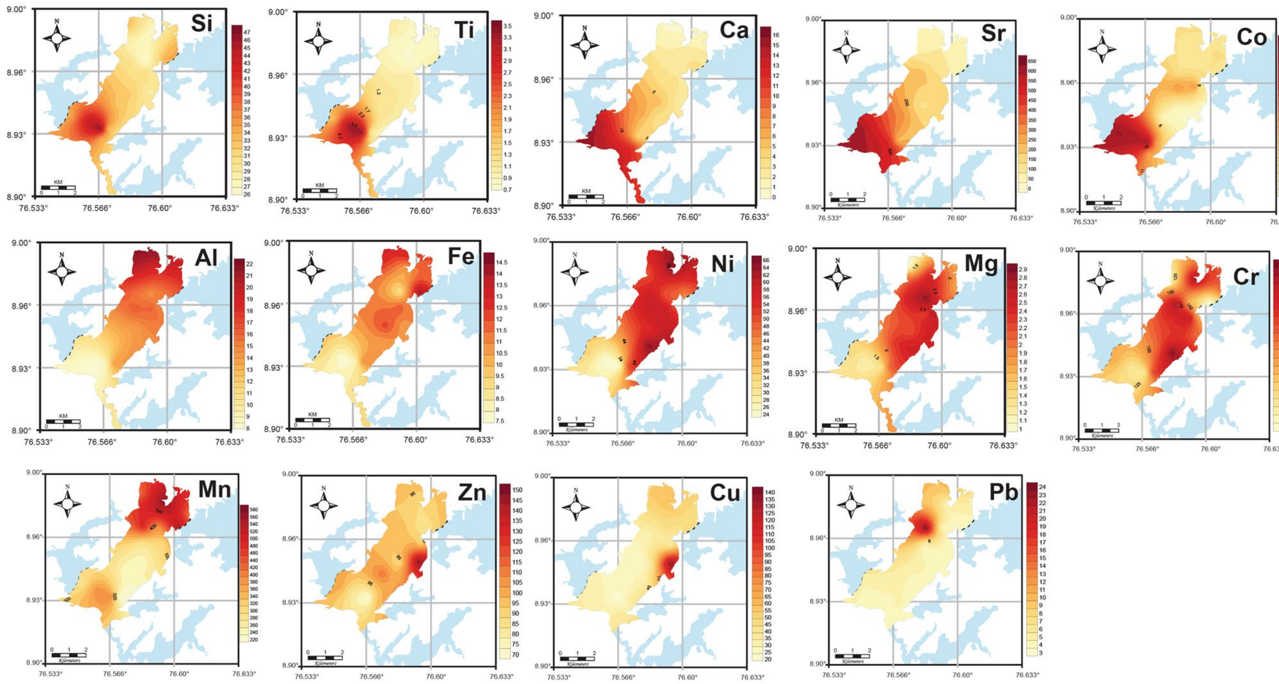

Fig. 2 Spatial distribution of various geochemical parameters

largely composed of calcium carbonate (Jennerjahn et al. 2008; Ankit et al. 2017). Therefore, the high Ca and Sr values and negative relationship with other terrestrial-derived elements (e.g., Al and Fe) indicate the high biological productivity observed in the marine-influenced lower estuary. Furthermore, cobalt concentration (2.5–20.0 ppm) shows an increase from the upper to the lower end of the estuary with an average value of 7.9 ppm. In an estuarine environment, Co is derived from both natural as well as anthropogenic effluents including chemical factories, sewage effluents, urban runoff and agricultural activities (Sanzgiri and Moraes 1979; Nagpal 2004). The reason for relatively higher Co in Ashtamudi Estuary could be related to human activity, as the downstream of the estuary is heavily disturbed by the discharge of industrial effluents (Sitaram 2014).

The second group of elements (e.g., Al, Fe and Ni) is characterised by negative loadings of PC1 with higher values towards the upstream of the estuary due to their terrestrial origin (Fig. 3a). The upper catchment of the estuary in the north is marked by Archean rocks with the dominance of mafic minerals enriched in Fe and Ni. The high concentration of Fe and Ni in the upper estuary is attributed to the weathering in the upper catchment of the basin. Furthermore, the high concentration of Al in the upper estuary is related to the dominance of clay minerals.

