JOURNAL GEOLOGICAL SOCIETY OF INDIA
Vol.86, July 2015, pp.107-114

Geochemical and Mineralogical Studies in Recent Clastic Sediments from Upper Godavari River in Peninsular India

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Abstract: Detailed geochemical and clay mineralogical investigations were carried out on bed sediments from upper Godavari river of peninsular India. The results suggest that these sediments are mainly derived from weathering of basaltic rocks of Deccan volcanic province. Strong relationship between Fe_2O_3 and TiO_2 is attributed to their co-occurrence in weathering resistant minerals and/or scavenging of Ti by Fe oxy-hydroxides during weathering of basalts. The concentrations of Na_2O, K_2O and MgO in these upper Godavari river sediments are relatively less compared to parent basalt. The abundance of minor elements like V, Cr, Cu and Zn shows significant scatter within the range of Deccan basalts. Vanadium and nickel shows significant correlation with Fe_2O_3 due to their association with iron minerals or their sequestration with iron oxy-hydroxides. Higher Zn concentrations at some locations of Godavari river may have been due to anthropogenic contamination. The REE pattern is greatly influenced by the degree of source rock weathering. Light rare earth elements (LREE) show greater fractionation compared to the heavy rare earth elements (HREE) during weathering and transportation. Strong positive Ce-anomaly in some sediment samples suggests oxidizing conditions of deposition. Chemical Index of Alteration (CIA) varies significantly from 37.01 to 60.16, indicating large spatial variability in the intensity of chemical weathering of Deccan basalts. Low CIA values are observed in samples that are enriched in CaO probably due to semi-arid climate which facilitates $CaCO_3$ precipitation and restricts chemical weathering.

Keywords: Godavari river sediments, Elemental compositions, Rare earth elements (REE), Chemical weathering, Clay minerals.

INTRODUCTION

Chemical and mineralogical compositions of river sediments are extremely important in understanding the geochemical cycling of elements (Martin and Meybeck, 1979; Milliman and Meade, 1983). Weathering of rocks largely depends on the geology of the region, topographic setting, climate and anthropogenic activity (Nesbitt, 1979). The effect of weathering on major, trace and rare earth elements of eroded products has been studied by several workers (Nesbitt, 1979; Duddy, 1980; Taylor and McLennan, 1985; Cullers et al. 1987; Sholkovitz, 1988; Biksham et al. 1991; Vital and Stattegger, 2000; Sharma and Rajamani, 2000; Singh and Rajamani, 2001). Major and trace element composition of river sediments are controlled by lithology, weathering, diagenesis and sedimentary sorting in catchment areas and therefore can be used to identify provenance of clastic sediments. Similarly, REEs are useful to decipher the sedimentary environments as sediments derive their REE compositions from source rocks (Piper, 1974; Alexander and Gibson, 1977; Condie, 1991; Ross et al. 1995; Sensarma et al. 2008; Kessarkar et al. 2010; Shynu et al. 2011).

The REEs show a unique behavior during chemical weathering and transportation because of the similarity in their electronic configuration. REEs are very mobile, particularly during the early stages of weathering (Nesbitt, 1979; Condie, 1991). In particular, LREE/HREE ratio is useful in studying sediment origin and their mobility in the crust (Ross et al. 1995). During the weathering process the LREE are preferentially scavenged, whereas HREE are retained in dissolved state (Sholkovitz, 1995; Yang et al. 2002; Caccia and Milero, 2007). Moreover, REEs entering into the river are fractionated due to the removal of certain elements because of complex chemical reactions (Andersson et al. 2005). REEs are generally mobilized under acidic pH and get adsorbed on particles or precipitated when the pH is neutral or alkaline (Nesbitt, 1979). All fractions in sediments derived from an intensely weathered source show negative Eu anomaly (Cullers et al. 1987). Under reducing conditions, Eu is reduced from Eu^{+3} to Eu^{+2} state and this

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effect leads to distinctive geochemical behaviour as compared to the other REEs. On the other hand, under oxidizing environment Ce^{+3} is oxidized to Ce^{+4}. Therefore, the abundance of these two elements is important to infer redox conditions of geochemical systems.

