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Central Himalayan crystallines as the primary source for the sandstone–mudstone suites of the Siwalik Group: New geochemical evidence

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ABSTRACT

Geochemistry of the Sub-Himalayan foreland basin Siwalik sediments has been used for interpreting the nature of the source rocks. This study has shown that the compositional changes are a function of stratigraphic height, demonstrated by the upward increase of P_2O_5, Na_2O, CaO, MgO and SiO_2 content from Lower to the Upper Siwalik rocks. On the other hand, K_2O, Fe_2O_3, TiO_2 and Al_2O_3 show decrease with the increasing stratigraphic height. These trends are a clear reflection of time-controlled changes in the source lithology. Ratios such as Eu/Eu*, $(La/Lu)_{cn}$, La/Sc, Th/Sc, La/Co, and Cr/Th suggest a prominent felsic source area for the Siwalik sediments. Chondrite-normalized REE pattern with LREE enrichment and moderately flat HREE pattern with sharp negative Eu anomaly are attributed to a felsic source. Contrary to the existing belief, this study has ruled out any contribution from the mafic sources and highlighted the compositional similarities of Siwalik sediments with the crustal proxies like PAAS, NASC and UCC. The geochemical data point to a significant role played by the Precambrian and early Paleozoic granitic rocks of the Himalayan tectogene in shaping the composition of the foreland sediments. The variable CIA values and marked depletion in Na, Mg and Ca exhibited by the Lower, Middle and Upper Siwalik sediments reflect variable climatic zones and variations in the rate of tectonic uplift of the source area. Our results demonstrate that in the Lower Siwalik and part of the Middle Siwalik, Higher Himalayan Crystalline sequence (HHCS) was the primary source area with minor contributions by the meta-sedimentary succession of the Lesser Himalaya. Later, during the deposition of the upper part of the Middle Siwalik and Upper Siwalik, the source terrain switched positions. These two prominent source terrains supplied sediments in steadily changing proportion through time.

1. Introduction

The Siwalik Group, probably the largest quartzolithic petrosome in the world with a collisional-orogen provenance, consists of a Neogene sedimentary succession of 5–8 km thickness, deposited as the Himalayan foreland sequence and developed following collision (~54–55 Ma) between the Indian and Eurasian continents (Molnar and Tapponnier, 1975; Tapponnier et al., 1986; Dewey et al., 1989; Harrison et al., 1992; Critelli and Ingersoll, 1994). This foreland basin package (~18.3–4.9 Ma) of Middle Miocene to Late Pleistocene age is primarily fluvial in origin and is represented by a shale, sandstone–mudstone sequence in the lower part and a thick sandstone and conglomerate succession in the upper part.

In recent years, a wide range of geological studies in different sectors of the Himalayan Range has enriched our geotechnical database. Works of Gouzu et al. (2006) on Tso Morari eclogite, geochronological studies by Yoshida and Upreti (2006) and seismic and tectonic studies by Purnachandra Rao et al. (2006) provided new insights to the world of Himalayan geology. The mantle dynamics of the Laddakh Himalaya in relation to the secular evolution of fluids in Earth history was evaluated by Santosh and Omori (2008). Geochemical studies across the Precambrian–Cambrian boundary rocks in the Gondwana supercontinent and evolution of the Himalayan motif were discussed by Banerjee and Mazumdar (1999). For the Nepal Himalayan sector, Paudel and Arita (2006) proposed a new thermal evolutionary model while Goscombe et al. (2006) gave a detailed account of the crustal architecture. Sequence stratigraphic approach (Catuneanu and Eriksson, 2007; Eriksson et al., 2007) to solve sedimentological and stratigraphic riddles in the Himalaya is rather rare (Jiang et al., 2002, 2003) Stratigraphy and tectonics of the Siwalik Group have been studied by Burbank et al. (1986, 1988), Johnson et al. (1982,1986), Burbank and Raynolds (1988), Baker et al. (1988), Burbank and Beck (1989, 1991); magnetostratigraphy by Johnson N.M. et al. (1982, 1985, 1988), Appel et al. (1991), Cande and Kent (1995); sedimentology by Sinha and Sastri (1973), Tandon (1976), Parkash et al. (1980), Behrensmeyer and Tauxe (1982), Raiverman et al. (1983), Mulder and Burbank (1993), DeCelles et al. (1998), Critelli and Ingersoll (1994), Huyghe et al. (2001), Shikha et al. (2001) and Kumar et al. (2003). Cerveny et al. (1988), Harrison et al. (1993), and chemical

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analyses of detriral particles by Najman et al. (2001) and Najman (2006) have enriched the database on Himalayas. The geochemical signature of bulk sedimentary rocks is a complex function of several geological variables such as source terrain composition, weathering environment, transport induced sorting and diagenesis (McLennan, 1989). So far, a detailed chemistry of the Siwalik sediments has not been carried out. Most of the provenance related studies have utilized petrography and heavy minerals as the proxies. In this paper we have identified the characters of the provenance, interpreted the nature of weathering in the source region and inferred the tectonic setting of the basin with the help of newly generated chemical analyses along with available petrologic data. The chemical contribution of the Higher Himalayan metamorphics to the foreland basin sediments has also been evaluated using data from Islam et al. (1999).

Like most fluvial deposits, the Neogene Siwalik sediments of the Himalayan foreland basin display dominance of sandstone alternating with fine grained mudrocks. The ratio of these two lithological associations varies with change in the depositional sites. A majority of recent workers have studied the sand dominant components of the lithosome (Shikha et al., 2001 and references therein) with passing references to the fine grained portion of the litho-association. The present study focuses on the chemistry of the muddy component of the sandstone assemblage, which tends to better preserve the geochemical signatures of the primary association.

2. Geological setting

The Lesser Himalaya composed of low-grade metasediments of Mesoproterozoic to Lower Cambrian age is separated from the Siwalik succession by the Main Boundary Fault (MBT; Fig. 1). A few fossiliferous Cretaceous and Tertiary rocks are exposed in stratigraphic windows. Proterozoic Higher Himalayan metasediments and igneous rocks include several young leucogranites of uppermost