The elemental group (Mg, Cr, Na, P and K) is characterised by high negative loadings of PC2 (Fig. 3b). Because of the low concentration of P (0.09–0.36 wt%) and K (1.07–2.08 wt%), these elements were not considered in our interpretation. The negative axis of PC2 shows the group of elements controlled by dual sources in the basin. For example, Mg is contributed by both geogenic (from the mafic rocks) and biogenic sources (Mucci and Morse 1982; Mishra et al. 2014). However, in Ashtamudi Estuary, the spatial distribution of Mg shows high concentration towards the upper end of the estuary. We attribute weathering in upper catchment and fluvial processes as the key factor controlling the Mg concentration in the basin. Similarly, a higher concentration of Na in the upstream of the estuary demonstrates that the contribution of Na is largely governed by the fluvial input into the basin (Table 1). Likewise, Cr shows a high concentration towards the upper end of the estuary (Fig. 2). Cr is enriched in mafic–ultramafic rocks which are dominant constituents in the upper catchment of the estuary. Meanwhile, the high concentration of Cr in an estuarine environment is also related to various anthropogenic activities such as small-scale industries, e.g., electroplating, textile industries (Krishnan and Jaya 2014; Karim and Williams 2015).

Cu and Zn show a low significant correlation with PC1 and PC2 and are only explained by PC3 (Fig. 3c). The spatial distribution of these elements indicated anthropogenic sources. Copper and zinc are potentially used in antifouling paints and galvanic anode in boats (Turner 2010) (Table 1). Near Trikkaruva, the high concentrations of these elements indicated the role of anthropogenic activities in controlling their concentration. This interpretation is in line with human-induced enrichment of Cu and Zn in Ashtamudi sediments (Krishnakumar et al. 2015).