Several workers have carried out geochemical studies on suspended and bed load sediments from rivers flowing through the Deccan traps (Subramanian et al. 1985; Ramesh et al. 1989; Dekov et al. 1998). These studies were aimed to decipher behavior of various elements during weathering and transportation. Das and Krishnaswami (2007) have done detailed investigations on elemental chemistry of sediments from different rivers flowing through the Deccan traps to investigate nature of chemical weathering and mobility of major and minor elements. The present study focusses on geochemical and mineralogical characteristics of upper Godavari river sediments flowing through an important basaltic province to know the behaviour of various major and trace elements during weathering, transportation and deposition. Hence, the main aim of this study is to determine (i) the major, minor and trace elemental composition of sediments, (ii) to utilize the geochemical data set for understanding the relative mobility of elements from the parent rocks during weathering and transportation, and (iii) to evaluate the effects of anthropogenic contamination on elemental concentrations of bed sediments is determined.

GEOLOGICAL SETTING

Godavari river is one of the largest rivers of India. It originates in the Deccan traps and drains an area of 3.1 x 10^5 km^2. The river flows in east and south-easterly direction for a distance of 1465 km before joining into the Bay of Bengal. The sediments discharged by Godavari river is estimated to be 170 x 10^6 ty^{-1} (Biksham and Subramanian, 1988). The upper reaches of Godavari basin are occupied by the Deccan traps of Cretaceous period and the middle part consists of Archaean granites, phyllites, quartzites, amphibolites and gneisses (Fig. 1). River Godavari flows nearly 48% on Deccan traps, 11% Precambrian and Gondwana sediments and 2-3% recent alluvium. The Deccan basalts are mainly tholeiitic lavas made up of plagioclase, pyroxene and olivine. Among pyroxenes, augite and pigeonite are more common (Sethna and Sethna, 1988). There are reports of zeolites in the cavities of basalt (James and Walsh, 1999). There is evidence of occurrences of

