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Major, trace and REE geochemistry of the Ganga River sediments: Influence of provenance and sedimentary processes

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A B S T R A C T

The sediments of the Ganga River from different depositional regimes in the Plain region such as the river channel, active flood-plain and the older flood-plain sediments from the inter-fluve region were analysed for major, trace and the rare earth elements (REEs). These are compared with catchment zone sediments of the river and probable source rocks in the Himalaya. The lower CIA values between 48 and 54.7 for the catchment sediments indicates that the sediments supplied to the Ganga Plain are chemically immature and subjected mostly to physical weathering due to higher erosion rates in the Himalaya. The CIA values ranging between 55 and 74, with average value of 59, 61.4 and 67 for sediments from the Plain's bed-load, active flood-plain and older flood-plain from the inter-fluve region indicates that silicate weathering of Ganga River sediments has occurred only after entering into the plains. This is likely because of higher residence time and change in the climate from cold-frigid in the Himalaya to tropical sub-humid in the plains. Therefore, the use of geochemical data on ancient system to infer climate in their source region may not always be true. Although the CIA values indicate a moderate chemical weathering in the plains, it is far from impressive. Dominance of physical weathering in the catchment region and lower degree of chemical weathering in the Plains indicate that weathering of sediments supplied by Himalayan Rivers, particularly the Ganga River may not have affected the atmospheric CO_2 to a significant level as is generally believed. Thus the net effect of the Himalaya on the CO_2 sequestration and consequent global cooling needs a re-evaluation.

The plots of sediments in ternary diagram among La, Th, Sc and ratios involving Co/Th, La/Sc and Sc/Th indicate granitic to granodioritic source rocks to the sediments. The ratio plots involving relatively immobile Al_2O_3, TiO_2 and FeO along with REE plots suggest that out of the major Himalayan lithologies, gneisses and Cambro-Ordovician granites of HHCS have acted as the dominant source to the sediments.

The plots of $LogNa_2O/K_2O$ vs. $LogSiO_2/Al_2O_3$ and FeO/SiO_2 vs. Al_2O_3/SiO_2 diagrams show that the combination of processes including erosion, weathering, sorting and aeolian activity has together played a major role in progressively changing the chemistry from source rock to catchments bed-load to Plains bed-load, active flood-plains and the older inter-fluve sediments in the Ganga River system. The above plots demonstrate that as a result of above processes the ratios between the elements generally thought to be immobile and used in provenance studies does not always remain invariant and the linear trend line in the scatter gram between the two immobile elements show rotation around the fine grained end member.

1. Introduction

The chemical record of clastic sediments has been widely used for the determination of provenance, tectonics and weathering in the source region (Bhatia, 1983; McLennan and Taylor, 1983; Taylor and McLennan, 1985; Wronkiewicz and Condie, 1987; Cullers, 1988; Feng and Kerrich, 1990; Condie et al., 1992; Garver and Scott, 1995; Fedo et al., 1996; Nesbitt and Young, 1996; Holail and Moghazi, 1998; McCann, 1998; Singh and Rajamani, 2001a,b; Tripathi and Rajamani, 2003). Particularly the use of immobile major and trace elements which are thought to be carried in the particulate load, such as Al, Fe, Ti, Th, Sc, Co, Zr and the rare earth elements (REEs), have been found to be useful indicators of the source (Taylor and McLennan, 1985). The use of these immobile elements in provenance determination is based on the assumption that these elements undergo little geochemical fractionation during denudational processes. The immobile elements such as Al, Fe, Ti, Th, Sc, Co, Zr and the REEs are commonly concentrated in the fine grained sediments because their host minerals including accessory primary minerals and secondary minerals occur in that size range. Therefore during the process of sediment transport and deposition, these immobile elements tend to concentrate in suspended load of the river leaving the bed-load sediments depleted in them with higher proportions of quartz and feldspars. Because of the size and mineral fractionation, geochemical fractionation is expected.

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In a binary diagram of two immobile elements or their ratios, it is expected that the plots for finer sediments and coarser sediments will move away in opposite direction and the tie lines joining them will pass through their common source (Fralick and Kronberg, 1997). However, many post depositional changes such as weathering, pedogenesis and diagenesis could modify the source, coarse and fine grain tie-line to different extent, depending on the extent of mobility of certain elements under the changed environmental conditions of deposited sediments (Sawyer, 1986; Taylor and McLennan, 1985; Milliken and Mack, 1990; Milliken et al., 1994; Fralick and Kronberg, 1997; Duddy, 1980; Suttner et al., 1981; McLennan, 1989; Cullers and Stone, 1991; Berner, 1992; Stallard, 1992; Johnsson, 1994; White and Blum, 1995; Nesbitt and Young, 1996). For a given group of young sediments of a river system, any large deviation from the tie line may imply either a selective chemical fractionation or multiplicity of source to the sediments. Therefore identification of group of elements which has remained immobile in a given group of sediments is essential in the study of sediment provenance (Fralick and Kronberg, 1997).

The Ganga River is a classic river with vast terraced flood-plains of different ages in the middle and lower reaches and has been receiving sediments from the Himalaya, a tectonically active highland, at least from the Cenozoic onwards (Dewey and Bird, 1970; France-Lanord et al., 1993; Hodges, 2000). It encompasses areas that are diverse both in terms of relief and climate. The sediments carried and deposited by it have varied residence time in different parts of the river and are exposed to different climate regimes (cold and frigid in the highlands to humid and sub-tropical in the Plains). Thus, the Ganga River provides an opportunity to study the effect of en-route climate change and various processes on sediment chemistry, which has significance for the use of sediment chemistry as a proxy in interpretation of source region climate in ancient rocks.

There are few studies carried out on the geochemistry of Ganga River sediments which include the study of sediments from the lower reaches with the intention of understanding the erosion rates in the Himalayas (Galy and France-Lanord, 2001). Some recent studies have been conducted on channel, suspended-load and flood-plain sediments (Ramesh et al., 2000; Tripathi et al., 2007) to understand the effect of weathering, sediment source and recycling on the sediments. These studies are based on data-bases generated from samples collected from limited or randomly scattered locations without any data on the precursor sediment from the source region to evaluate the changes that occurred in the Plains. In the present study I discuss the texture, mineralogy, and geochemical characteristics (major, trace and REEs) of the entire spectrum of sediments deposited by the Ganga River. This study encompasses sediments from the source region in the Himalayas to different depositional regimes in the Plain. This study consists of a large number of samples providing a wider database from all the possible locations and regimes. This enables us to critically evaluate the effect of weathering and different fluvial processes on the sediment's chemistry in different parts of the Ganga River and also to assess and identify the group of elements suitable for provenance identification. This study also provides some information on geochemical differentiation by surface geological processes during transport and deposition and also investigates post-depositional changes in the sediment chemistry due to an en-route change in the climate from source to the Plains.

The collision of Indian plate with the Eurasian plate ~50 Ma resulted in uplift and rapid erosion of the Himalaya. The volume of material eroded after the beginning of collision is thought to have resulted in reduction of atmospheric CO_2 in Cenozoic (Raymo and Ruddiman, 1992; Raymo et al., 1988; Molnar et al., 1993; Edmond, 1992; Singh et al., 1998; Krishnaswami et al., 1992) leading to global fall in temperature. Other workers (Pearson and Palmer, 1999; Palmer and Edmond, 1992) find no dramatic change in atmospheric CO_2 during the same period. The above conclusions are based on different estimates made for the relative proportions of Himalayan Sr fluxes derived from the carbonate and the silicate sources. All the above studies are based upon the solute chemistry and have not involved the particulate load carried by the rivers which are thought to hold the signature of the extent of chemical weathering of the source. The present study on the sediments collected from different locations in mountainous tract and the Plains, also aims at determining the nature and extent of chemical weathering experienced by the silica-clastic sediments in different parts of the Ganga River, to infer its role in past climate change related to the Himalayan uplift.

2. Geology and geomorphology

The Ganga River coming out of the foothills at Haridwar is formed as a result of joining of several prominent tributaries, namely the Bhagirathi, Alaknanda and Mandakini, flowing in the hilly tract through rocks of the Higher Himalayan Crystalline Series (HHCS), Lesser Himalayas Series (LHS) and the Siwaliks also known as the Siwalik foreland basin. All of these are at different elevations and have different relief (Valdiya, 1980). All the major tributaries originate in the Higher Himalayas, which are permanently snow capped and form extremely rugged mountainous ranges with high relief. The river gradient is very high in this part of the mountains, rising from an elevation of 3000 m to more than 8000 m. South of this is the LHS, which is a gentle undulating terrain with the elevation ranging from 600 to 2500 m. The mountains in this part are rounded, having gentle slopes and undulating topography. South of the LHS is the Siwalik range, 250 m to 800 m high, which form the southern front of the Himalaya. These tributaries join at various places in this region finally forming the main trunk of the Ganga River at Devprayag. The total catchment area of the Ganga River is 7811 km^2, out of which 2328 km^2 is snow bound.

On coming out of the hilly tract the Ganga River flows in a S–E direction almost through the central part of the Gangetic Plain and forms the major trunk channel crossing the Ganga Plain and is joined by a number of tributaries on its way to the Bay of Bengal. The Ganga Plain is an active foreland basin formed as a result of continent–continent collision of the Indian plate with the Eurasian plate. The Ganga River passes through a wide range of climatic variation, from cold and frigid to sub–humid, from north to south in the Himalaya. The climate in the Plain region is humid sub-tropical and is divided into three seasons: the cold season (November to February), the hot season (March to mid-June) and the monsoon season when it receives the maximum rainfall (mid-June to October).