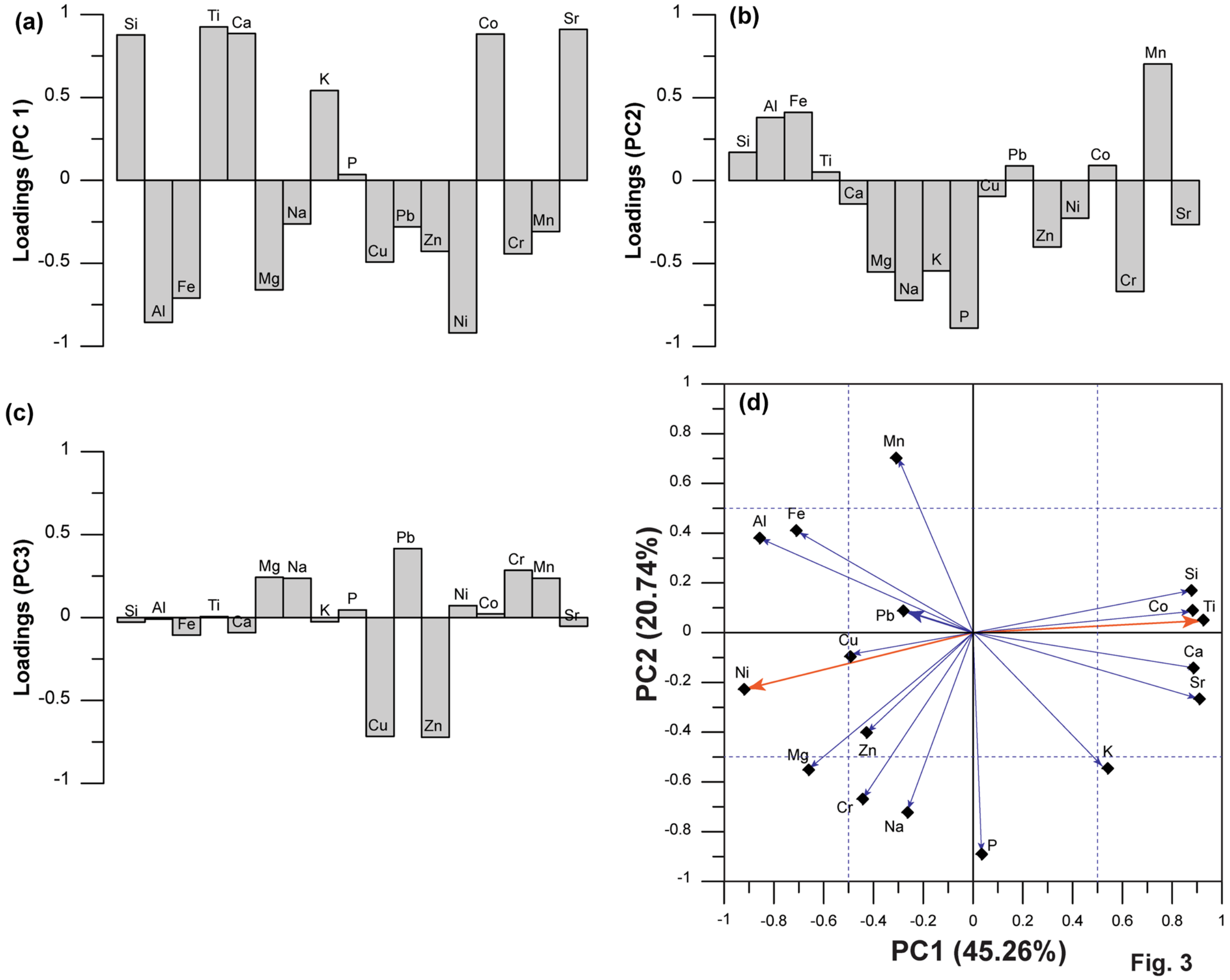

Fig. 3 **a–c** Loadings of principal components (PCs) derived from elemental data; and **d** biplot between PC1 and PC2

Statistical approach to understanding grain-size distribution

The grain-size distribution in the estuarine system is mostly controlled by waves and tidal currents, intensity of fluvial discharge as well as anthropogenic activities (Allen 1991; Anithamary et al. 2011; Liria et al. 2009; Wang et al. 2010; Oyedotun 2016). The combined effects of these activities modify the original grain-size distributions in the basin. Therefore, it is essential to unmix the grain-size distribution in terms of the various depositional processes in the basin. The end-member modeling analysis of the grain size provides quantitative estimation of sediment transport processes from multimodal grain-size distributions (Dietze et al. 2012).

The result of EMMA is shown in Fig. 5. The EMMA of the grain-size distribution in Ashtamudi sediments provides four EMs suggesting the various modes of deposition/sedimentation in the basin. EM1 (mode = 7ϕ) and EM2 (mode = 6ϕ) with a dominant mode in fine silt demonstrate the highest occurrences toward the upstream and southeastern end of the estuary (Fig. 5b, c). The result is also corroborated with mean grain-size distribution in the basin which shows the finest sediment towards the riverine-influenced upstream area of the estuary (Fig. 4b). The grain-size variability of riverine inputs in estuarine environment depends on the intensity of discharge and geomorphological constraint which includes flow velocity, water turbulence and channel length (Allen 1971; Dessai et al. 2009). The coarser sediments are usually trapped in the flood plain of the river with finer fractions being transported into the estuary (Dyer 1995). The trapped coarser fractions are released into the estuaries during high-energy fluvial discharge events (Dyer 1995; Ip et al. 2007). The fine-grained sediments in the upstream area of the Ashtamudi Estuary can be attributed to the variability in the fluvial discharge.