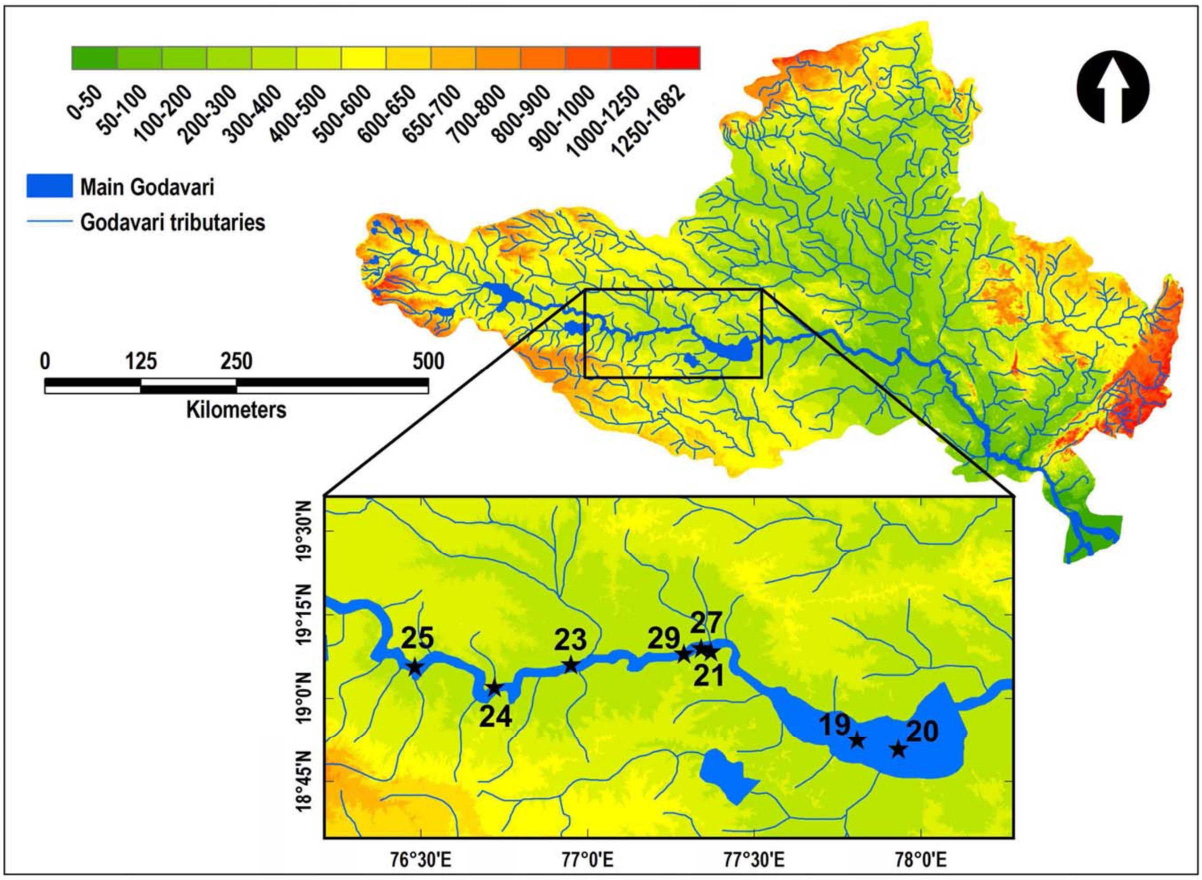

Fig.1. Location map of upper Godavari region indicating sampling stations.

calcium carbonate in the basin as a component of bed sediments (Das et al. 2005). Calcite is also reported as a minor phase in some of the basalts (Jeffery et al. 1988). Lateritic and black coloured soils are predominant at many locations of Deccan traps. These black soils are mostly fine textured with high content of smectite clay and contain low concentrations of Ca, Na, Mg and several trace elements because of the continental leaching of parent basalts (Widdowson and Gunnell, 2004). The sediments from Godavari are plastic and fine grained compared to the Himalayan rivers. This difference is due to the variation in weathering regimes, as Godavari river sediments are mainly derived by the chemical weathering as against both physical and chemical weathering in the Himalayan region. The climate of the study area is subtropical with semi-arid conditions. Under modern conditions rainfall in the study area is about 70 cm/yr. Most of the rainfall (~ 85%) occurs during the southwest monsoon (June-September). River Godavari has several dams both in upstream and downstream of sampling locations.

MATERIALS AND METHODS

Major, trace and rare earth elements were determined in bulk sediment samples by using X-ray fluorescence spectrometer (XRF) and Inductively Coupled Plasma spectrometer (ICP-OES). Major elements were estimated by wavelength dispersive XRF (model Philips MagiX PRO, PW 2440) using pressed pellets. Pressed pellets were prepared by using collapsible aluminium cups. These cups were filled with boric acid and about 1g of the finely powdered sample and then pressed under a hydraulic press. Details of the sample preparation and instrumental parameters used for major elemental analysis by XRF are provided elsewhere (Krishna et al. 2007). Trace and rare earth elements were estimated by Agilent ICP-OES (Model 725 ES) at NGRI, Hyderabad. Sediment samples were digested overnight in PTFE Teflon beakers using an acid mixture of HF, HNO_3, and $HClO_4$ in 7:3:1 proportion. The beakers were then heated on a hot plate at 180 °C for about 1 hour and evaporated to near dryness. The evaporation process was repeated after adding 5 ml of the above acid mixture in each sample. Finally, the residue was dissolved by gently heating in 20 ml of 1:1 HNO_3. Clear solutions were obtained for all samples. After cooling to room temperature, the volume was made up to 50 ml, and these final solutions were stored in polyethylene bottles. Trace and REE analysis was carried out by using international rock standards like AGV-1, MAG-1, MRG-1 and BRS in order to calibrate and monitor precision and accuracy of ICP-OES analysis. Routine measurements of blank and standards were carried out after every five samples for better precision. The overall relative standard deviation (TSD) is better than 5% for trace and rare earth elements.