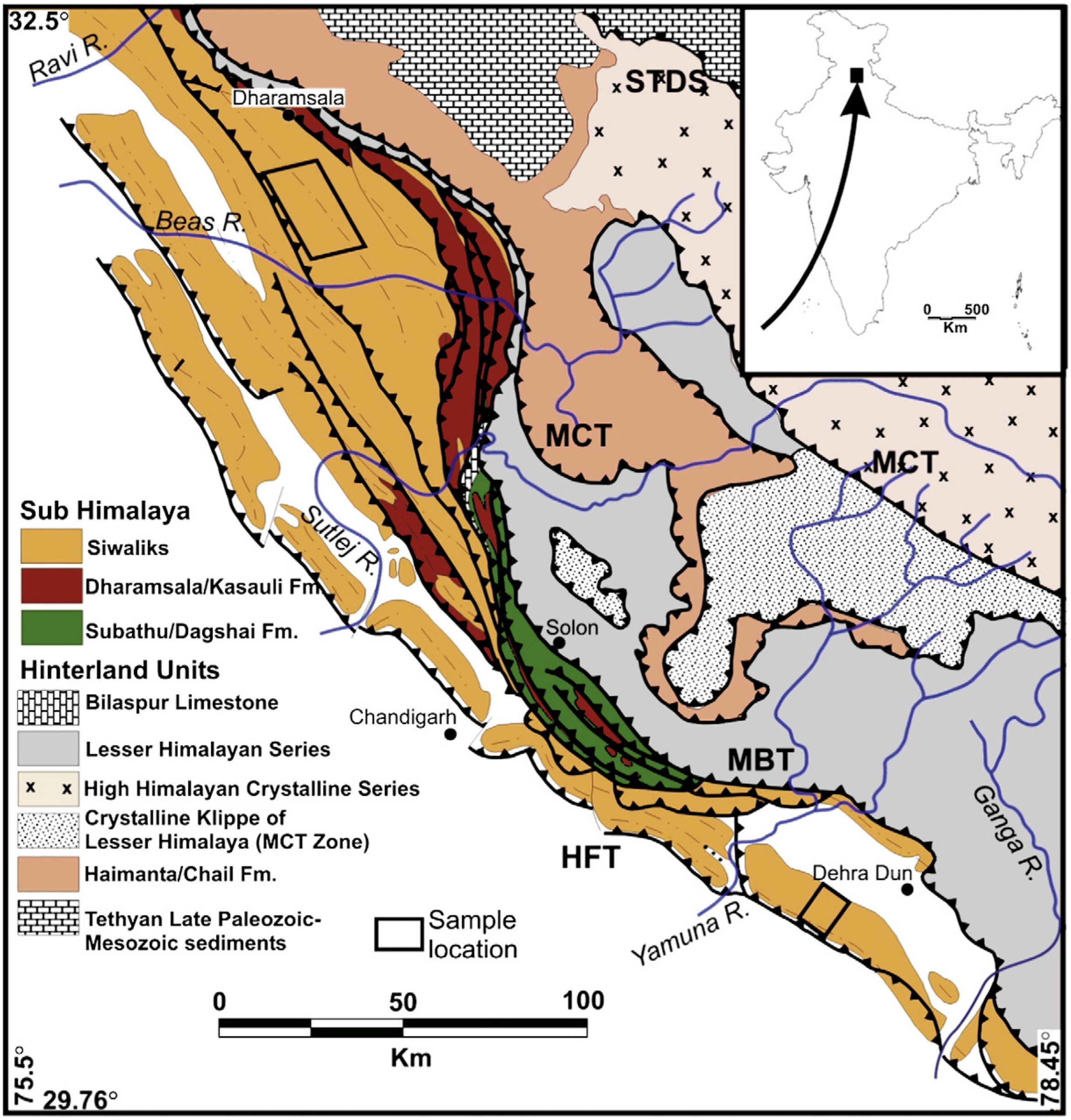

Fig. 1. Geological map of the northwestern part of the Himalaya. Box location in Kangra sub-basin and Dehradun sub-basin shows the location of the Siwalik samples. Major Himalayan faults are marked as; STDS—South Tibetan Detachment System; MCT—Main Central Thrust; MBT—Main Boundary Thrust; MFT—Main Frontal Thrust (modified map adopted from Powers et al., 1998).

crustal composition (Treolar and Searle, 1993). Un-metamorphosed sedimentary deposits of Cambrian to Paleocene age represent the Tethyan Zone in the north. The Trans-Himalayan Zone exposes the Upper Cretaceous to Eocene calc-alkaline plutons (LeFort, 1996). The Indus Suture Zone marks the area where the Indian and Eurasian plates are believed to have collided.

The high-grade crystalline rocks and granitoids of the Higher Himalaya along the Main Central Thrust (MCT) override the low-grade Proterozoic rocks of the Lesser Himalaya (Fig. 1). This thrusting event was concomitant to the convergence of India with Eurasia which was accompanied by propagation of a series of south-directed thrusts in Middle to Late Miocene times (Najman and Garzanti, 2000). One of the thrusts, the Main Boundary Thrust (MBT), which parallels the Krol Thrust is of present concern. As a result of such thrusting the Lesser Himalayan rocks slipped past many sub-Himalayan formations. The Siwalik sedimentary prism is confined between the Main Boundary Thrust (MBT) in the north and the Himalayan Frontal Thrust (HFT) in the south. The Main Frontal Thrust (MFT) separates the sub-Himalayan succession from the Indo-Gangetic alluvial plains to the south.

3. Materials and methods

Fine-grained sediments are known to preserve chemical signatures of the source terrain rather accurately. Such sediments are better mixed and chemically homogenous in comparison to the coarse grained fraction (Wronkiewicz and Condie, 1987; Cullers et al., 1988). The fine grained sandstone samples ($n = 35$) from Lower, Middle and Upper stages of the Siwalik Group were therefore studied at the Kotla and Ranital Sections of the Kangra sub-basin (Lower Siwalik) and at the Mohand Rao section (Middle and Upper Siwalik) of the Dehra Dun re-entrant in NW Himalaya. These sections were systematically sampled along well defined traverses through the Siwalik succession exposed in this region. The Lower Siwalik formation consists of three facies associations; sand dominant; sandy–mud-dominant and predominantly silty-heterolithic. The sampling bias clearly reflected in favor of the sand–mud dominant association. The Middle Siwalik, fine-grained multistoried sand bodies from the Mohand Rao section were sampled for the sand–mud interface. In the conglomerate-dominated Upper Siwalik, fine-grained sandstone and conglomerate matrix were sampled for this investigation. Published chemical analyses of the Himalayan granitoids and the Higher Himalayan Crystalline rocks (HHC) were used for evaluating the role of these formations in producing the Siwalik sedimentary package.

Major and some trace elements were determined by a sequential X-ray fluorescence spectrometer (Philips PW 1400 XRF) using fused beads and pressed pellets. Simultaneous analysis of the Siwalik sediments and the international reference standards show the confidence limits to be better than 95%. Trace and rare earth elements were analyzed in duplicate by Perkin-Elmer SCIEX model ELAN® DRC II ICP-MS using 50-mg sample aliquot and processed in standard laboratory format. The operative parameters are given in Balaram and Gnaneshwer (2003). The precision and accuracy of analyses were better than ±5% for all trace elements.