The Plain region has been differentiated into several geomorphic surfaces as a result of river incision during different times in response to neotectonic activity during the late Quaternary (Singh, 1996). These geomorphic surfaces are depositional surfaces and are associated with almost all the rivers on the Ganga Plain. Accordingly, the surfaces along the Ganga River are identified as: (i) Upland terrace surface (T2) which forms the upland inter-fluve surface beyond the reach of the floods of the present Ganga River (the sediments of this surface are generally fine-grained and at places show prominent development of calcretes); (ii) the river valley terrace (T1) which is situated slightly above the present day active channel and is flooded only during the high flood events; (iii) the T0 surfaces, which are flooded almost every year; and (iv) the channel, which contains sediment bars is here-after referred as bed-load sediments. The study area comprises the upper and middle part of the Ganga River in the Plains and also refers to Ganga and its tributaries in the upper reaches in the Himalayas catchment region. In the present study I have grouped the sediments in accordance with the surfaces from which the samples were collected in the Plain region, i.e. the older flood-plain sediments of T2 surface are grouped as 'inter-fluve sediments'; Sediments sampled from T1 and T0 surfaces which still get flooded are together grouped

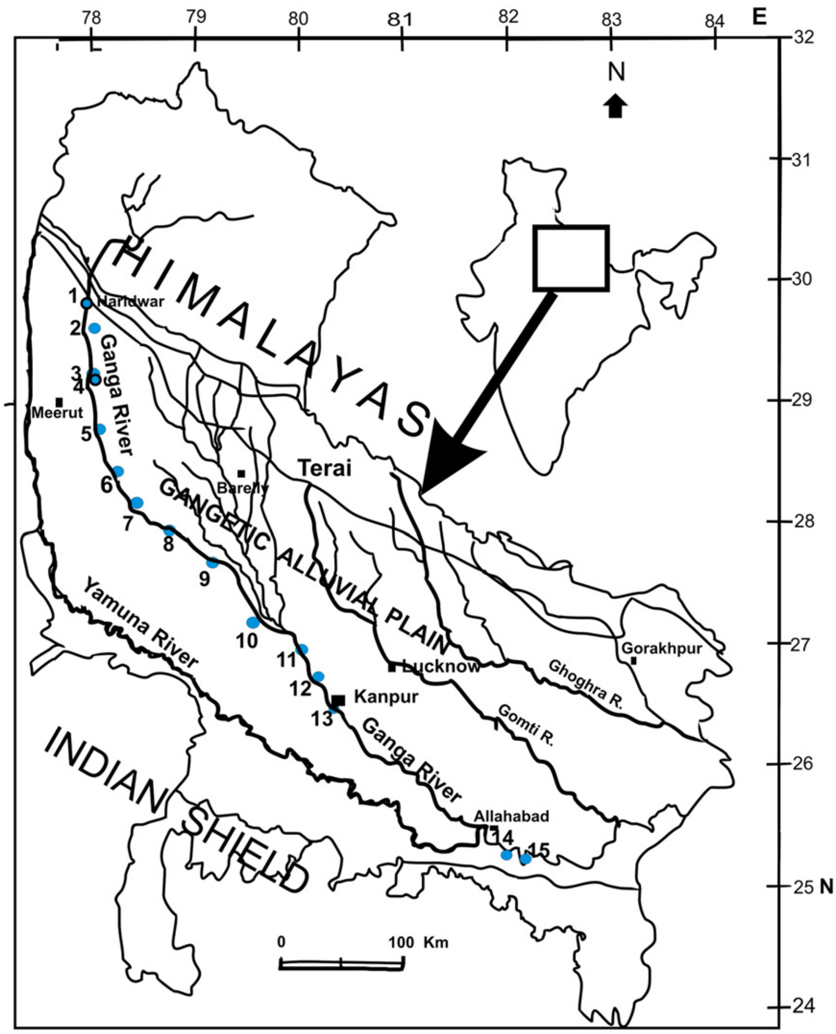

Fig. 1. Simplified map of Gangetic plain (modified from Singh, 1996) showing sampling sites (1–15) along Ganga River. For detailed sample nomenclature and locations refer Table 1.

as 'active flood-plain sediments' and channel sediments sampled are grouped as 'bed-load sediments'.

3. Sampling and analytical methods

Systematic sampling of sediments from various geomorphic surfaces was done from a total of fifteen locations along the Ganga River for a distance of about 800 km from Haridwar to Mirzapur (Fig. 1). The samples and their locations are listed in Table 1. The interfluve sediments (T2 surface) were collected from vertical profiles of several meter thickness along the channel which was exposed due to incision of this surface by the present day channel. Similarly active flood-plain sediments, T1, were collected from the profile cut into this surface and T0 sediments were collected from the flood deposits on the margin of the present day channel. Approximately 2 to 3 kg of sediments were collected from a meter-long groove made after cleaning the exposed surface in the flood-plain. The samples were analysed for texture, mineralogy and geochemistry including REEs on selected samples (Supplementary Table 2 and Table 2).

The samples were dried in an oven at 70 °C for approximately 24 h and then homogenized. One hundred grams of each homogenized sample was ground to −200 mesh (0.075 mm) size in an agate mortar for geochemical and XRD mineralogical study. Grain size was measured following the standard pipetting method (Gale and Hoare, 1991) for the silt and clay fraction after treating 50 g of the sample with cold 1 M HCl and H_2O_2 to remove carbonates and organics. Deflocculating the sample was accomplished by adding sodium hexametaphosphate, and the sand was separated by wet sieving. The size fractions above 0.063 mm were determined by dry sieving. Mineral identification was performed using a Phillips XRD (PW1140). Heavy minerals were separated using bromoform and identified under a binocular microscope. Clay minerals in the clay fraction, separated by the Atterberg method (Müller, 1967), were identified by X-ray

Table 1
Sampling locations with corresponding sample ID for ~800 km stretch of Ganga River in the plain region.

Location no.	Location name	Latitude	Longitude	Sample ID
1	Haridwar	nd	nd	G1B, G1AFP
2	Nagal	29° 40′	78° 12′	G2B, G2IF
3	Bijnor	nd	nd	G3B, G3AFP1, G3AFP2, G3IF
4	Doranagar	29° 19′	78° 5′	G4B, G4AFP
5	Tigri	28° 49′	78° 9′	G5B, 5AFP, G5IF
6	Anupsahar	28° 21′	78° 16′	G6AFP1, G6AFP2
7	Naurora	28° 12′	78° 23′	G7AFP
8	Soron	27° 55′	78° 52′	G8B, G8AFP
9	Kampil	27° 37′	79° 17′	G9AFP
10	Fathegarh	27° 22′	79° 38′	G10B, G10AFP, G10IF
11	Kannauj	27° 3	79° 56′	G11B, G11IF
12	Bilhaur	26° 51′	80° 4′	G12B, G12AFP
13	Kanpur	26° 27′	80° 20′	G13B, G13AFP, G13IF
14	Manda	25° 10′	82° 14′	G14B, G14AFP1, G14AFP2, G14IF
15	Mirzapur	25° 9′	82° 34′	G15B, G15IF1, G15IF2

Table 2
Rare earth element (ppm) composition of the Ganga River plain sediments from various locations and depositional regime, along with various elemental ratios.

Bed-load sediments								Active flood-plain sediments												Interfluve sediments				
	G3B	G4B	G8B	G10B	G12B	G14B	G15B	G3AFP1	G3AFP2	G4AFP	G5AFP	G6AFP1	G6AFP2	G7AFP	G10AFP	G12AFP	G14AFP1	G14AFP2	G3 IF	G10 IF	G14 IF	G15 IF 1	G15 IF 2
	Bijnor	Doranagar	Soron	Fatehgarh	Bilhaur	Manda	Mirzapur	Bijnor	Bijnor	Doranagar	Tigri	Anupsahar	Anupsahar	Naurora	Fatehgarh	Bilhaur	Manda	Manda	Bijnor	Fatehgarh	Manda	Mirzapur	Mirzapur
La	19.4	26.0	31.8	34.0	17.9	21.3	10.2	25.2	31.2	27.6	27.1	31.7	33.2	35.4	37.4	40.8	21.3	32.6	35.0	28.0	27.3	28.4	23.6
Ce	38.1	55.5	63.5	62.5	35.5	40.9	18.2	46.7	63.2	55.1	54.3	61.3	64.0	68.4	78.3	81.6	40.9	65.2	70.4	55.9	57.5	57.8	48.3
Nd	15.6	24.0	28.5	27.8	14.5	19.7	8.9	21.0	29.0	24.0	25.5	27.8	27.3	29.6	32.4	34.1	19.7	30.7	30.8	23.6	24.0	26.4	22.1
Sm	4.5	5.1	5.9	6.9	4.2	3.4	1.9	5.1	5.4	5.3	5.1	5.8	7.2	6.7	7.0	8.6	3.4	6.0	5.6	6.1	5.1	6.0	4.2
Eu	0.6	0.9	0.8	0.9	0.5	0.5	0.2	0.6	0.8	1.0	0.7	0.9	1.0	0.9	1.2	1.1	0.5	0.9	0.9	0.9	0.8	0.8	0.5
Gd	3.0	4.2	5.0	5.1	2.7	3.3	1.6	4.4	5.4	4.5	5.1	5.0	5.1	5.0	6.2	6.4	3.3	6.0	6.2	4.7	5.2	5.0	4.1
Dy	2.6	3.6	4.4	4.5	2.6	3.0	1.7	3.6	4.9	3.9	4.2	4.4	4.8	4.5	5.2	5.5	3.0	4.7	5.1	3.9	4.2	4.5	3.4
Yb	1.6	2.2	2.7	2.5	1.6	1.4	0.6	1.7	2.3	2.2	2.1	2.6	2.8	2.6	3.0	3.1	1.4	2.3	2.8	2.2	2.0	2.1	1.5
(La/Yb)*n*	8.2	7.9	8.1	9.1	7.8	10.4	11.2	10.2	9.1	8.7	8.8	8.4	8.1	9.1	8.4	9.0	10.4	9.7	8.5	8.6	9.4	9.2	10.7
(La/Sm)*n*	2.7	3.2	3.4	3.1	2.7	3.9	3.4	3.1	3.6	3.3	3.3	3.4	2.9	3.3	3.3	3.0	3.9	3.4	3.9	2.9	3.4	2.9	3.5
(Gd/Yb)*n*	1.5	1.5	1.5	1.6	1.4	1.9	2.1	2.1	1.9	1.7	2.0	1.6	1.5	1.5	1.7	1.7	1.9	2.1	1.8	1.7	2.1	1.9	2.2
Eu/Eu*	0.52	0.58	0.46	0.45	0.48	0.43	0.32	0.41	0.47	0.59	0.44	0.52	0.48	0.50	0.55	0.46	0.43	0.44	0.45	0.53	0.49	0.46	0.37

Number after first letter of sample corresponds to location number given in map.

diffraction after mounting the samples on glass slides and running the untreated, glycolated, and heat-treated (at 550 °C) samples. Homogenized powders of sediment samples were digested following the open beaker acid digestion procedure using HF, HNO_3, and $HClO_4$, and analyzed for major and trace elements. The samples for REE analysis were digested by the NaOH and Na_2O_2 fusion method in nickel crucible. REE separation and preconcentration was done using HNO_3 and HCl cation exchange resin (Bio-Rad AG50-X8) columns (for details see Singh and Rajamani, 2001b). All the elements were analysed using a JY ICP-AES (ULTIMA 2) using international and laboratory working standards. SiO_2 was determined by a Systronics spectrophotometer following the method of Shapiro and Brannock (1962). Elemental analyses have a precision and accuracy better than 5% and 2% respectively. For low REE samples (<5X chondrite) the precision was 5–10% for Ce and Nd. Concentrations of Zr, Th, Sc, Co, and U on representative samples were analyzed by the XRF method (Siemens SRS3000) at Wadia Institute of Himalayan Geology, Dehradun.