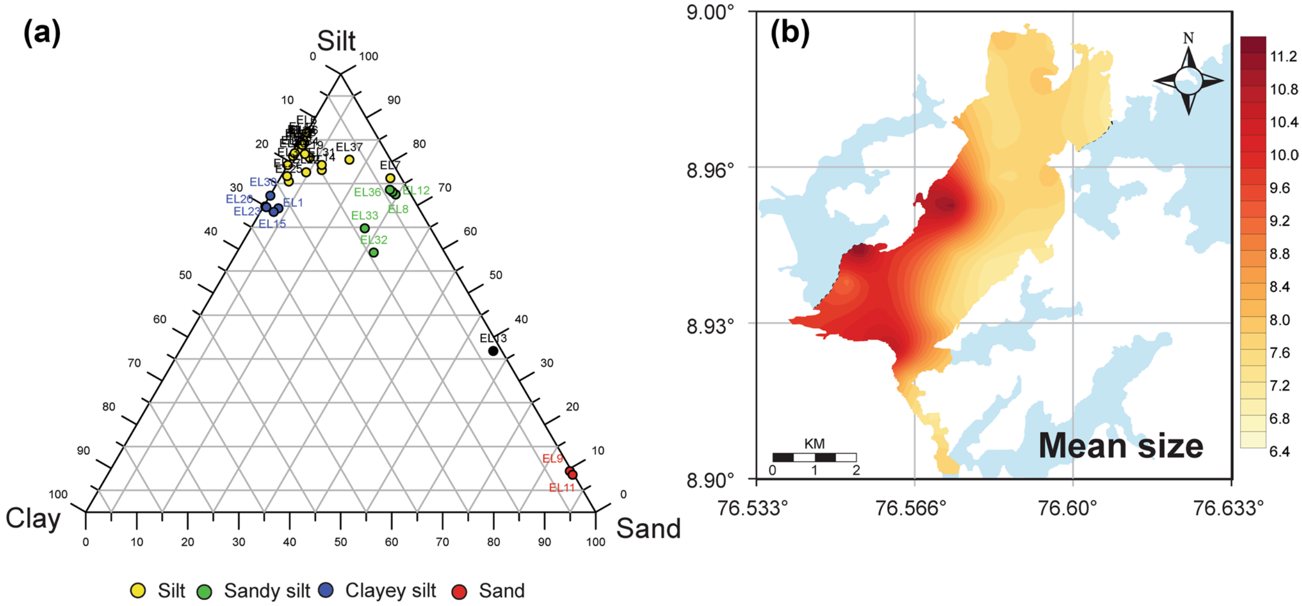

Fig. 4 **a** Ternary diagram of grain-size distribution of the Ashtamudi estuarine sediments; and **b** spatial distribution of mean grain size

The EM3 concentration with a dominant mode of 4.5ϕ (~45 µm) lies in the range of coarser silt. The EM3 shows higher values towards the tidal-dominated seaward side of the estuary (near Neendakara) (Fig. 5d). The tidal-induced enhanced erosion in the downstream estuary has resulted in increased supply of coarser sediment into the Ashtamudi Estuary. Furthermore, the presence of coarsest sediments (EM4; mode = 3.3ϕ or 100 µm) near Thekkumbhagam (a region with maximum human interventions) indicate a possible role of anthropogenic alterations. The area around Thekkumbhagam has been subjected to several dredging and construction activities (Nair et al. 1983; Rajan and Bindhu 2011) resulting in poorly sorted coarse sediments represented by EM4 (Fig. 5e). The end-member modeling analysis of grain-size distribution delineates the natural versus anthropogenic factors in controlling the sediment distribution in Ashtamudi Estuary.

Conclusions

The integrated geochemical and sedimentological analyses of Ashtamudi surface sediments were conducted to disentangle the natural (fluvial/marine) versus anthropogenic influence in basin sediments. The sediments in the upper stream of the estuary are characterised by Al, Ni, Cr, and Fe derived from the weathering of catchment rocks in the Kallada basin. Additionally, the high contribution of EM1 and EM2 indicates relatively fine-grained sediments carried by the fluvial input in the suspended form. A high concentration of Si, Ti Ca, Sr and EM3 (phi = 4.5ϕ) was observed in the seaward side of the estuary indicating tidal interaction. The anthropogenic impact of the Ashtamudi Estuary is indicated by the high concentration of Cu, Zn, Co and EM4 (phi = 3.3ϕ) in the basin. Several small-scale industries, the influx of domestic discharge, and pollution from the mechanized fishing boats in the lower estuary and vicinity of Thekkumbhagam are the regions of the high anthropogenic inputs in the basin. The integration of geochemical analysis with the end-member analysis on grain-size parameters has provided a sound framework to understand sedimentary processes along the continental margin of Southern India.