4. Results

4.1. Petrography

The Siwalik sedimentary pile forms a coarsening upward succession with gradual increase of channel bodies and decrease in overbank facies attributed to progradation. Thirty four medium to fine grained unaltered sandstone samples representing Lower, Middle and Upper Siwalik formations were petrographically analyzed. Nearly 300 grain points in each thin section were counted following the Gazzi-Dickinson method (Ingersoll et al., 1984). The point counts were plotted on standard ternary diagrams (Fig. 2a and b). These sandstones are predominantly quartzolithic with ubiquitous monocrystalline to fine grained polycrystalline quartz. K-feldspar and plagioclase along with lithic fragments of gneisses and schists are common. Sedimentary rock particles include limestone, dolomite, chert, shale and crystalline quartzite. Volcanic fragments are rare. Tiny crystals of zircon, tourmaline, garnet, apatite and opaque minerals show an increase in abundance along traverses from Lower to Upper Siwalik rocks. The Middle and Upper Siwaliks are more quartzolithic compared to the Lower Siwaliks. Intergranular spaces are filled with siliceous detritus and occasional authigenic calcite. Mica and iron oxide minerals are minor constituents. The finer component of the Siwalik succession is represented by mudstone composed of illite, chlorite, smectite, sepiolite and mixed clays (Chaudhri & Gill, 1983; Raiverman and Suresh, 1997; Suresh et al., 2004). The chlorite, montmorillonite and mixed clay layers are more common in the upper part of the succession.

4.2. Major elements

The major element composition (Table S1: available as electronic supplementary item) exhibits significant elemental variation with 62 to 78% SiO_2 and 10 to 22% Al_2O_3. The K_2O/Na_2O ratio shows progressive

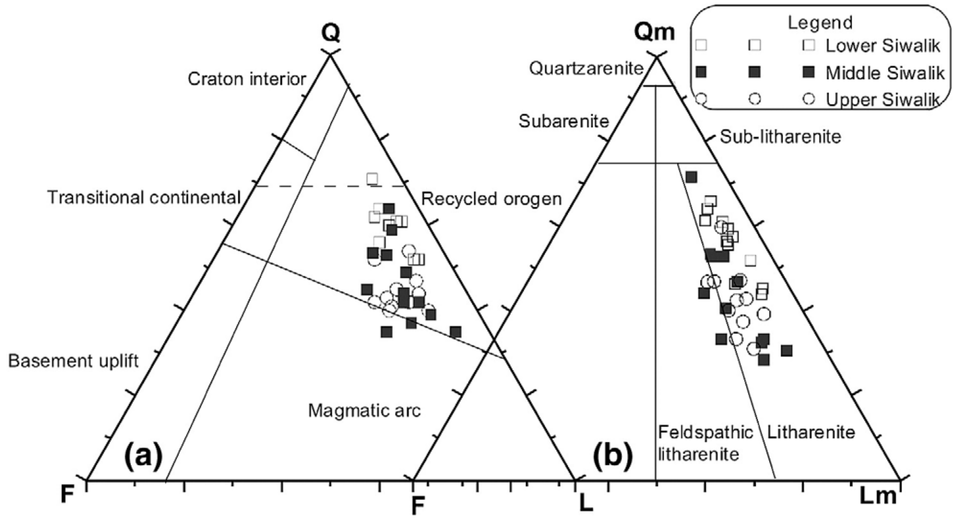

Fig. 2. (a) and (b) The QFL and Qm-F-Lm plots of the Siwalik sandstones (provenance fields after Dickinson et al., 1983). Q—Quartz, F—Feldspar, L—Lithic fragments, QM—Monocrystalline quartz, Lm—Lithic fragments + Quartzites, Polycrystalline quartz and chert.

decline from Lower to Middle Siwalik rocks while SiO_2 content increases along the same path. The average K_2O/Na_2O ratio of composite Siwalik sediments is ~3.78, marginally higher than the NASC value of 3.5. The SiO_2/Al_2O_3 ratio shows an increase from 3.29 in the Lower Siwalik to 4.77 in the Middle Siwalik and finally to 5.48 in the Upper Siwalik, suggesting progressive increase in the quartz content with younging of the succession. Small Na_2O contents (<1%) point to rarity of Na-rich plagioclases. On the other hand, the high K_2O/Na_2O ratio is attributed to the predominance of K-feldspar over plagioclase. A high concentration of Fe_2O_3 and TiO_2 in average Siwalik sediments is due to the abundance of a Ti-bearing heavy mineral suite. The TiO_2/Al_2O_3 ratio shows systematic increase in the younger sediments. Compositional changes in the Siwalik sediments as a function of stratigraphic younging are demonstrated by the upward increase of P_2O_5, Na_2O, CaO, MgO and SiO_2 concentrations. On the other hand, K_2O, Fe_2O_3, TiO_2 and Al_2O_3 show decreasing trends with increasing stratigraphic height of these rocks. These trends reflect time-controlled changes in the lithology of the source terrain. Increase in Fe_2O_3 and TiO_2 values could be attributed to the presence of heavy minerals like ilmenite.

All major elements in the sandstone–mudstone suite were plotted against Al_2O_3 values (Fig. 3). Crustal proxies like NASC (Gromet et al., 1984), PAAS and UCC (Taylor and McLennan, 1985; McLennan, 2001) were also plotted in order to assess the variations observed in the Siwalik sediments. Chemical composition appears to be dependent on the particle size, with Al_2O_3 (as constituent of clay minerals) showing an increase in the fine grained sediments. Other major elements show a conformable relationship with Al_2O_3. In other words, clay minerals dominate the fine fraction while quartz rules over the coarse fraction.

4.3. Alteration indices

The CIA (Chemical Index of Alteration) values exhibited by different levels of the Siwalik stratigraphy suggest a wide variety of climatic conditions in the source area. Such variability may have links with the tectonics affecting the region. Metamorphism does not seem to affect the CIA values (cf., Nesbitt and Young, 1982), hence the CIA index relative to 'Average Shale' provides significant information about the weathering scenario in the source area. The Lower Siwalik sediments with 82–87 CIA values when compared with 'Average Shale' (CIA: 70–75, Visser and Young, 1990) suggest an intensely weathered source terrain, while the Middle (CIA: 70–75) and Upper Siwalik (CIA: 71–82) sediments indicate moderation in the weathering conditions. High Al and low Ca and Na in the Lower Siwalik sediments reflect derivation from an intensely weathered terrain. Like CIA, the CIW (Chemical Index of Weathering: Harnois, 1988) based on an unaltered upper crustal value of 55–60, shows higher values for the Lower (96.6), Middle (86.1) and Upper Siwalik (87.6) sediments. These CIW values further strengthen the interpretation of a highly weathered source area. The PIA (Plagioclase Index of Alteration: Fedo et al., 1995) values for the Lower (60.8–78.2), Middle (67.4–78.2) and Upper (61.8–74.7) Siwalik exceed 60, which is the threshold value for moderate to low weathering environments. On the A–CN–K [Al_2O_3 ($CaO^* + Na_2O$) K_2O] plot of Nesbitt and Young (1984) the Siwalik sediments show depletion of Na and Ca compared to upper continental crust (UCC) values. A negative correlation between SiO_2 and CIA in the Siwalik sediments suggests grain size control on individual CIA values.