Clay mineral estimation has been carried out at the CSIR-National Institute of Oceanography, Goa in separated finer fraction (< 2 μm) of sediments by using X-ray diffraction (XRD) technique (Rao and Rao, 1995). Clay fractions were separated from the total sediment samples by standard settling procedure before XRD analysis (Folk, 1968). Carbonate content and organic matter was removed by treating with acetic acid and hydrogen peroxide respectively. Oriented clay slides were then prepared by pipetting concentrated solutions onto glass slides and allowed to dry in air. These slides were glycolated at 100°C for 1 hour to carry out X-ray diffraction studies on Philips X-ray diffractometer using nickel filter CuKα radiation (Kessarkar et al. 2010).

MAJOR, TRACE AND REE GEOCHEMISTRY

The concentrations of major and minor elements in sediments are given in Table 1 and 2 respectively. Major and trace elements in upper Godavari river sediments suggest that they are mainly derived from mafic minerals of Deccan basalts. The SiO_2 content in sediments ranges from 40.59 to 51.87% and Al_2O_3 from 9.51 to 11.59%. There is strong positive correlation between SiO_2 and major oxides like Na_2O, Fe_2O_3, MgO and TiO_2 (Table 1). The MgO content is relatively uniform (1.57 - 2.35%). CaO concentrations vary significantly from 4.59 to 15.33% indicating variations in degree of weathering of source rock and/or precipitation of $CaCO_3$. Previous work in bed load sediments from Bhima and Krishna tributaries flowing through Deccan traps have also reported a large variation in $CaCO_3$ contents (from 0 to ~ 17%) (Das and Krishnaswami, 2007). Similarly Mahi River in Deccan Trap has higher concentrations of K_2O and CaO (Sharma et al. 2013). The TiO_2 concentrations vary considerably from 0.57 to 1.91% (Table 1), whereas Na_2O (2.05-2.60%) and K_2O (0.11-0.56%) are relatively uniform. In most of the upper Godavari sediments Na_2O, K_2O and MgO are relatively less compared to the parent basalt (Table 1).

The mobility of elements during weathering of Deccan basalts follows the trend (Na H" Ca e" Mg H" Sr) > (K e" Ba) > (Al e" Fe H" Ti) (Das and Krishnaswami 2007). During the chemical weathering, Na, K and Ca are removed by surface fluid as ion to form predominantly smectite clay mineral. However there is a considerable spatial variability in the intensity of chemical weathering in Deccan traps.

Table 1. Major elemental concentrations with calculated CIA values. (wt. %).

Element (ppm)	Station 19	Station 20	Station 21	Station 23	Station 24	Station 25	Station 27	Station 29	Avg. Parent basalt*
SiO_2	44.92	47.38	47.09	49.07	40.59	51.87	49.13	41.39	47.78
TiO_2	0.79	1.23	1.02	1.37	0.57	1.91	1.68	0.60	2.36
Al_2O_3	9.51	11.13	9.90	11.25	10.27	11.59	10.96	10.30	12.35
Fe_2O_3	5.23	9.07	6.09	9.16	3.92	10.25	9.39	4.14	14.53
MnO	0.14	0.22	0.18	0.18	0.20	0.19	0.18	0.20	0.19
MgO	1.76	1.99	1.89	2.18	1.59	2.35	2.12	1.57	5.03
CaO	13.29	4.59	13.27	5.94	15.05	8.06	6.42	15.33	8.08
Na_2O	2.24	2.22	2.37	2.32	2.05	2.60	2.34	2.08	2.41
K_2O	0.16	0.56	0.11	0.39	0.13	0.16	0.48	0.12	0.35
P_2O_5	0.18	0.19	0.20	0.19	0.17	0.22	0.21	0.17	0.23
LOI	17.30	18.44	14.94	14.99	20.17	8.35	14.10	19.91	6.23
Total	95.52	97.02	97.06	97.04	98.14	97.55	97.01	98.38	99.52
CIA	37.74	60.16	38.60	56.53	37.35	51.72	54.26	37.01	55.66