4. Results

4.1. Texture and mineralogy

The channel sediments of the Ganga River consist almost entirely of sand sized particles with little silt and almost no clay. On average it is composed of 89% of sand-sized particles, 10% of silt-sized particles and <1% of clay particles. The channel sediments have a mean size falling into the very fine sand size (2–3.5Φ) right from Haridwar at the base of the Himalayas (Supplementary Table 2), which suggests that the fine size of sediments is not only due to the degree of transport but is also because of the inherently fine-grained nature of the grains in the source rock itself. The active flood-plain sediments predominantly contains silt-sized particles and on an average contain 27% of sand-sized particles, 67% of silt sized particles and 6% of clay sized particles and have mean grain sizes ranging between 3.5Φ and 5.5Φ . The samples from older flood-plain surfaces also contain predominantly silt-sized particles with little clay content and a small amount of sand. In general the flood-plain sediments are finer than the bed-load sediments but the bed-load sediments are also not very coarse grained. The presence of all size fractions in various flood-plain sediments indicates their unsorted nature. During flooding due to the high energy state of the stream water, part of the sand from the channel bed is also taken into suspension along with the normal suspension population, comprising silt and clay, and is together deposited as overbank sediments resulting from the overtopping or breaching of the bank at the time of flooding. In general clay was found to be less in all the sediment samples analyzed. Less clay contents suggest that there has been little chemical weathering in the source region. The channel bed-load sediments are good to moderately well sorted, whereas the flood-plain sediments are poorly sorted.

The minerals in the bed-load and flood-plain sediments include quartz, plagioclase, K-feldspar, muscovite and biotite as major phases and garnet, zircon, monazite as heavy minerals. The bed-load contains a higher relative percentage of quartz compared to the flood-plain sediments. Although mineral proportions may vary between bed-load and flood-plain sediments, the general mineralogy of the sediment is not found to vary either longitudinally or laterally along the channel. The clay minerals identified by X-ray diffraction are dominantly illite followed by chlorite, smectite and kaolinite in order of decreasing abundance.

4.2. Geochemistry

Geochemical composition (major and trace elements) of sediments of the Ganga River in the Plains region, along with ratios of important elements are shown in Supplementary Table 2. The numbers stated after the first letter of the sample correspond to their locations as given on the

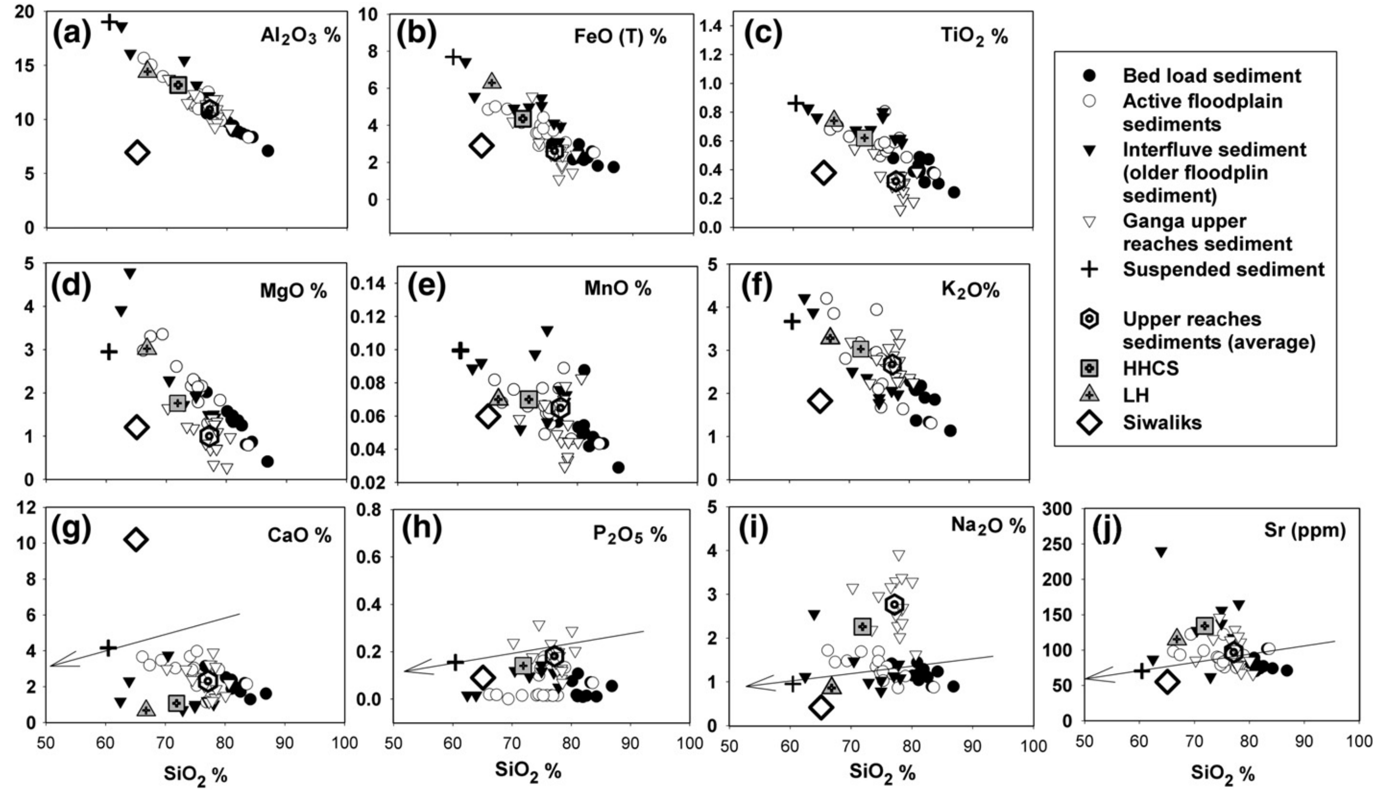

Fig. 2. Variation diagram of major and trace elements against % SiO_2 for all the group of sediments studied, along with average composition of suspended-load, bed-load in catchment region and the source rock (Galy and France-Lanord, 2001). a–f represent plot for elements which concentrate in fines and tend towards 100% SiO_2; g–j represent plot for elements which concentrate in coarse fraction and tend towards 0% SiO_2.

map (Fig. 1). REE data for the above set of sediments are given in Table 2. For comparison is given the geochemical composition of the three catchment region sediments of the Ganga River after the confluence of all its major tributaries (Table 4). The samples studied have been categorised into three groups: (1) bed-load sediments (mostly sandy), (2) active flood-plain sediments from the T0 and T1 surfaces (which are overtopped by present day flooding), and (3) inter-fluve sediments from T2 surfaces which no longer receives sediments from the present river (these are sediments ranging in age from Pleistocene to several thousand years before present, Srivastava et al., 2000, 2003). In general

Table 3
Correlation coefficient among textural and chemical parameters for the entire group of sediments of Ganga River.

	SiO_2	Al_2O_3	TiO_2	FeO	MgO	MnO	CaO	K_2O	Na_2O	P_2O_5	Mz	Ba	Sr	Ni	Zr	Sc	Co	Th	U	La	Ce	Nd	Sm	Eu	Gd	Dy	Yb
SiO_2	1.0																										
Al_2O_3	−1.0	1.0																									
TiO_2	−0.8	0.9	1.0																								
FeO	−0.9	0.9	0.9	1.0																							
MgO	−0.9	0.8	0.7	0.8	1.0																						
MnO	−0.6	0.7	0.7	0.7	0.5	1.0																					
CaO	−0.4	0.1	0.2	0.2	0.4	0.0	1.0																				
K_2O	−0.8	0.7	0.5	0.6	0.8	0.4	0.3	1.0																			
Na_2O	−0.4	0.3	0.2	0.1	0.6	0.1	0.2	0.4	1.0																		
P_2O_5	0.1	−0.1	0.1	0.1	−0.2	−0.1	0.3	−0.4	−0.1	1.0																	
Mz	−0.9	0.8	0.8	0.8	0.8	0.6	0.2	0.8	0.3	−0.2	1.0																
Ba	−0.7	0.7	0.6	0.7	0.6	0.4	0.3	0.6	0.3	−0.1	0.6	1.0															
Sr	−0.5	0.4	0.5	0.5	0.5	0.3	0.4	0.2	0.3	0.2	0.4	0.4	1.0														
Ni	−0.6	0.8	0.7	0.7	0.4	0.7	−0.3	0.4	0.0	−0.2	0.6	0.4	0.2	1.0													
Zr	−0.5	0.6	0.7	0.5	0.3	0.6	−0.3	−0.1	0.5	−0.1	0.6	0.4	0.0	0.6	1.0												
Sc	−0.9	0.9	0.9	0.9	0.7	0.8	0.1	0.4	0.0	0.0	0.7	0.7	0.7	0.7	0.5	1.0											
Co	−0.8	0.8	1.0	0.9	0.4	0.8	−0.1	0.1	−0.2	−0.1	0.6	0.7	0.7	0.8	0.5	0.9	1.0										
Th	−0.5	0.7	0.5	0.5	0.2	0.6	−0.5	0.0	0.3	−0.2	0.4	0.3	−0.3	0.7	0.8	0.4	0.4	1.0									
U	−0.6	0.8	0.5	0.5	0.5	0.5	−0.3	0.2	0.4	−0.5	0.6	0.5	−0.2	0.6	0.7	0.4	0.4	0.8	1.0								
La	−0.6	0.6	0.6	0.5	0.6	0.5	0.2	0.5	0.3	0.0	0.6	0.5	0.1	0.6	0.6	0.6	0.4	0.6	0.6	1.0							
Ce	−0.7	0.7	0.6	0.6	0.6	0.6	0.2	0.5	0.3	0.0	0.7	0.6	0.2	0.6	0.5	0.7	0.4	0.6	0.6	1.0	1.0						
Nd	−0.7	0.7	0.6	0.6	0.6	0.5	0.3	0.4	0.3	0.1	0.7	0.6	0.2	0.6	0.5	0.6	0.4	0.6	0.5	1.0	1.0	1.0					
Sm	−0.5	0.5	0.5	0.4	0.5	0.4	0.1	0.3	0.4	−0.1	0.5	0.4	0.1	0.5	0.6	0.7	0.5	0.7	0.7	0.9	0.9	0.9	1.0				
Eu	−0.6	0.6	0.5	0.4	0.6	0.5	0.2	0.5	0.5	0.0	0.5	0.4	0.1	0.5	0.5	0.7	0.4	0.6	0.6	0.9	0.9	0.9	0.9	1.0			
Gd	−0.8	0.7	0.8	0.7	0.7	0.7	0.3	0.5	0.3	0.1	0.8	0.6	0.3	0.6	0.6	0.8	0.6	0.6	0.6	0.9	1.0	1.0	0.8	0.9	1.0		
Dy	−0.7	0.7	0.7	0.7	0.7	0.6	0.2	0.5	0.3	0.0	0.7	0.6	0.2	0.6	0.5	0.7	0.5	0.6	0.5	1.0	1.0	1.0	0.9	0.9	1.0	1.0	
Yb	−0.6	0.6	0.5	0.5	0.6	0.5	0.1	0.5	0.4	−0.1	0.6	0.5	0.0	0.5	0.5	0.5	0.3	0.6	0.5	1.0	1.0	0.9	0.9	0.9	0.9	0.9	1.0