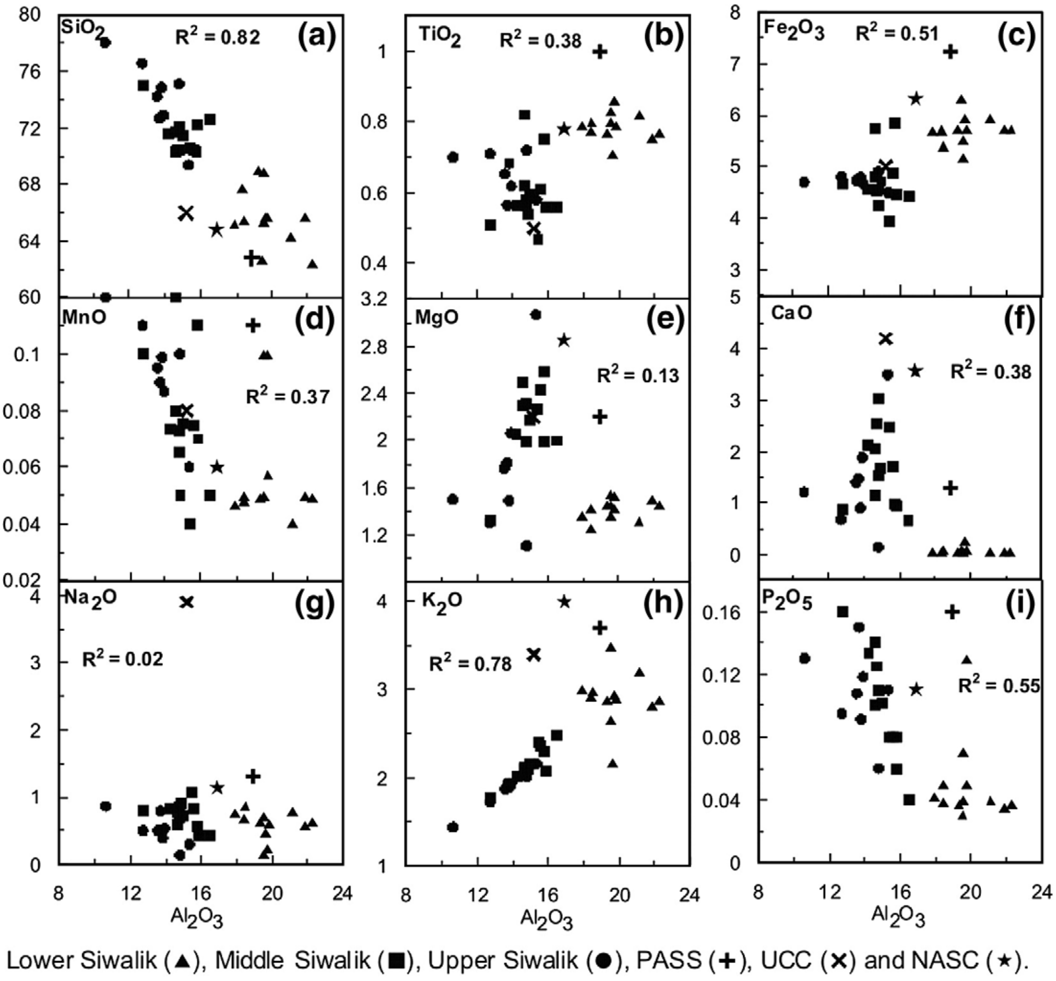

Fig. 3. The variation diagrams of the major elements plotted against Al_2O_3. Data for PAAS, NASC and UCC are plotted for comparison (Taylor and McLennan, 1985; Gromet et al., 1984).

4.4. Minor elements

The trace and rare earth element concentrations are given in Table S2 (available as electronic supplementary item). The concentration of *high field strength elements* (HFSE), Y, Nb, Zr, Hf and Ta shows variability within groups and also with reference to NASC and UCC values. Zr (12.6 ppm) and Hf (3.5 ppm) concentrations show a depleted character. Y concentration (37 ppm) is greater than the average UCC value (McLennan, 2001), but is closely similar to the NASC value of 35 ppm. Near-constant Ta (1.2 ppm) is slightly enriched compared to UCC (1 ppm: McLennan, 2001). Concentrations of *large-ion lithophile elements* (LILE) vary widely in these rocks, apparently due to greater mobility of these elements during weathering, diagenesis and low-grade metamorphism (Wronkiewicz and Condie, 1987). Rb (127.7 ppm) content is similar to NASC (125 ppm) and close to the revised UCC value of 112 ppm (McLennan, 2001). Ba values are in the range of 403–849 ppm (average 584 ppm) while an average Sr value of ~81 ppm shows a highly depleted pattern with reference to UCC (350 ppm) and PAAS (200 ppm). The average Th value of 17.6 ppm tied to 2.7 ppm of U and 12.5 ppm of Nb are similar to UCC values (McLennan 2001). The Pb value shows an abrupt change in concentration. The concentration of *ferro-magnesian trace elements* (FMTE) deviates within the ±25% range. The average values of V, Cr, Co and Ni are 77, 40, 12 and 30 ppm, respectively, which reflect a systematic change in the concentration levels from Lower to Upper Siwalik successions. These elements show a decrease in their concentrations along the younging path of the stratigraphic sequence. The result of *rare earth elements* (REE) analysis (Table S2: available as electronic supplementary item) and the chondrite and PAAS normalized patterns of mean concentrations in the Lower, Middle and Upper Siwalik are presented in Fig. 4a,b. Total REE plots show a broad scatter, while the chondrite-normalized plots display LREE and HREE enrichment and a sharp negative Eu spike. The $\sum$REE ranges from 158 to 260 ppm (mean 208.5 ppm), substantially higher than the values for UCC, PAAS and NASC, but closely similar to the European Shale value of 204 ppm (Taylor and McLennan, 1985). Siwalik sediments show a moderately high LREE/HREE ratio (5.9–11.3, mean 7.72) and a sharp negative Eu anomaly (0.57–0.79, mean 0.65).