LOI- Loss of Ignition, CIA- Chemical Index of Alteration *Chavan (2012)

Table 2. Trace and REE concentrations (ppm)

Element (ppm)	Station 19	Station 20	Station 21	Station 23	Station 24	Station 25	Station 27	Station 29	Parent basalt*
Ba	268.72	225.99	287.7	197.44	410.14	168.99	200.94	400.6	-
Cd	1.35	0.75	1.07	4.81	0.82	1.47	2.69	2.3	-
Cr	214.32	104.15	258.74	159.2	201.72	359.59	185.34	139.73	-
Cu	72.09	110.84	86.18	137.09	46.84	140	146.67	57.03	-
Ni	36.08	43.32	34.31	55.72	33.42	50.35	46.33	32.25	-
V	210.05	223.34	288.87	368.85	169.89	494.19	422.08	180.56	-
Zn	55.62	86.63	72.94	109.48	35.34	143.95	138.66	44.35	-
La	10.21	6.59	9.12	6.92	12.90	6.75	6.45	12.60	11.86
Ce	30.24	60.08	24.96	46.01	29.57	30.55	44.61	26.35	40.9
Pr	2.23	2.07	2.25	2.53	2.62	2.97	2.73	2.51	3.52
Nd	12.25	14.15	14.74	15.69	10.97	19.09	17.07	10.83	21.0
Sm	3.97	3.65	3.87	3.58	4.71	3.58	3.35	4.80	5.49
Eu	1.01	0.79	0.99	0.97	1.25	1.08	0.93	1.18	1.87
Gd	2.01	2.37	2.28	2.86	2.03	3.12	2.84	2.03	6.75
Tb	0.22	0.18	0.20	0.23	0.17	0.24	0.30	0.19	1.08
Er	0.51	0.58	0.70	0.86	0.48	1.14	1.06	0.50	3.15
Yb	0.69	0.64	0.67	0.67	0.78	0.78	0.66	0.71	2.48
Lu	0.05	0.07	0.05	0.08	0.03	0.09	0.08	0.04	0.36
LREE	58.90	86.54	54.95	74.72	60.78	62.94	74.21	57.10	82.77
HREE	4.48	4.62	4.89	5.67	4.74	6.45	5.87	4.65	15.69
SREE	63.38	91.16	59.84	80.39	65.51	69.39	80.07	61.75	98.46
LREE/HREE	13.15	18.71	11.23	13.19	12.83	9.76	12.65	12.27	5.28

*Chavan (2012)

Lower values of calcium and magnesium in riverine sediments are generally attributed to their higher mobility during chemical weathering. Chemically immobile elements like TiO_2 and SiO_2 provides insight into chemical behavior of major mineral phases. The major element composition of sediments is displayed on ternary diagram of Al_2O_3, (CaO + Na_2O) and K_2O (A-CN-K) (Fig. 2). All the points lie along the mixing line of basalt and laterite. This spread suggests that the bed sediments have been subject to varying degree of weathering. A positive correlation between Al_2O_3 and Fe_2O_3 suggests the alteration of biotite into aluminisilicates and iron oxides (Table 1). At some locations CaO concentrations are found to be significantly high (13.27 to 15.33%). These samples exhibit very low CIA values ranging from 37.01 to 38.60 (Table 1). The source of Ca could be from calcretes and tufas in soils (Patil and Surana, 1992; Pawar and Kale, 2006). Calcretes and tufas are formed by precipitation during dry climate. Alternatively high CaO could be due to the semi-arid climate of the region, which facilitates $CaCO_3$ precipitation and restricts chemical weathering. It is possible that river water at some locations in Deccan traps is supersaturated with reference to calcite,