the bed-load sediments show higher SiO_2 concentration and lower concentration of all other elements compared to active flood-plain and inter-fluve sediments (Fig. 2), which, is because of their higher quartz percentage. The bed-load sediments show no particular downstream variation in elemental concentration and appear to be controlled by mean grain size of the sediments. The flood-plain sediments of both groups do not show any secular trend with respect to distance from the source and their chemistry remains more or less similar for the entire stretch sampled. A similar observation was also reported for the flood-plain sediments of the Kaveri River by Singh and Rajamani (2001a). The flood-plain sediments have higher Al_2O_3, FeO, MgO, MnO, TiO_2 and K_2O compared to the bed-load sediments. This is a consequence of the higher quartz percentage in the bed-load sediments and a higher content of aluminous minerals, such as mica and clays, in the active flood-plain and the inter-fluve sediments, as reflected in the linear trend from bed-load to flood-plain and inter-fluve sediments in Fig. 2. This is also reflected in the strong positive correlation amongst the above group of elements, and their negative correlation with silica (Table 3). However Na_2O, CaO, P_2O_5 and Sr show different patterns.

Moderately good correlation between Ba and K, and of both with Al_2O_3, FeO, MgO, MnO and TiO_2 suggests that the distribution of K and Ba are mainly controlled by clay (probably illite) and mica. Strong correlation of mean grain size (Mz) with Al_2O_3, FeO, MgO, MnO, TiO_2, K2O, Ni, Ba indicates grain size control on these elements and also suggests that the fine sediments have a higher percentage of mica and clay minerals. Poor correlation of Na_2O with mean grain size and SiO_2 suggests that feldspars have no preferred grain size.

4.2.1. Rare earth elements (REEs)

All three groups of sediments (bed-load, active flood-plain and older flood-plains/inter-fluve) have similar chondrite-normalised REE pattern with overlapping abundances (Fig. 3a, b, c). The average REE concentration shows an increase in order from bed-load to active flood-plain to inter-fluve although within a narrow range. All groups of sediments show fractionated, parallel to sub-parallel REE patterns, with $(La/Yb)_N$ ratios ranging between 7.8 and 11.1 (Table 2), with averages of 8.80, 8.97, and 9.14 for bed-load, active flood-plain and inter-fluve sediments respectively. Both LREE and HREE show variable fractionation with $(La/Sm)_N$ values ranging between 2.6 and 3.9 with averages of 3.1, 3.26, and 3.29 for bed-load, active flood-plain and inter-fluve sediments respectively. The $(Gd/Yb)_N$ value ranges between 1.4 and 2.2 with averages of 1.61, 1.75, and 1.92 for bed-load, active flood-plain and inter-fluve sediments respectively. This indicates that although the REE content among various groups of sediments vary within a narrow limit, their fractionation increases in order from bed-load to active flood-plain to inter-fluve sediments mainly because of minor increases in both $(La/Sm)_N$ and $(Gd/Yb)_N$ ratios of the same order. All the samples show consistently uniform negative Eu anomalies with $(Eu/Eu^*)_N$ ratios ranging between 0.32 and 0.59. For comparison and to facilitate interpretation, the average REE pattern of the sediments from different parts of the mainstream Ganga River in the Himalayan region, after the confluence of all its tributaries and before entering into the Ganga Plain (Table 4) are also plotted (Fig. 3). It is interesting to note that the REE patterns of the sediments from the Ganga River in the Himalayan region are similar to the REE patterns of the sediments from the Plain region including the Eu anomalies.

The PAAS (Taylor and McLennan, 1985) normalised REE patterns of average of bed-load, active flood-plain and inter-fluve sediments from the Ganga Plain and the sediment from the Himalayan region are plotted in Fig. 4. A significant feature of the shale normalised pattern is the depletion of Eu in all the groups of sediment, and the similarity of the REE patterns of all the groups of sediments from the Plain region with the sediments from the Himalayan region of the Ganga. One thing to note is that the bed-load in the Plains region has distinctly lower abundances of REEs unlike the bed-load sediments from the Himalayan region, which show abundances similar to the flood-plain and inter-fluve sediments (Fig. 4). This indicates that the bed-load sediments from the Himalayan region are texturally and mineralogically immature and efficient size separation has not taken place as they are just the ground up debris from the source rock. This is evident from the similar patterns and abundances of REEs in bed-load and suspended-load sediments from this region. This further indicates that the inter-granular and mineral separation of Ganga River

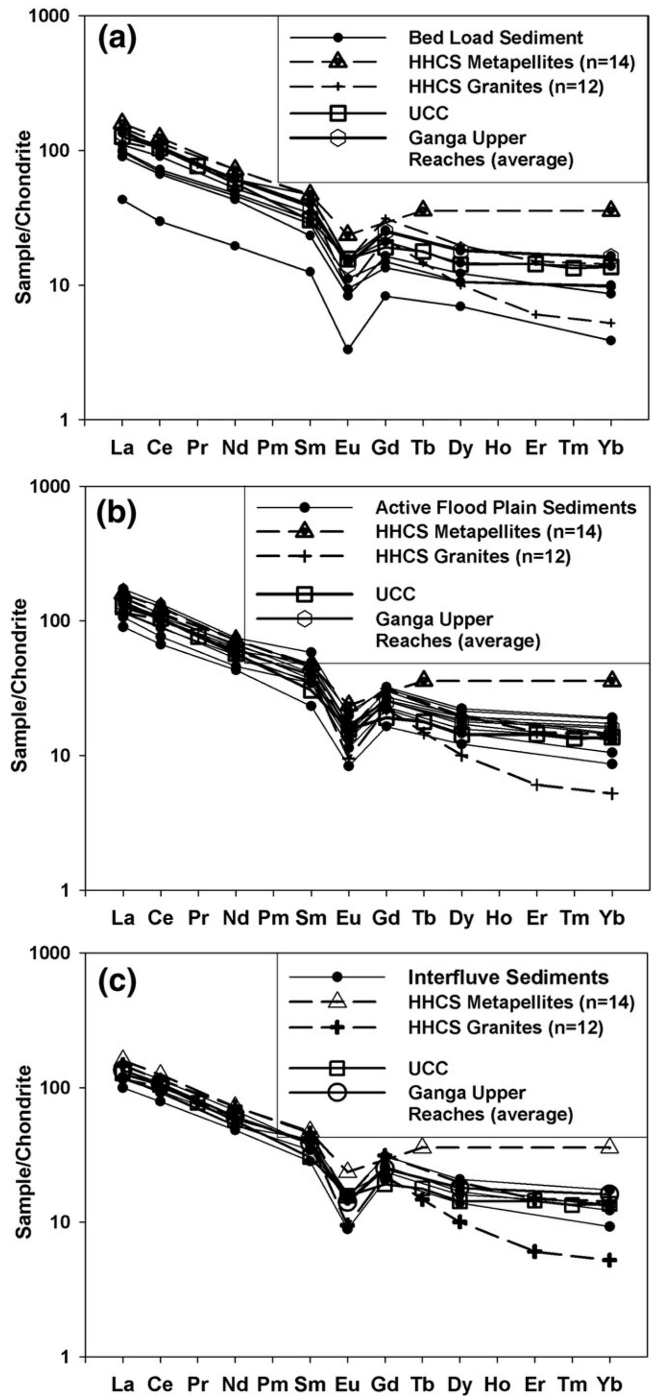

Fig. 3. Chondrite (McDonough and Sun, 1995) normalised REE patterns of the Ganga river sediments from different depositional regime in the plains. For comparison average of three samples from Ganga in upper reaches after confluence of all the tributaries are also plotted along with the averages of two probable source rocks from HHCS [metasediments (Harris et al., 1992; Ayres and Harris, 1997), Cambro-Ordovician granites(Miller et al., 2001)]. Note the uniformity in REE pattern for all sediments in the plain region and its similarity with sediments from upper reaches of Ganga before entering into the plains.

Table 4
Major-element (in wt.% oxide) and trace-element and rare earth element (ppm) composition of selected sediments from the Ganga River in the Himalayan cathment region from various locations after the confluence of all its major Tributaries at Devprayag and before Haridwar along with their CIA and Mean Size (Mz).

	G1	G2	G3		G1	G2	G3
SiO_2	77.66	77.38	78.41	Co		8	
$Al2O_3$	9.81	9.87	9.67	Th		12	
TiO_2	0.36	0.31	0.30	U		1	
FeO	2.68	2.55	2.36	Zr		201	
MgO	1.41	1.31	1.34	Y	29	28	24
MnO	0.07	0.07	0.05	La	35.3	34.0	26.3
CaO	3.24	3.24	3.11	Ce	74.0	70.2	52.2
Na_2O	2.28	2.52	2.34	Nd	31.1	29.5	22.5
K_2O	2.39	2.58	2.29	Sm	6.4	6.0	4.6
P_2O_5	0.12	0.15	0.12	Eu	0.8	0.9	0.7
CIA	48.4	46.2	47.9	Gd	5.7	5.0	4.2
Mz	2.6	3.0	2.7	Dy	4.9	4.5	3.9
				Yb	2.9	2.5	2.3
Ba	392	447	406	Lu	0.4	0.4	0.3
Sr	101	129	111	$(La/Yb)_n$	8.2	9.1	7.8
Cr	21	73	60	$(La/Sm)_n$	3.5	3.5	3.6
Ni	10	21	19	$(Gd/Yb)_n$	1.6	1.6	1.5
Sc		7	19	Eu*/Eu	0.4	0.5	0.5

sediments is achieved only after it enters the Plains region. Thus, to a first approximation, the uniformity of REE patterns of the sediments from the Himalayan region suggests that the sediments are thoroughly homogenised even before entering the Plains, and the textural control on REE abundance becomes effective only after entering into the Plains. Increasing degree of transport, with greater separation of individual minerals, makes the sorting more effective.