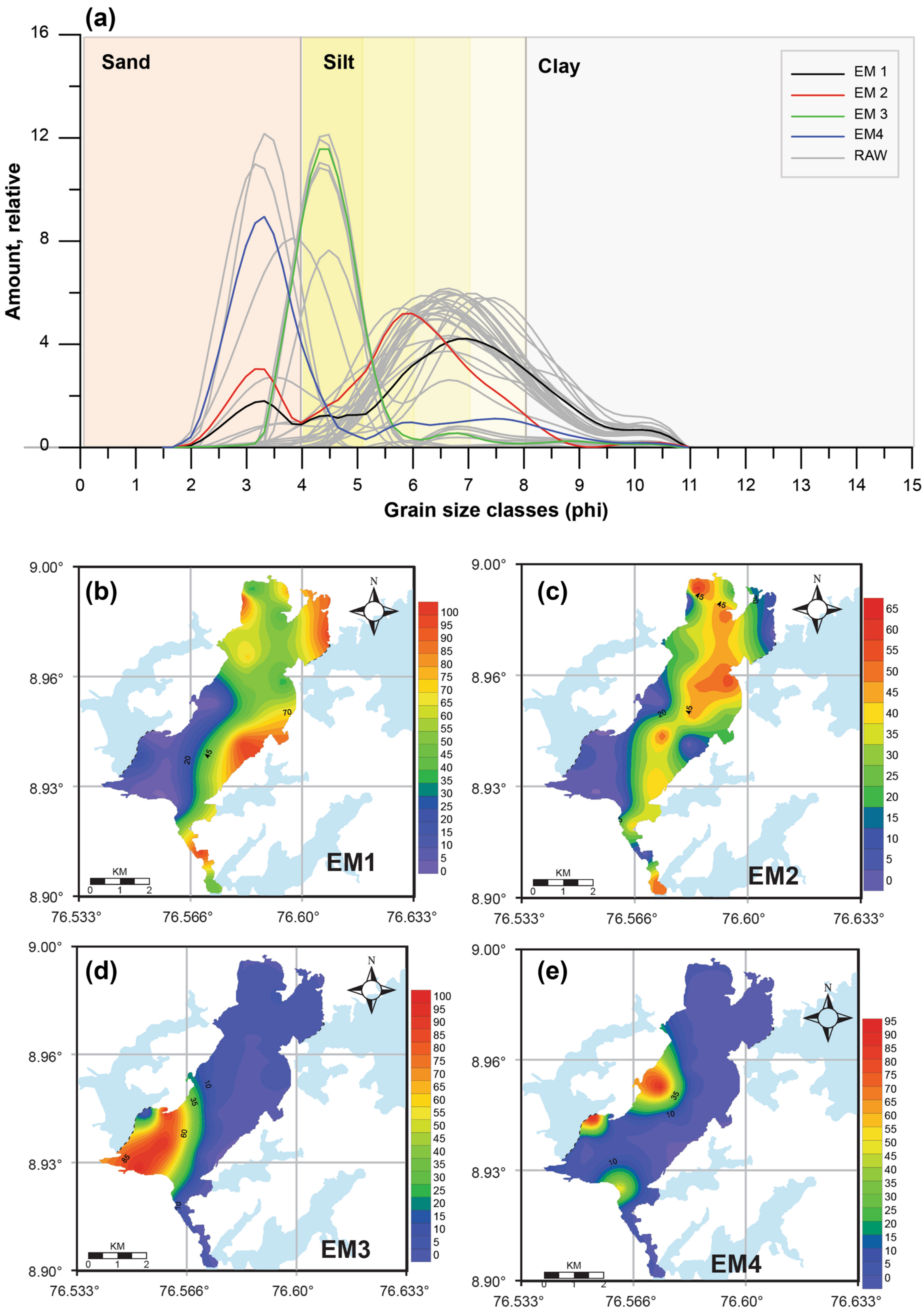

Fig. 5 **a** End-member loadings of grain-size distribution; **b**–**e** spatial distribution of end members (EMs)

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Compliance with ethical standards

Conflict of interest The authors confirm that they have no conflict of interest.

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