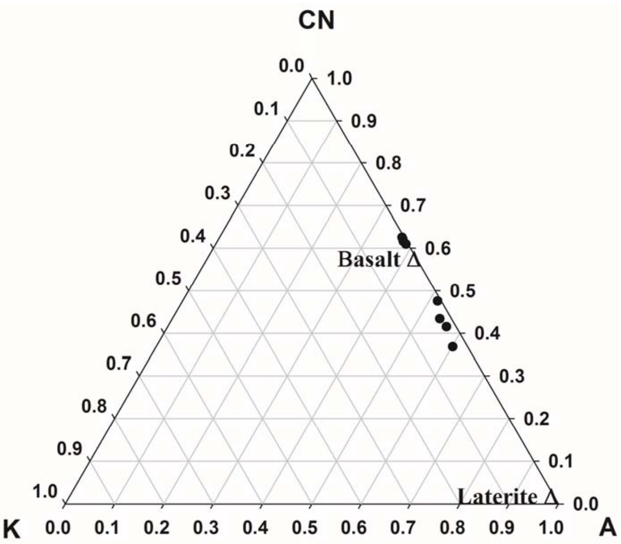

Fig.2. Ternary diagram K-CN-A [K_2O-($CaO+Na_2O$)-Al_2O_3].

resulting in precipitation of calcium carbonate from river water (Das et al. 2005). The chemical indexes of alteration (CIA) values in these sediments vary from 37.01 to 60.16 with an average value of 46.67 (Table 1). Very low CIA values in some samples indicate presence of basaltic fragments indicating incipient weathering under semi-arid climate. It may be noted that CIA values are sensitive to land surface temperature, latitude at river mouth and soil depth in a drainage basin. Interestingly our CIA values are identical (37 to 59) to Mahi river sediments (37 to 59) that flow through Deccan traps (Sharma et al. 2013).

The abundance of minor elements like V, Cr, Cu and Zn shows significant scatter within the range of Deccan basalts. The concentrations of V (169.9 to 494.2 ppm), Ni (32.3 to 55.7 ppm), Cr (104.2 to 359.6 ppm), Cu (46.8 to 146.7 ppm) and Zn (35.3 to 144.0 ppm) vary significantly (Table 2). Vanadium and Nickel shows significant correlation with Fe_2O_3 due to their association with Iron minerals or their sequestration with Iron oxy-hydroxides (Table 2). Zn contents are high (72.9 to 144.0 ppm) at some locations probably due to anthropogenic activities. Human activities such as agriculture and industry may have resulted in higher Zinc concentrations. Ba concentrations are ranging from 169.0 to 410.1 ppm. High barium contents in these sediments are attributed to the presence of K-feldspar. Nemee (1975) has reported high Ba content in K- feldspar megacrysts.

Total rare earth elements concentrations in upper Godavari sediments vary between 64 and 95 ppm (Table 2). There is a significant increase in LREE compared to HREE with high LREE/HREE ratios (5.87 to 10.46). River borne sediments usually have REE pattern that are strongly depleted in HREE (Sholkovitz, 1988). The difference in REE content, particularly LREE in river sediments may be due to the variations in the relative abundance of coarse and finer fractions at different locations. Cullers et al. (1988) have demonstrated that coarser sand fractions have lower REE content relative to the silt and clay size fractions and showed varying shapes of REE pattern in different size fractions. LREE are fractionated significantly in upper Godavari river sediments during weathering, transportation and deposition with respect to parent basalt as illustrated in Fig. 3a & b. Previous workers have shown that that REE are fractionated during weathering processes and the weathered residual material is generally enriched in LREE and depleted in HREE. River sediments have uniform REE patterns with enrichment of LREE and depletion of HREE relative to NASC (North American Shale Composite) (Goldstein and Jacobsen, 1988; Sholkovitz, 1995). The extent of LREE fractionation in our sediment samples is also evident by high $(La/Yb)_N$ ratios (ranging from 8.65 to 17.75). In contrast, HREE shows less fractionation as seen by the low $(Gd/Yb)_N$ ratios (2.86 to 4.30). Interestingly the $(La/Yb)_N$ ratio in weathered parent rocks at a nearby location of our sampling stations is also low 4.78 (Chavan, 2012) indicating fractionation of REE during weathering process.