4.5. Constraints of Himalayan granitoids

In the Himalayas, granitoids of different ages occur at various tectonic levels, whose petrogenesis is linked to the tectonic evolution of respective terrains (Sinha-Roy and Bhargava, 1989). Proterozoic granites are well exposed as concordant slabs along the entire spread of the Lesser Himalaya (Islam et al., 2005), either as a belt of deformed granites at the base of the crystalline rocks of the axial zone along the Main Central Thrust (MCT), or as detached tectonic sheets and slivers within the Lesser Himalayan meta-sedimentary sequences (Sinha-Roy and Sen-Gupta, 1986). Published chemical data for the NW Himalayan granitoids have been utilized for comparing the inherited geochemical traits of the crustal protolith.

According to Islam et al. (2005) these Lower Himalayan Proterozoic granites of the Lesser Himalaya are enriched in silica and potash with high A/CNK value (>1.1) and show the presence of normative corundum. These authors have depicted an enriched LREE and moderately depleted HREE pattern with sharp negative Eu anomaly. With the help of a primitive mantle normalized spider diagram, these authors demonstrated lower values of Ba, Nb, Sr, P and Ti and higher concentrations of Rb, Th, and U. The elemental concentration in the Siwalik sediments normalized in respect to Proterozoic Himalayan granitoids, when plotted parallel to the ‘average Proterozoic granites’ of Condie (1993) and UCC of Taylor and McLennan (1985) show marked depletion of Rb, Th, U, Ta and Sr, similarity with Ba, Nb, Ce, Sm and Yb values and large enrichment of Ti (Fig. 5a).

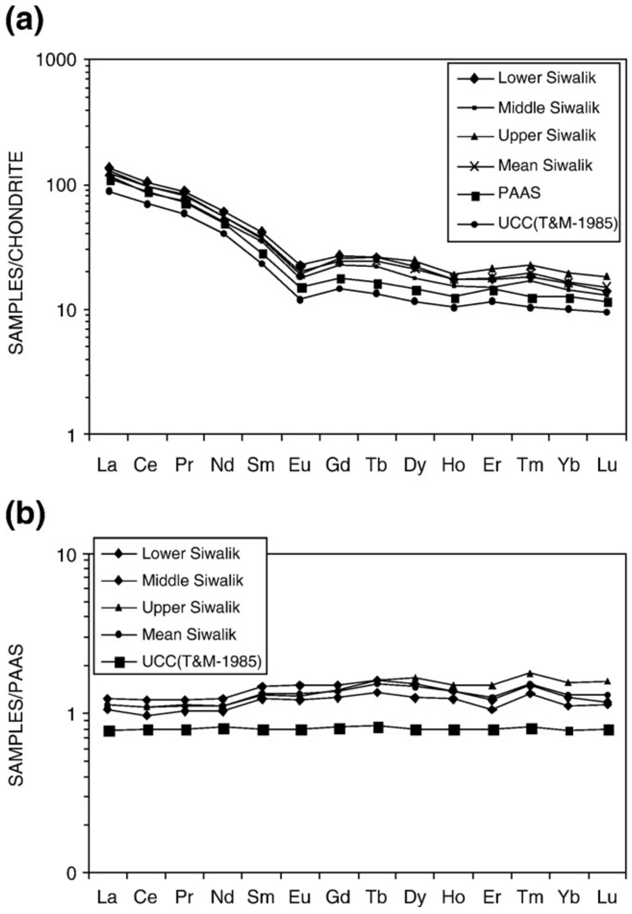

Fig. 4. (a) The chondrite-normalized REE pattern for Siwalik foreland sediments. UCC and PAAS values plotted for comparison. (b) PAAS normalized REE pattern for Siwalik sediments and UCC (Taylor and McLennan, 1985).

Early Paleozoic granites (550 ± 50 Ma) of the NW Himalaya with high A/CNK value (>1), enrichment of LILE and LREE and depletion of Eu show remarkable similarity to the Siwalik sediments in terms of elemental abundance of Th, Nb, Zr, Sm and Y. A Paleozoic granite-normalized multi-element spider diagram of the Siwalik sediments (Fig. 5b) shows marked depletion of Rb, Sr and P.

Tertiary intrusive anatectic leucogranites, the product of the collision tectonics in the Higher Himalayan range, show remarkable uniformity in their structural setting and their geochemical traits. These rocks are enriched in SiO_2, K_2O, Na_2O, P_2O_5 and Rb and depleted in Ba, Th, K, REE, Sr, Ti, and Y concentrations (Islam et al., 2005). Elemental data for the Siwalik sediments normalized to Tertiary leucogranite, when plotted as a spider diagram, show enriched values of Ba, Th, La, Ce, Nd, Sm, Zr, Ti and Y, and are depleted in Rb, Sr and P concentrations (Fig. 5c). A similar pattern with a certain degree of mismatch has been noticed in the UCC trend (Taylor and McLennan, 1985).

5. Discussion

Observable petrographic evidence revealed by the coarse granularity of the average Siwalik sediment points to a prominent high-grade metamorphic provenance. On the other hand, preponderance of quartzolithic components in the Middle and Upper Siwaliks suggests an increasing contribution from plutonic rocks and granitoids with declining influence of the metamorphic provenance. QFL and Qm-F-Lm triangular plots (Fig. 2a and b) when superposed on the provenance fields of Dickinson et al. (1983) show a recycled orogen provenance for the entire Siwalik sedimentary pile, with increasing influence of granitoids. The type of clay minerals present in the upper