Moderate to strong positive correlation of REEs with Al_2O_3, FeO, MgO, MnO, TiO_2, Ni, Ba and the mean grain size, along with Th and Zr (Table 3), indicates alumino-phyllosilicates (mainly clay minerals and micas) and accessory phases such as zircon and monazite are holding the REEs, and their sorting has controlled its abundances in different groups of sediments. This is in contrast to the poor correlation observed between Al_2O_3, FeO, MgO, MnO, TiO_2, Ni, Cr and the REEs in the sediments from the various parts of the Ganga River and its tributaries in the catchment region in the Himalayas up to Haridwar as observed by the author in another study. In the upper reaches, REEs have a strong correlation only with Th, P_2O_5 and Zr (monazite, apatite, zircon). In the Plains region P_2O_5 looses its correlation with REEs. This would suggest that part of the apatite and/or monazite containing REEs in the upper reaches have weathered after entering into the Plain region and their REEs have been released and taken into the clays and other phyllosilicates. It is interesting to note that even after this exchange of phases the REE pattern of sediments from the catchment and the Plain remain similar (Figs. 3 and 4). This would indicate that weathering has resulted neither in loss nor fractionation of REEs in the Plains.

5. Discussion

5.1. Provenance

Previous studies on the Ganga Plains, mostly based on bed-load and suspended-load composition (Galy and France-Lanord, 2001), have suggested that HHCS form the major source to the sediments and Lesser Himalayas contribute only between 10 and 20% to the sediment load. Whereas, Tripathi et al. (2007) have suggested the Ganga Plain is similar to the Siwaliks and it is its Quaternary extension. These studies were based on either small number of samples or samples collected from a few or randomly scattered locations. Here I re-examine the source for the sediments and the suitability of different elements in the source terrain using a larger data set.

Some elements, and particularly their ratios, are useful indicators of provenance as they are least effected by processes such as weathering, transport and sorting. In particular the commonly immobile elements Al, Fe, Ti, Th, Sc, Co, Cr, REEs and their ratios have been found to be useful (Taylor and McLennan, 1985). These elements even if mobilised have short residence time in water and hence are almost quantitatively transferred into the sediments.

To understand the chemical change experienced by the sediments during the process of erosion, transportation and after deposition, it is necessary first to evaluate element mobility in the given set of sediments. This helps us in separating the changes brought by physical process of sorting from that due to chemical weathering. Fralick and Kronberg (1997) have put forward a graphical method for determining element mobility by means of binary diagrams. This method allows identification of immobile elements in a given set of sediments and is based on the premise that plots of pair of immobile elements hosted in common mineral or minerals having similar hydrodynamic behaviour from sediment suite derived from common source will form a linear array of coordinates pointing to the origin on binary diagrams. Any large deviation from the above may imply either selective chemical fractionation or multiplicity of source to the sediments.

To test for immobile elements in the studied group of sediments, pairs of elements suspected to be immobile, e.g., Al_2O_3, TiO_2, FeO, and

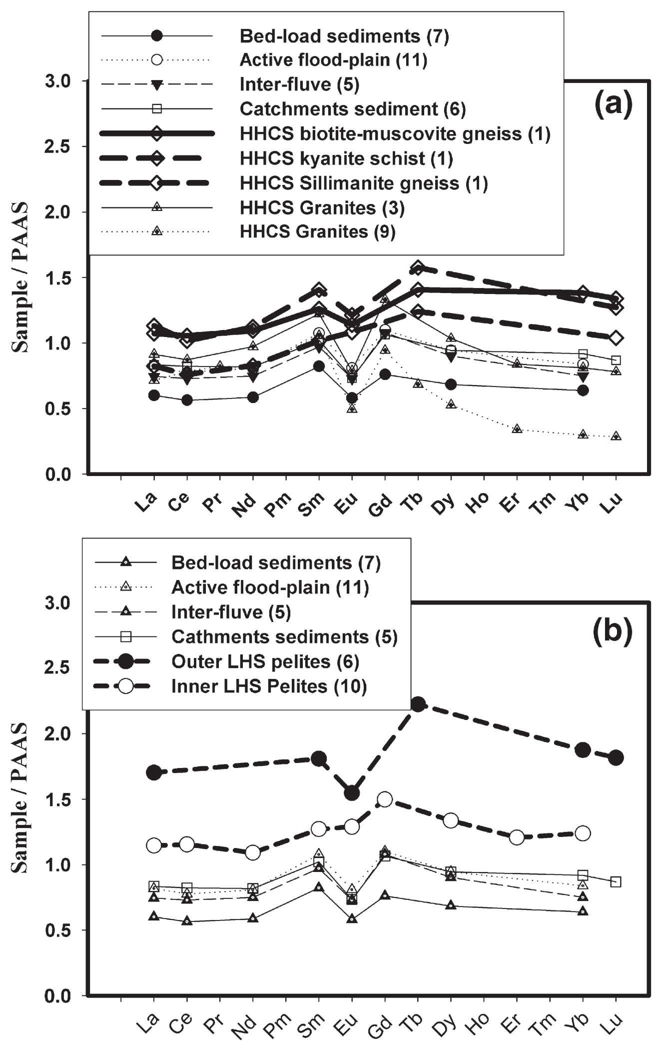

Fig. 4. Averages of rare earth element concentrations from different depositional regimes from the Ganga River plain normalised to PAAS (Taylor and McLennan, 1985) along with the plot for sediments from the upper reaches of Ganga River and (a) the HHCS source rocks (HHCS biotite–muscovite gneiss- Ayres and Harris, 1997; Kyanite schist and sillimanite gneiss- Harris et al., 1992; granites- Miller et al., 2001) (b) LHS source rocks (Rashid, 2002, 2005).

MnO are plotted in the binary diagram (Fig. 5). The diagram shows the effect of hydraulic sorting whereby the above group of elements concentrate in finer fraction and are depleted in coarser fraction. The linear arrays of points along lines extending towards the origin in FeO–TiO_2, FeO–Al_2O_3 and Al_2O_3–TiO_2 binary diagrams demonstrate that the above elements were immobile, and also hydraulically fractionated in a similar manner. MnO indicates slight mobilization (Fig. 5d), particularly in the bed-load sediments.

The plots of elements against SiO_2 provide further insight into element mobility and hydrodynamic behaviour of their major mineral phases (Fralick and Kronberg, 1997). SiO_2-immobile element plots result in a linear arrangement of points extending either towards 0% (if major mineral phases containing the elements are concentrated in sand) or 100% (for elements concentrated in fine fraction). Fig. 2a, b, c, d, e, f describes typical trends for elements which are concentrated in the fine fraction. Trend of Al_2O_3, TiO_2, FeO and K_2O towards 100% SiO_2 indicates them to have remained chemically immobile in the present study. MgO and MnO are concentrated in fine fraction, but are also lost from the system particularly from the bed-load sediments although to different extent. Fig. 2g, h, i, j indicates Na_2O, CaO, P_2O_5 and Sr which are concentrated in the sand fraction. The distribution of points in it demonstrates that these elements are also lost from the system to

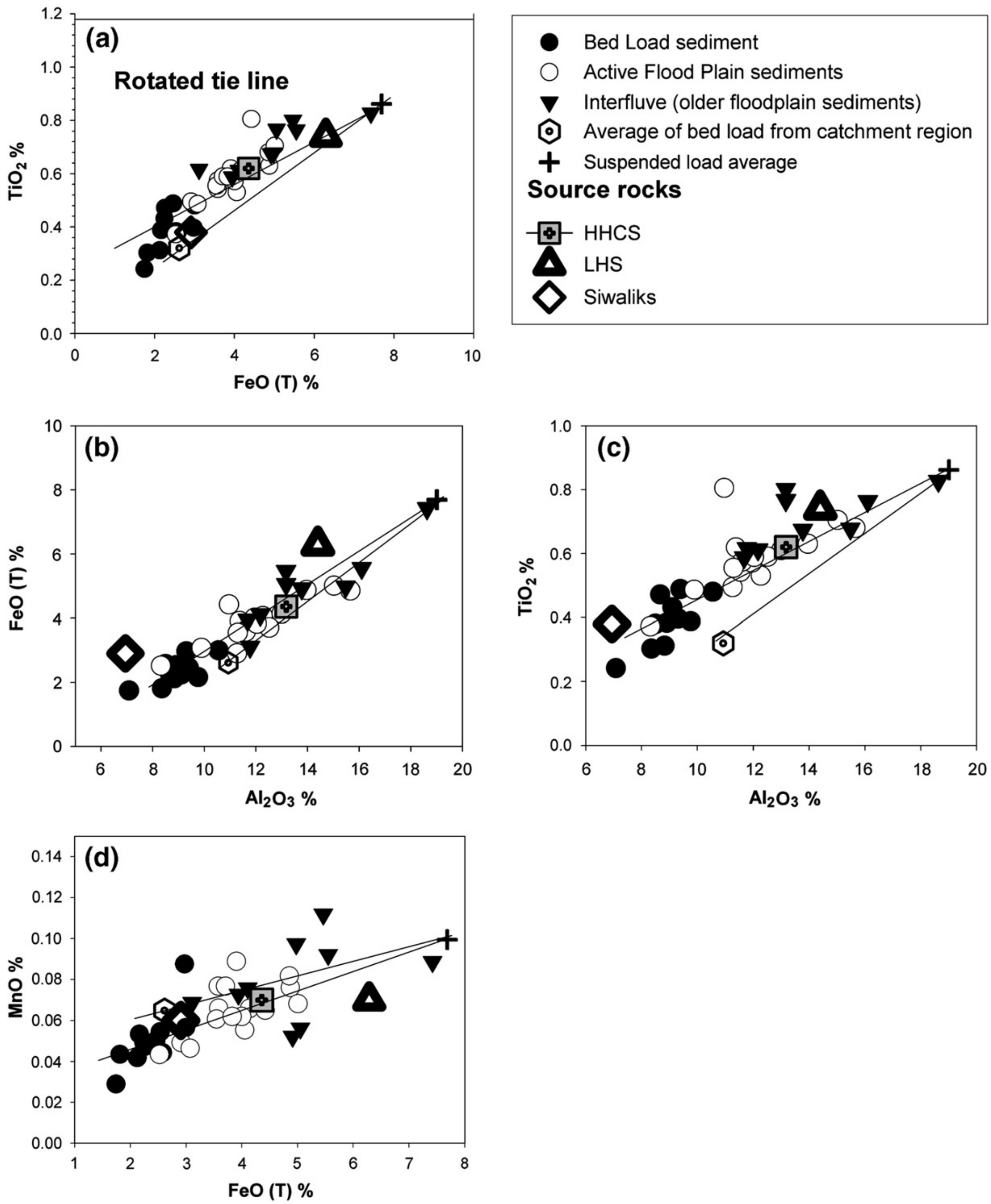

Fig. 5. Plots of sediments in the bivariant diagram between different elements. See the rotation of tie line joining the plots for bed load in catchment region and suspended load. For details see text. For comparison average of Higher Himalaya, Lesser Himalaya and Siwalik rocks (Galy and France-Lanord, 2001) are also plotted.