Chondrite normalized REE distribution pattern of parent basaltic rock in Fig. 3a is essentially flat and linear with no Eu-anomaly and a weak positive Ce-anomaly. A significant feature of the chondrite normalized REE pattern in upper Godavari river sediments is the presence of weak to strong positive Ce-anomaly (Fig. 3a). The REE pattern exhibits a shape characteristic of Deccan basalt with more depletion in LREE content (Fig. 3a) (Taylor and McLennan, 1985). Some samples exhibit moderate positive Ce-anomaly and a few samples show a strong Ce-anomaly. Ce-anomaly has been observed earlier in basaltic soils of Deccan basalt near our sampling locations (Chavan, 2012). Positive Ce-anomalies have been reported in many weathering profiles with different types of source rocks. For example lateritic ferricretes from the Ivory Coast have shown strong positive Ce-anomaly (Steinberg and Courtis, 1976). Positive Ce-anomalies have been reported on chondrite normalized REE patter from four laterite profiles in Cameroon beneath the zone of iron oxide accumulation (Braun et al. 1990). Cerium concentrations in the weathering environments are important because it can occur as Ce^{+3} or Ce^{+4} under the oxidizing conditions. Consequently, the sediment shows a positive Ce-anomaly. Under the prevailing chemical weathering conditions in the catchment areas of upper Godavari floodplains, the bulk of the REE are derived by the weathering of mafic minerals. It appears that the contribution

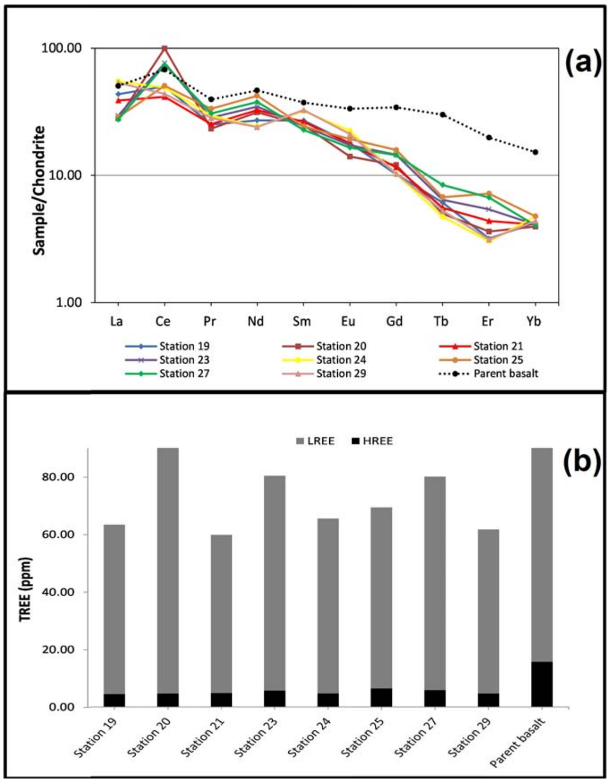

Fig.3. (a) Chondrite-normalized REE pattern of upper Godavari river sediments. (b) Bar diagram showing abundances of LREE and HREE with parent basalt composition of LREE and HREE (Chavan, 2012).

from basaltic soil and laterite is greater at some locations resulting in the strong Ce-anomaly. A close examination of REE bar diagram (Fig. 3b) suggests that the bulk of HREE were retained in the parent basaltic rocks during the weathering process. The control of titaniferous minerals on REE geochemistry in upper Godavari river sediments may be ruled out as TiO_2 is not related to total REE content (Tables 1&2).