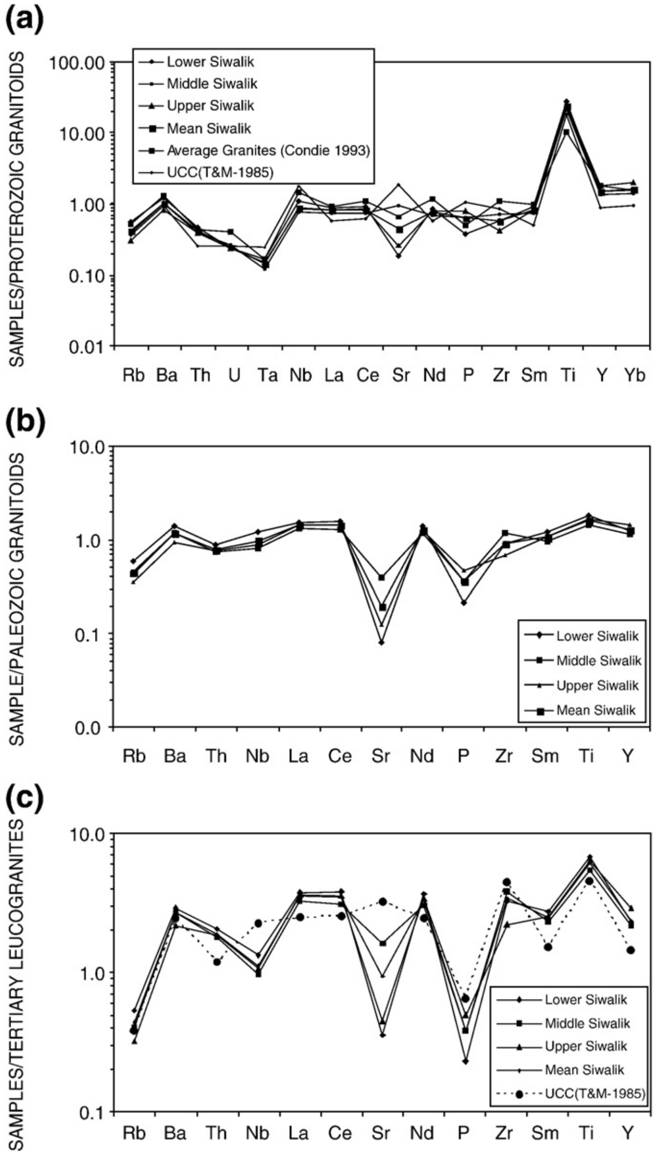

Fig. 5. (a) Proterozoic granitoid (NW Himalaya) normalized spider diagram of the Lower, Middle, Upper and "Mean" Siwalik sediments. Average Proterozoic granites (Condie, 1993) and Upper Continental Crust (Taylor and McLennan, 1985) values plotted for comparison. Average NW Himalayan granitoid data taken from Islam et al. (2005). (b) Early Paleozoic granitoid (NW Himalaya) normalized spider diagram of the Lower, Middle, Upper and "Mean" Siwalik sediments from this study. (c) Tertiary leucogranite (NW Himalaya) normalized spider diagram of the Lower, Middle Upper and "Mean" Siwalik sediments from this study. Upper Continental Crust composition (Taylor and McLennan, 1985) plots for comparison.

part of the Siwalik succession indicates the presence of igneous rocks in the source terrain.

Several studies in the past have shown that sandstones formed in different tectonic settings possess characteristic chemistry. Using Miocene sediments as a proxy, Roser and Korsch (1986) demonstrated that sandstones originating in variable tectonic domains can be successfully discriminated on the basis of SiO_2 content and K_2O/Na_2O ratios after recalculating the composition on CaO free basis. Using these criteria, the chemical composition of the average Siwalik sediment falls in the passive margin field of the (Fig. 6b) discrimination diagram of Roser and Korsch (1986). The K_2O/Na_2O vs. SiO_2/Al_2O_3 plot (Maynard et al., 1982) further confirms the passive margin setting for the source terrain (Fig. 6a).

The major element distribution pattern demonstrated by the elemental data statistics and environment discriminatory plots reveal interesting information about the provenance. As mentioned before, major elements like SiO_2 and Al_2O_3 (Table S1) exhibit large compositional variation (62 to 78% and 10 to 22%, respectively). Likewise, the K_2O/Na_2O ratio shows progressive decline from Lower to Middle Siwalik rocks while SiO_2 content shows an increment along the same path. Such a coupled variation suggests a systematic change in the provenance. A gradual and systematic variation in the alkali ratio is also suggestive of change in the provenance, from high-grade metamorphics to low-grade metasediments. The negative correlation of most major elements with SiO_2 is due to wide variation in the source rock composition. Depletion of CaO and Na_2O with respect to NASC and PAAS reflects moderate to strong weathering and widespread recycling in the source area. Significant depletion of CaO in the Siwalik rocks, particularly in the silica-rich Lower Siwalik sediments suggests a silica dilution effect. As stated before, K_2O, Fe_2O_3, TiO_2 and Al_2O_3 values show gradual increase from top to bottom of the Siwalik succession. Such variations in the lithology of the source terrain suggest an influence of the geological age of the individual unit.

A bivariate plot of most major oxides relative to Al_2O_3 demonstrates a range of correlation factors (Fig. 3): positive correlations for Fe_2O_3 (R^2—0.51), K_2O (R^2—0.78), P_2O_5 (R^2—0.55) and SiO_2 (R^2—0.82) and partial correlations for TiO_2 (R^2—0.38) and CaO (R^2—0.38). CaO in

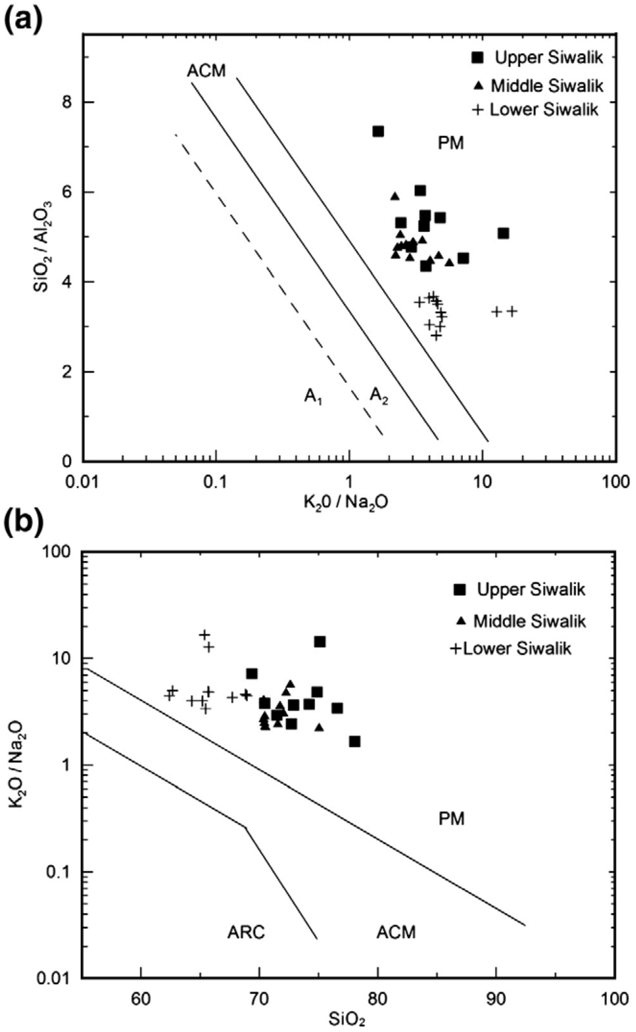

Fig. 6. (a–b) Tectonic setting discrimination diagrams of the Lower, Middle, and Upper Siwalik sediments: (a) K_2O/Na_2O–SiO_2/Al_2O_3 plots (Maynard et al., 1982). (b) SiO_2–(K_2O/Na_2O) plots (Roser and Korsch, 1986). Abbreviations PM = passive margin; ARC = active arc setting: A_1 = arc setting with basaltic and andesitic detritus, A_2 = evolved arc setting with felsic plutonic detritus; ACM = active continental margin. Siwalik sediments plot in the passive margin (PM) field in both the diagrams.

these sediments is primarily derived from the authigenic carbonate minerals, with alkalis originating from the dissolution of feldspars and clays.