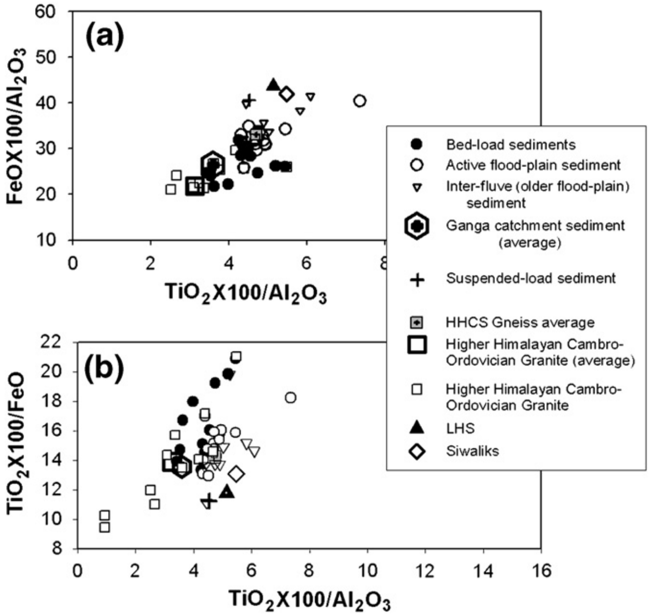

Fig. 6. (a). FeO/Al_2O_3 vs. TiO_2/Al_2O_3 ratio diagram comparing the Ganga plain sediments with various source rocks (HHCS, LHS and Siwaliks) (b) FeO/TiO_2 vs. Al_2O_3/TiO_2 ratio diagram comparing the Ganga plain sediments with various source rocks (HHCS, LHS and Siwaliks from Galy and France-Lanord (2001); Cambro-Ordovician granites from Miller et al. (2001).

variable extent particularly in Plains sediments. The concentrations of Na_2O and Sr in the catchments bed-load and few active flood-plain and inter-fluve sediments is higher than that expected by hydrodynamic sorting.

The binary plots discussed in the preceding section suggest that Al_2O_3, TiO_2, FeO and K_2O were immobile and affected by sorting in similar manner. These elements are concentrated in finer fraction as suggested by their plots against SiO_2. Since the above group of elements are immobile and show hydrodynamic similarity of the mineral phases holding them, their ratio should be same as the average ratio of these elements in their source.

The Ganga River before entering into Plains traverse through rocks of three major tectonic units of the Himalaya from north to south such as the Higher Himalayan Crystalline Series (HHCS), Lesser Himalayan Series (LHS) and the Siwaliks exposed at different elevations decreasing from north to south. The dominant lithologies of HHCS include Precambrian metasedimentary rocks comprising high grade kyanite/sillimanite gneiss and biotite–muscovite gneiss. These were intruded by numerous Cambrian to Ordovician granites and to the north by Miocene–Pliocene leucogranites. The LHS comprise largely low-grade metasediments composed of schist and phyllites of Precambrian–Palaeozoic age. The Siwaliks consist of unmetamorphosed sediments. In the present study we have compared the sediment chemistry with the above mentioned lithologies to establish the dominant source of the sediments.

The plot of suite of all sediments in the present study in FeO/Al_2O_3–TiO_2/Al_2O_3 diagram (Fig. 6a) show clustering of data points around average composition of HHCS rocks with the exception of bed-load sediments and two samples of inter-fluve sediment. The scatter in bed-load indicates less thorough mixing of source region detritus in it and higher contribution from Cambro-Ordovician granite. Higher component of Cambro–Ordovician granite from HHCS is expected in bed-load because of their inherent coarse texture compared to the gneiss and schist. This indicates gneiss and Cambro-Ordovician granites of HHCS formed the sediment source. The separation of two inter-fluve and one active flood-plain sediment (from location 14 and 15, Fig. 1) from the cluster may probably be the result of higher proportion of biotite in them, caused by reworking or aeolian input, as these are older sediments. The average compositions of LHS and Siwalik rocks plot outside the sediment cluster although they are not very distinctly separated. This is expected, as in-spite of higher and lower concentration of Al_2O_3, FeO and TiO_2, in LHS and Siwaliks respectively, compared to HHCS, they plot close to the trend line of sediments (Fig. 5a, b, d).

In the TiO_2/FeO–TiO_2/Al_2O_3 diagram (Fig. 6b) the average HHCS rocks plot in the centre of field defined by the sediments. Again for the same reason as stated in the preceding paragraph, the LHS and Siwalik rocks although separated, do not plot very far from the sediment cluster.

Only in the plot involving Na_2O/K_2O and SiO_2/Al_2O_3 ratios (Fig. 7a) we find the LHS and Siwalik rocks well separated from the sediment composition. I use this plot to argue that, due to feldspar sorting, even in physically weathered sediments, the fine grained fractions are always expected to have lower concentration of Na_2O. Therefore, they are expected to have lower Na_2O/immobile element ratio compared to the source. In no case can this ratio be higher in fine grained sediments compared to their source. In contrast we find lower Na_2O/K_2O ratio in LHS and Siwalik rocks average compositions in comparison to the entire sediment suite from the Ganga Plain (Fig. 7a). This would suggest that LHS and Siwalik rocks could not be the source to the sediments. On the contrary, the HHCS rocks is found to have similar or higher Na_2O/K_2O ratio in comparison to the sediments (Fig. 7a) and therefore could have formed the source to the Ganga Plain sediments. Thus using the above argument and the earlier discussed diagrams we can conclude that HHCS rocks were probably the dominant source to the studied sediments.

Th/Sc, La/Sc, La/Th ratios are especially sensitive to the nature of the source. They are useful in distinguishing mafic from felsic sources,

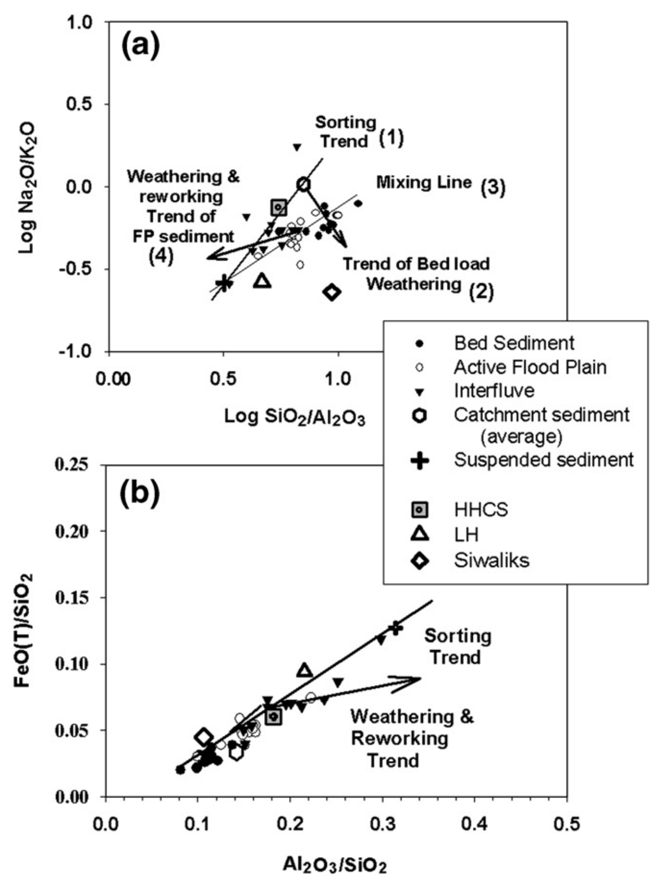

Fig. 7. Plots of sediments in (a) Log Na_2O/K_2O vs. Log SiO_2/Al_2O_3 and (b) $FeO(T)/SiO_2$ vs. Al_2O_3/SiO_2 diagram showing the pathways of chemical evolution of sediments (numbered 1–4 in Fig. 6a) during its passage from catchment to the plains. For explanation refer text. Average source and suspended sediment composition after Galy and France-Lanord (2001).

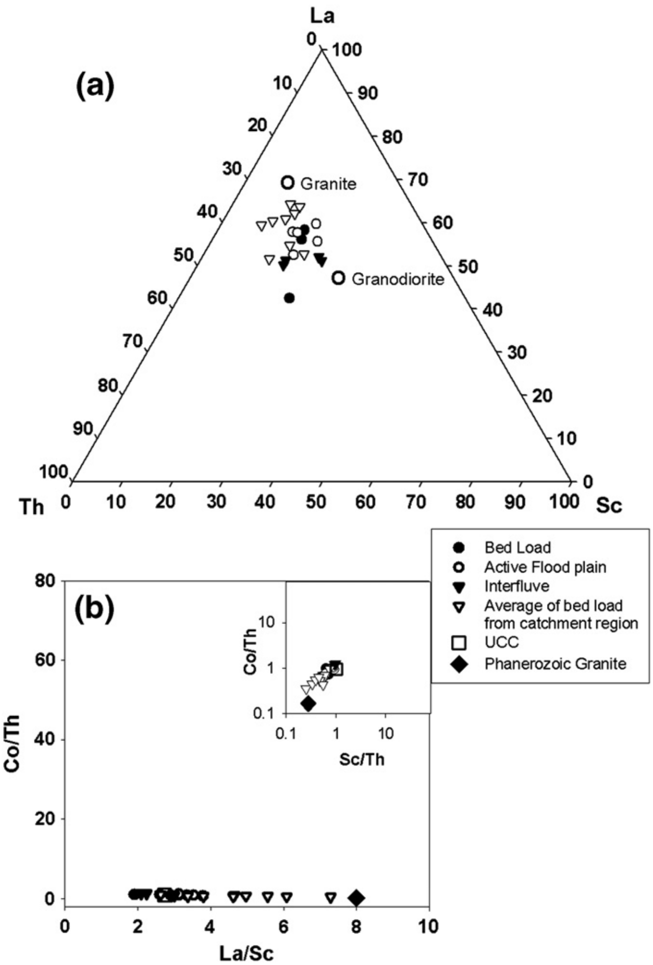

Fig. 8. Plots of selected sediments samples from all the groups in (a) La–Th–Sc and (b) Co/Sc vs. Sc/Th and Sc/Th vs. La/Sc diagrams. Note that in all the diagrams the sediments plot in-between the field of Granite and Granodiorite.