CLAY MINERALOGY

The clay fractions of upper Godavari river sediments have predominantly smectite clay. Other clay minerals like kaolinite and chlorite are present in small quantities and illite is absent in all samples (Table 3). The smectite content in upper Godavari river sediments is very high (85.2 - 90.6%) compared to the lower Godavari sediments (24.6 – 54.0%) (Vuba et al. 2013). Also kaolinite content is very low (4.1-8.9%) compared to lower Godavari river sediments indicating incipient weathering under semi-arid climate. The variations in the relative abundances of clay minerals in upper and lower reaches of Godavari may be related to the variations in the source rock composition, degree of alteration, type of vegetation, topography of the region. The smectite and kaolinite are derived by chemical weathering of basalts and granites (Chamley, 1989). The total content of smectite and kaolinite in Godavari river sediments vary from 91.6 to 95.9% (Table 3). The highest smectite concentrations in the shelf sediments off east coast of India are reported to be due to the sediment load from the Godavari and Krishna river systems (Rao et al. 1988; Rao, 1991).

Table 3. Clay mineralogical compositions in < 2 μm fraction of sediments

Station No.	Smectite %	Iilite %	Kaolinite %	Chlorite %	K/C %	S+K %
19	85.2	-	8.9	5.9	1.51	94.1
20	90.6	-	5.3	4.1	1.29	95.9
21	90.3	-	4.1	5.6	0.72	94.4
23	88.6	-	4.2	7.3	0.57	92.8
24	85.9	-	5.7	8.4	0.68	91.6
25	88.6	-	7.3	4.1	1.77	95.9
27	88.3	-	7.6	4.1	1.86	95.9
29	89.5	-	5.6	4.9	1.13	95.1

Godavari River passes through regions that have black cotton soils and thick vegetation which contain a large percentage of smectite clay. Deccan traps in the upper reaches of Godavari River are usually altered by weathering to laterite or bauxite and the resulting soils contain about 80% smectite with kaolinite as minor clay mineral (Somayajulu et al. 1993). The drainage area in the upper reaches of Godavari River is a smectite rich zone, but in the middle and lower reaches the river passes through khondalites and charnockite-type of rocks and granites of Archean age. These rocks on weathering under humid conditions produces mixed clay (Vuba et al. 2013). Raman et al. (1995) reported that Godavari and other Peninsular Indian rivers transport clay with high smectite content with minor amounts of kaolinite and illite. They have suggested that Deccan basalts and Archaean rocks are the source of smectite in the Peninsular Indian Rivers. Kulkarni et al. (2014) have suggested that the high smectite content in Godavari river sediments are due to the weathering of basaltic rocks. The smectite introduced through Godavari in shelf region of the Bay of Bengal is poorly crystalline probably due to its basic volcanic parent rock.

CONCLUSIONS

- The chemical and mineralogical investigations in the upper Godavari river sediments suggest that these sediments are predominantly derived by the incipient weathering of basaltic rocks with contribution from lateritic soil in the hinterland.
- Major, trace and rare earth elemental compositions

indicate evidence of sedimentary sorting during weathering, transportation and deposition.

- Strong relationship between Fe_2O_3 and TiO_2 is attributed to their co-occurrence in weathering resistance minerals and/or scavenging of titanium by iron oxy-hydroxides formed during weathering of basalts.
- Chondrite-normalized REE pattern demonstrate a weak to strong positive Ce-anomaly suggesting oxidizing environment of sediment deposition. No Eu-anomaly is observed in any sample indicating insignificant contribution from plagioclase.

Acknowledgements: We acknowledge financial support from 12th plan projects INDIAS IDEA, SHORE and INDEX. We are thankful to Mr. Netramani Sagar, Mr. Waseem Raza, Mr. K. Venkatesham, Mrs. G. Suseela, Mr. Mahjoor Lone and Mr. Tabish Raza for their help during the course of this study. We express our sincere gratitude to Dr. V. Purnachandara Rao and his colleagues at CSIR-NIO, Goa for XRD analysis and to Dr. Keshav Krishna for XRF analysis. The financial assistance received through ISRO-GBP project and the Department of Science & Technology, Govt. of India is thankfully acknowledged.

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(Received: 17 July 2014; Revised form accepted: 11 March 2015)