Variation in the degree of weathering of the source terrain tends to influence the dispersal of alkali and alkaline earth elements in the resultant terrigenous deposits (Nesbitt et al., 1980; Wronkiewicz and Condie, 1987, and references therein). Elements with larger ionic radii, such as Cs, Rb and Ba, are recorded from many weathering profiles the world over, where they preferentially get exchanged and adsorbed on clay particles. On the other hand, smaller cations such as Na, Ca and Sr get selectively removed in similar environmental conditions (Nesbitt et al., 1980). Marked depletion of Ca, Na and Sr content (Tables 1 and 2) reflects an intensely weathered source terrain. The degree of weathering from the lower to the upper part of the Siwalik sedimentary pile produces tell-tale changes in the intensity corresponding to the changing energy level during the depositional phase. In other words, the grain size factor played a significant role in restricting weathering to a certain level. The dominance of mudstone–sandstone suites and clay beds in the lower part reflects a low energy depositional environment, while the sandstone–conglomerate association in the upper part can be related to a high-energy condition of deposition. Such gradual rise in the energy level of the depositional phases from Lower to Upper Siwaliks was apparently linked to the tectonics affecting the provenance. On the other hand, varying concentration of the mobile elements along the entire stratigraphic profile suggests changing tectonic activity in the source region.

Provenance of the terrigenous sediments is best understood by studying the behavior of least mobile elements during weathering, transport, diagenesis, and metamorphism. Examples of such elements, having relatively short residence times in seawater, are REE, HFSE, Th, Sc, Hf, and Co (Taylor and McLennan, 1985). These elements are believed to get transferred as such into the clastic sediments during weathering and transportation and retain the signatures of the parent material (Bhatia and Crook, 1986; McLennan, 1989; Condie, 1993). Proterozoic and Paleozoic granites in the Higher and Lesser Himalayas which have higher concentrations of these elements may have acted as the source rock for Y- and Th-rich Siwalik sediments. Contrary to the general view that the contribution of granites and gneisses is not significantly perceptible until the youngest detritus was deposited in the Upper Siwalik times (Garzanti et al., 1996; Srikantia and Bhargava, 1998), the geochemical evidence presented here suggests that even Lower and Middle Siwaliks received significant inputs from the granites and gneisses of the Lesser and Higher Himalayas. This inference is of vital significance and constitutes the focal theme of this paper.

The analyses of trace element data show that the ratios like Eu/Eu* (~0.65), La/Lu (~9.04), La/Sc (~3.79), Th/Sc (~1.54), La/Co (~3.59), and Cr/Th (~2.26) are very similar to the sediments derived from the weathering of felsic rocks (Table 1). Relative REE distribution patterns, the extent of the Eu anomaly and LREE/HREE ratio in a particular rock suite are invariably used for provenance interpretations (Taylor and McLennan, 1985; Wronkiewicz and Condie, 1989). The LREE/HREE ratios in the Siwalik sediments (5.9 to 11.3, mean ~ 7.72) and their distribution pattern shows a prominent negative Eu anomaly (0.57–0.79, mean ~ 0.65) similar to the UCC (0.63) and NASC (0.66) values (Gromet et al., 1984; Taylor and McLennan, 1985). A UCC-type source terrain is therefore inferred for rocks of the Siwalik succession. Stratigraphic variations in the LREE/HREE ratios can be explained by sedimentary recycling of diversified lithologies and intense weathering in the source terrain. Enriched LREE and broadly flat HREE pattern with $(Gd/Yb)_n$ ratios ranging from 1.08 to 2.15 (mean ~ 1.56) with a sharp negative Eu anomaly indicates an original felsic provenance, while the negative Eu anomaly on its own reflects differentiated silicic sources, like that of granite (Taylor and McLennan, 1985). The Ce_N/Yb_N value ranges from 3.8 to 9.8 (mean 6.24), similar to the ratio observed in the felsic as well as reworked sedimentary source rocks. In the $Eu/Eu^* - (Gd/Yb)_{cn}$ diagram (Fig. 7) Siwalik samples plot close to the UCC domain. A $(Gd/Yb)_{CN}$ ratio less than 2, except for two samples LS-1, MS-6, suggests these are derived from less HREE depleted Proterozoic sources.

Table 1
Range of elemental ratios in Siwalik foreland sediments compared to ratios in similar fractions derived from felsic rocks, mafic rocks and UCC.

Elemental ratios	Range of sediments from Siwalik Group[a]	Range of sediments from felsic source[b]	Range of Sediments from mafic source[b]	Upper Continental crust[c]
Eu/Eu[b]	0.57–0.79 [0.65]	0.40–0.94	0.71–0.95	0.63
(La/Lu)[d]	5.5–1.43 [9.04]	3.00–27.0	1.10–7.00	9.73
La/Sc	2.1–5.0 [3.79]	2.50–16.3	0.43–0.86	2.21
Th/Sc	0.7–2.2 [1.54]	0.84–2.5	0.05–0.22	0.79
La/Co	2.2–4.9 [3.59]	1.80–13.8	0.14–0.38	1.76
Th/Co	0.9–1.9 [1.46]	0.67–19.4	0.04–1.40	0.63
Cr/Th	1.1–4.77 [2.26]	4.00–15.0	25–500	7.76

[a] This study.
[b] Cullers (1988, 1994, 2000), Cullers and Graf (1984),Cullers and Podkovyrov (2000), Cullers et al. (1988).
[c] McLennan (2001), Taylor and McLennan (1985).
[d] Chondrite-normalized.

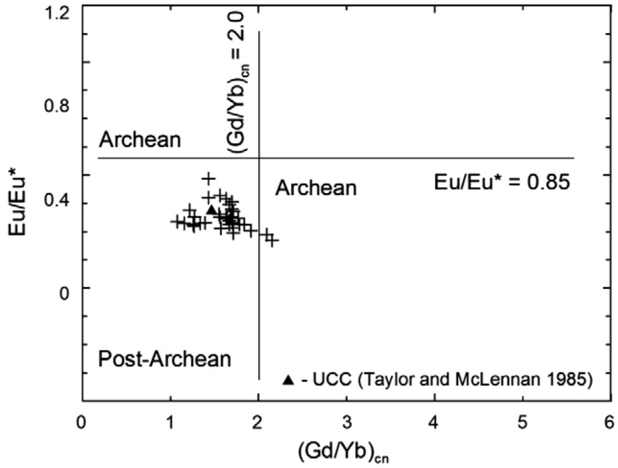

Fig. 7. Plot of Eu/Eu* vs. $(Gd/Yb)_{CN}$ for composite Siwalik sediment samples. Discrimination fields drawn after Taylor and McLennan (1985).