Sc and Co being preferentially concentrated in the former because of their compatibility with early fractionating phases. Whereas, Th and La are preferentially partitioned into melts, being incompatible elements, and are associated more with the later fractionating minerals and hence are more concentrated in felsic rocks (Taylor and McLennan, 1985). Thus for fractionated crust Th/Sc, Th/Co, La/Sc ratios are high and for mafic rocks they are low. Typically, for post Archean UCC the ratio of Th/Sc is ~1, for granitic rocks it is higher and for Archean and mafic rocks it is less than 1.

The entire sample group of Ganga Plain sediments shows higher Th/Sc (1–1.9) and similar La/Sc (2–3.5) ratios (Supplementary Table 2) compared to the UCC suggesting a granitic to granodioritic source. In the triangular plot for La, Th, Sc (Fig. 8a) all the sediments in the present study are plotted in the field corresponding to silicic rocks between granite and granodiorite composition. Similarly in the plot of elemental ratios, such as Co/Th versus La/Sc, and Co/Th versus Sc/Th (Fig. 8b) the samples plot between UCC and granite. The above results imply that the sediments have been derived dominantly from granitic to granodioritic rocks corroborating our observation as discussed earlier in this section.

The rare earth elements (REE) are also considered to preserve the source rock composition as they are carried almost entirely by the detrital component (Taylor and McLennan, 1985; Wronkiewicz and Condie, 1987). They are useful in differentiating between the felsic and mafic source. Mafic rocks have less fractionated chondrite normalised REE pattern with low $(La/Yb)_N$ ratios and little or no Europium anomalies, in contrast the felsic rocks generally show fractionated chondrite normalised REE patterns with higher $(La/Yb)_N$ ratios and prominent Eu anomalies (Taylor and McLennan, 1985). All the sediments in this study show fractionated chondrite normalised REE patterns (La_N/Yb_N average 8.80, 8.97, and 9.14 for bed-load, active flood-plain and inter-fluve sediments respectively) (Table 2) with prominent Europium anomalies ranging between 0.32–0.59 (Fig. 3). This would suggest felsic source rock to the sediments. Deeper Eu anomalies compared to UCC once again suggest more silicic source probably between Granite and granodiorite. One thing to note here is the similarity in the REE patterns of the Ganga River sediments from the Himalayan region with various group of sediments namely bed-load, active flood-plain and the inter-fluve from the Ganga River in Plains region (Fig. 3). This suggests that the REE patterns in the present case are neither affected by fluvial process of sorting nor weathering, and the homogenisation of at least the phases holding REE, has occurred in the hilly tract itself even before entering into the Plain region and the uniform REE pattern is not a result of multiple recycling and mixing in the Plain region.

The PAAS normalised REE patterns of the sediments are compared with the average patterns of different lithologies from HHCS and LHS (Fig. 4a,b). The entire group of Ganga Plain sediments have LREE patterns similar to various lithologies of HHCS and overlapping abundances, but different HREE patterns and content (Fig. 4a). The Cambro-Ordovician granites show a higher degree of HREE fractionation with lower contents in contrast to the gneiss/schist which show higher content and HREE enrichment. The sediments show intermediate fractionation and content of HREE compared to the above HHCS end member source rocks. Thus the HREE of sediments require admixture of above end member source rocks to attain their patterns.

The LHS lithologies show enrichment of both LREE and HREE with higher content compared to the sediments, and have negligible Eu anomalies (Fig. 4b). Any significant contribution of the LHS rocks to the sediment would result in reduced Eu anomalies and LREE enrichment of the sediments compared to the HHCS source rocks, which is not found to be true. This and the results of the ratio plots discussed earlier indicates greater similarity of sediments to granite and gneiss of HHCS rocks suggesting them to have acted as their dominant source. This is expected as the HHCS form the region of

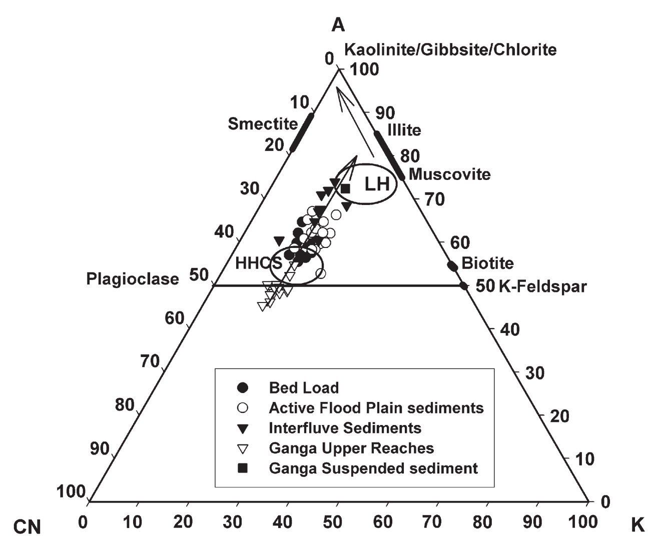

Fig. 9. A–CN–K plots (Nesbitt and Young, 1984) for sediment samples from different depositional regimes of Ganga river in plains along with the plots for sediments from the catchment tributaries of Ganga river (Singh, communicated), Suspended load (Galy and France-Lanord, 2001), and field for Higher Himalaya and Lesser Himalaya source rocks (Ahmad et al., 2000; Ayres and Harris, 1997; Galy and France-Lanord, 2001; Harris et al., 1992; Miller et al., 2001; Rashid, 2005; Richards et al., 2005).

higher elevation and relief ensuing greater erosion compared to the LHS which have much lower elevation and relief.

5.2. Geochemical changes related to weathering and sorting

The chemistry of the sediments is a result of the net influence of provenance, weathering and the fluvial processes involved in sorting (Rollinson, 1993, McLennan et al. 1993). The bulk change in the chemistry can be related to weathering by an index called the chemical index of alteration (CIA), which is used to quantify the weathering (Nesbitt and Young, 1984). $CIA = (Al_2O_3/[Al_2O_3 + CaO^* + Na_2O + K_2O]) \times 100$, where Al_2O_3, CaO^*, Na_2O and the K_2O are molecular concentrations (CaO^* is CaO in the silicate fraction). The CIA of all the sediments range between 55 and 74, which indicates low to moderate weathering. It is interesting to note that the order of increasing intensity of weathering is from bed-load (range: 55–64, average CIA = 59) to active flood-plain (range: 58–67, average CIA = 61.4) to inter-fluve sediments (range: 60.4–73.9, average CIA = 67.26). In another study by the author it has been shown that the bed sediments in the upper reaches of the Ganga River are immature product of the physically ground up source rocks and have suffered only minor chemical weathering and have even the bulk chemistry similar to the source rocks (see Fig. 9 and Table 4). To look into the changes brought about after these sediments enter into the Gangetic Plain, I have plotted the sediment from the upper reaches along with the sediments collected in the Plains region from the channel bed, active flood-plains and the inter-fluve region (older flood-plains) in the Al_2O_3–$CaO + Na_2O$–K_2O (A–CN–K) triangular plot (Nesbitt and Young, 1984; Nesbitt et al., 1996) which gives the weathering trend. The A–CN–K diagram is the plot of the molecular proportion of Al_2O_3, CaO, Na_2O, and K_2O. In the A–CN–K diagram (Fig. 9) all the above groups of sediments plot along the same weathering trend line suggesting their derivation from a common source. The sediments from the upper reaches of the Ganga River plot near to the feldspar line and on the lower side of the field of source rocks (HHCS). This indicates that these sediments are chemically unweathered and the slight shift towards the lower end of the source rock field is the result of little sorting whereby some amount of alumino-phyllosilicates have been removed, resulting in a slight reduction in both Al_2O_3 and K_2O (major components of muscovite) and consequent increase in Na_2O. On the other hand, the sediments from the bed-load in the Ganga Plains region plot at the higher side of the source field indicating that after entering into the Plains the sediments have suffered some amount of chemical weathering resulting in the breakdown of a minor amount of feldspars and hence loss of CaO and Na_2O. The active flood-plain sediments plot further up the weathering trend line than the bed-load sediments. This does not necessarily indicate a higher degree of chemical weathering of these sediments as the mineral sorting by fluvial processes can also produce similar results (Nesbitt and Young, 1989; Taylor and McLennan, 1985) whereby more phylo-silicates are incorporated into the flood-plain sediments resulting in an increase in Al_2O_3. It is interesting to note that the older flood-plain sediments from the inter-fluve region plot further up the trend line than the active flood-plain sediments (being deposited by present day floods) although both have been deposited by similar processes, though at different times. This would suggest that the inter-fluve sediments have suffered chemical weathering after their deposition in the Plain region. The foregoing discussion suggests that Ganga River sediments have suffered chemical weathering only after entering into the Plains and not in the catchment region mainly because of change in climate from cold and frigid in the catchments region to humid sub-tropical in the Plains. The above observation indicates that the use of sediment chemistry particularly the CIA in ancient sediments to infer the climate of their source region may not always be true due to en-route modification suffered by the sediment.

5.3. Evolution of sediment geochemistry from the source region to the Plain

To further define and differentiate the chemical changes brought about by a combination of sorting and weathering from that solely due to chemical weathering we have used binary plots between Log Na_2O/K_2O vs. Log SiO_2/Al_2O_3 (Fig. 7a) and $FeO(T)/SiO_2$ vs. Al_2O_3/SiO_2 (Fig. 7b). For comparison, we have also plotted the composition of the suspended-load (Galy and France-Lanord, 2001), the average composition of bed-load in the Himalayan region and the average of different source rocks (Galy and France-Lanord, 2001).

5.3.1. Influence of feldspar sorting and weathering on the sediment chemistry

The plot of Log Na_2O/K_2O vs. Log SiO_2/Al_2O_3 very clearly brings out the sequence of processes involved in the evolution of chemistry of the Ganga sediments. The sequence is indicated by numbers given in Fig. 7a. The first step, as indicated by number (1) in the figure is the linear trend line from bed-load in the source region to suspended-load, passing through the average source rock composition (HHCS) in-between. The linear array within the above group of sediments can be explained by physical separation occurring due to the process of hydraulic sorting of physically weathered sediments, whereby alumino phyllosilicate minerals (mostly mica, chlorite and illite) are preferably taken into suspension along with minor amounts of feldspars and quartz, as indicated by their higher percentage in the suspended load as reported by Chakrapani (2005). This results in an increase of K_2O and Al_2O_3 in the suspended load as seen in Fig. 7a. At the same time, because of selective removal of phyllosilicate minerals in suspension, there is a residual enrichment of feldspar and quartz in the bed-load compared to the source rocks and the suspended-load, which leads to an enhanced concentration of Na_2O and SiO_2 in the bed-load, as is clearly implicit in the above figure. Only a small shift of the plot of bed-load sediment in the Himalayan region from the average source rock composition indicates that, although sorting has taken place, it has not been very efficient. This is to be expected as the sediments are physically weathered and mineral separation has not been very efficient due to the lack of chemical weathering.

Once these sediments enter into the Plain, chemical weathering picks up as indicated by trend line (2) from the bed-load fraction in the catchment region to the bed-load fraction in the Plain, which indicates loss of both Na_2O and Al_2O_3 (Fig. 7a). This is mainly due to a higher residence time of sediments in the Plains because of a change in topography and gradient of the river and also a change in the climatic zone from cold in the catchment region to humid sub-tropical in the Plains, which favours chemical weathering. The decrease in Na_2O concentration is due to the chemical weathering of feldspar in the bed-load sediments which results in the decrease of the Na_2O/K_2O ratio. This leads to a simultaneous increase in the SiO_2/Al_2O_3 ratio in the bed-load sediments as the fine textured clay produced by the above weathering is incorporated into the suspended load resulting in a decrease in the Al_2O_3 content in the residual bed-load sediments. This process leads to a shift in the plot position of the bed-load sediments from the Plains away from the catchment bed-load of trend-line (1). The next step as specified by the trend-line (3) in Fig. 7a indicates the effect of the process of physical remixing of the Plains bed-load and the suspended-load, resulting in the deposition of unsorted sediments with a higher SiO_2/Al_2O_3 ratio, on the present day active flood-plain. At the time of flooding, due to higher energy conditions, part of the bed-load sediment is picked up in suspension and along with the fine-grained suspended sediment is deposited on the flood-plain. Therefore the flood-plain sediments attain a chemistry intermediate between the bed-load and suspended-load sediments and plot more closely to the source rock composition (HHCS) as seen in the figure. A slight downward shift of the plot for the active flood-plain sediments from the source composition is because of the mobilisation and loss of Na_2O to the solution due to feldspar weathering in the Plains, as observed in step 2. The flood-plain sediments, once deposited in the Gangetic Plain, have a long residence time, as indicated by the very old ages obtained for the inter-fluve sediments (Srivastava et al., 2000, 2003). Due to the long residence time and the Gangetic Plain having a humid sub-tropical climate for 5 to 6 months and hot–dry climate for 4 months in a year, these inter-fluve sediments are further modified by combined effect of different processes as indicated by step (4) in the diagram. Step 4 indicates decrease of the SiO_2/Al_2O_3 ratio and limited change in Na_2O/K_2O ratio. The plausible explanation for this may be the in-situ weathering of feldspars under alkaline conditions, leading to the loss of silica and alkali elements keeping the Na_2O/K_2O ratio buffered. Because the inter-fluve is older flood-plain, it may have also received significant aeolian component with cations, which on weathering as stated above may have lead to further residual enrichment of Al_2O_3. Inter-fluve and active flood-plain have similar mean size but inter-fluve plots further in the trend-line in A–CN–K diagram (Fig. 9), which supports higher weathering of inter-fluve sediments. Thus, the foregoing discussion clearly indicates that fluvial processes of mineral sorting and feldspar weathering in the Gangetic Plain has played a major role in the modification of the sediment chemistry of the Ganga River.

5.3.2. Influence of sorting of micas on the sediment chemistry

The FeO(T)/SiO_2 versus Al_2O_3/SiO_2 plot (Fig. 7b) brings out clearly the changes brought about by the sorting of micas on the sediment chemistry in the present study, along with the influence of feldspar weathering, on the trend of the plots of various groups of sediments. In this diagram the plots for source rock (HHCS), suspended-load and the bed-load from the catchment region all fall on the same trend line as expected by the hydraulic separation of physically weathered sediment. In the present case it leads to the enrichment of micas in the suspended-load in preference to other non-platy minerals resulting in an increase of FeO and Al_2O_3 in this sediment compared to the source. In contrast the bed-load has a decrease in FeO and Al_2O_3 due to a loss of mica and the higher retention of quartz and feldspar compared to the suspended-load. The shift in the plot of the Plains bed-load sediments away from that of the catchments bed-load towards the lower FeO(T)/SiO_2 and Al_2O_3/SiO_2 ratios indicates further loss of Al_2O_3 and FeO from the bed-load after entering into the Plains. This is due to the effect of sorting whereby phyllosilicates are lost to the suspended-load. During flooding due to mixing of the bed-load and suspended-load sediments the active flood-plain sediments again acquire a chemistry intermediate between these two end members, and closer to the source rocks (Fig. 7b). After deposition on the flood-plain in the Ganga Plain region, further change in sediment chemistry occur as indicated by the shift of inter-fluve (older flood-plains) sediments away from the active flood-plain sediments. Initial shift of plots of inter-fluve sediments along the trend-line is mainly brought about by feldspar weathering under alkaline conditions, which mobilises the SiO_2 and alkali elements, and results in residual enrichment of FeO and Al_2O_3 in same proportion. Further shift of inter-fluve sediments away from the trend-line towards either higher FeO(T)/SiO_2 or Al_2O_3/SiO_2 ratios can only result from disproportionate addition or removal of biotite, muscovite and feldspar. The possible reason for this may be disproportionate removal of biotite, muscovite and/or feldspar by hydraulic reworking by rain water, or addition/removal by the aeolian activity. This is expected because inter-fluve is an older flood-plain exposed to around four months of rainfall during monsoon, and aeolian activity for three months during hot and dry summer season in the Plains.

6. Implications to Himalayan weathering

The foregoing discussion suggests that the chemistry of sediments of the Ganga River in different parts of its traverse and depositional sub-environments has been influenced by different sets of processes. In the catchment region physical weathering has been the dominant process. On entering into the Plains region the sediments have been influenced both by sorting and chemical weathering. In the Plains region the chemistry of the bed-load sediments shows similar influence from sorting and chemical weathering. In the Plains region the chemistry of the active flood-plain sediments shows a greater influence from remixing of bed-load and suspended-load compared to little from chemical weathering. Whereas, the older sediments from the inter-fluve region have suffered more chemical weathering compared to recently deposited active flood-plain sediments, and also show some influence of hydraulic and aeolian activity. The above noticeable difference between the different groups of sediments deposited by the Ganga River is because of the variation in topography and climate along its long traverse from the Himalaya to the sea, and the higher residence time of sediments in the Plains region. Galy and France-Lanord (2001) in their study based upon the mass balance calculations have indicated that not all the sediments eroded from the Himalayan region by Ganga River reaches the Bay of Bengal and at least 50% of the sediments get locked into the Plain region. The cold arctic to sub-arctic climate, higher relief and topographic gradient, along with known active tectonics in the source region favour physical weathering and higher erosion rates. In contrast the flat undulating topography of the Plain region along with humid sub-tropical climate (at least for 5 to 6 months in a year) favours chemical weathering of the sediments locked up in the vast Gangetic Plain. Thus the above observations imply that the influx of elements into solution in the waters of the Ganga River and ultimately to the Bay of Bengal as a result of silicate weathering, dominantly occur in the Gangetic plain region compared to the source in the Himalaya where physical weathering and erosion are the dominant features. Although silicate weathering has occurred in the Plains, it is not to a significant level. This would imply that the erosion by Himalayan Rivers, particularly the Ganga, provided only unweathered sediments and therefore its efficacy in terms of CO_2 drawdown to cause any global cooling needs to be better understood.

7. Conclusions

The results and discussion of the present study leads to the following conclusions:

1 The silicate chemical weathering of Ganga River sediments at the source was insignificant and has occurred only after entering into the Plains due to change in the climatic system from the cold-frigid in the catchment region to humid sub-tropical in the Plain region and also because of higher residence time of sediments in the Plains.

2 The above observations imply that influx of elements into the water of the Ganga River and ultimately to the Bay of Bengal by the silicate weathering, dominantly occur in the Gangetic Plain region compared to the source in the Himalayas. The present study attracts caution on the use of sediment geochemistry to infer the climate of the catchment region especially for the sediments deposited by the large river systems.

3 The results from this study show that the fluvial process of mineral sorting and post depositional feldspar weathering in the Plains can play a major role in the modification of the sediment chemistry in the Plain region and result in rotation of trend line joining the suspended-load and the bed-load concentration of various elements and their ratios around the suspended-load as an axis. This is mainly due to the loss of Na_2O and other mobile elements.

4 In the present suite of sediments, Al_2O_3, FeO and TiO_2 have behaved as immobile elements. Their ratio plots along with REEs suggest that Gneiss and Cambro-Ordovician granites of HHCS have been the dominant source to the sediments.

5 The better clustering of points of flood-plain sediments in the ratio plots of immobile elements suggests them to be the most appropriate population for tracing the provenance.
6 Uniformity of the REE pattern of all the studied sediment samples from the Plain region and the catchment region indicates complete homogenisation of at least the REE hosting phases in the catchment region itself before entering into the Plains.
7 The supply of unweathered materials by Ganga River to the Plains and their subsequent lower chemical weathering in the Plains calls for a re-look into the efficacy of Himalayan uplift in terms of CO_2 drawdown to cause global climate cooling.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.chemgeo.2009.06.013.

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