Since hydraulic sorting in the sedimentation basin affects the sediment distribution, grain size and mineralogy control the REE abundance in a sedimentary deposit (Armstrong-Altrin et al., 2004 and references theirin). The variations in the $\sum$REE (158–260 ppm, mean 208.5 ppm) in the Siwalik rocks therefore appear to be governed by the nature of weathering in the source area. Sedimentary recycling in the orogen and the lithological variation in the source terrain could be additional controlling factors. Observations show that all REEs behave as a group of immobile elements during weathering, hence very little enrichment or depletion of REE could be expected. The variation observed in the $\sum$REE content of these foreland basin sediments can therefore be attributed to the abundance of a particular grain size fraction and to the influence of the dilution effect caused by the ubiquitous heavy and clay minerals.

While average Siwalik sediments with 40 ppm Cr and 30 ppm Ni show Cr/Ni ratios of 1.33, the individual sedimentary units (Lower Siwalik: 1.1–2.8 ppm; Middle Siwalik: 0.7–1.5 ppm; Upper Siwalik: 10–1.3 ppm) have variable concentrations. UCC on the other hand contains 83 ppm Cr and 44 ppm Ni (McLennan, 2001) with a Cr/Ni ratio of 1.89. It is well known that the mafic and ultramafic rocks are repositories of high Cr and Ni content. Hence, the low Cr and Ni contents of the Siwalik sediments suggest an insignificant role for the mafic sources. On the other hand, Garver et al. (1996) proposed that argillites with high Cr and Ni values and low Cr/Ni ratios indicate ultramafic rocks in the source region, even though the associated sandstone may show a higher Cr/Ni ratio. Since no ultramafic rocks are known in the potential source regions, the above contention is not tenable for the Siwalik sequence.

The UCC normalized trace element distribution pattern of the Siwalik sediments displays loss of Sr, Pb, Zr and Hf and enrichment of Cs and Ta (Fig. 8) possibly as a result of source terrain weathering and homogenization during mass transport from the orogen to the subsiding foreland basin. Lower Th/Sc and La/Sc ratios than NASC point to higher contribution by the felsic rocks to the Siwalik basin. Triangular discrimination plots using Th–Hf–Co and La–Th–Sc demonstrate an increased felsic contribution with reference to the crustal proxy (Fig. 9). Compared to the revised UCC values (McLennan, 2001) Siwalik sediments show depletion of Sr, Zr, Hf and Pb. The Zr/Hf value (36.53) is remarkably similar to the present day Australian alluvial sediments (36.9 ± 1.4: Kamber et al., 2005). Such a composition is generally ascribed to high rainfall in the catchments area. It is possible that the Siwalik basin was frequently filled with detritus produced during heavy precipitation in the catchment area.

Previous studies have revealed that the concentration in the post-Archean sedimentary rocks does not show any significant change but the U values show gradual decrease, leading to increase in the Th/U ratio with the passage of time (McLennan and Taylor, 1980). The average U and Th concentrations in the Siwalik sediments and Th/U ratio compare favorably to passive margin sediments derived from differentiated upper crust (McLennan et al., 1990). High Th/U ratios of ~6 are caused by the oxidation of U^{4+} to U^{6+} and its removal as soluble $(UO_2)^{2-}$ (McLennan and Taylor, 1980). Recycling of sediments in the depositional basin, repetitive weathering and re-sedimentation in the source area under oxidizing conditions may have resulted in substantial loss of U and consequent increase in the Th/U ratio. The average Th and U concentrations in the UCC are nearly fixed at 10.7 ppm and 2.8 ppm, respectively (Taylor and McLennan, 1985). It is interesting to note that U concentration in the Siwalik sediments is remarkably close to the UCC value of 2.8 ppm in spite of intense weathering in the source terrain and large scale homogenization of the derivative sediments. Higher Th (17.6 ppm) values in the Siwalik sediments can be explained in terms of their derivation from the Higher Himalayan Crystalline sequence, the Proterozoic granitoids (42 ppm) and Paleozoic granites (22 ppm) and gneisses.

Time and again, it has been reiterated that the sedimentary rocks of the Lesser and Higher Himalayas were generated in a Precambrian passive margin setting which subsequently got deformed and was intruded by several granitic bodies in Proterozoic and Paleozoic times (Brookfield, 1993). These passive margin rocks, after the Cenozoic uplift, assumed the status of positive terrain and produced sediments through weathering and erosion which filled up the frontal Tertiary orogen represented today by the clastic rocks of the Siwalik Group. In Western Nepal, Huyghe et al. (2001) estimated $\varepsilon_{Nd}(0)$ values between −14.6 and 19.2 for the Surai Khola and Karnali river sediments which have close similarity with the present day values for the modern Higher Himalayan rocks. Consequently, it can be proposed that the detrital input into the Siwalik sedimentation basin was largely from the Higher Himalayan rocks with minor contributions from the Lesser Himalayan terrain. The decrease in $\varepsilon_{Nd}(0)$ value in the Middle and Upper Siwalik rocks (Huyghe et al., 2001) indicates gradual increase in the proportion of Lesser Himalayan detritus. A similar explanation for the Lower Siwalik rocks was given using Sr–Nd isotopic composition (White et al., 2002) where the ^{87}Sr/^{86}Sr ratio lies confined to the field of the Higher Himalayan Crystalline series.

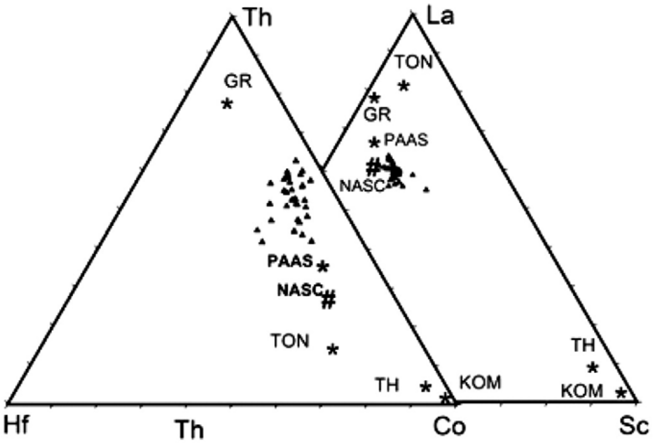

Fig. 9. Distribution of composite Siwalik sediments on Th–Hf–Co and La–Th–Sc diagrams. NASC, PAAS, Tonalite (TON), Tholeiite (TH), Komatiite (KOM) and Granite (GR) plots after Wronkiewicz and Condie (1987).

6. Synthesis

Chemical composition of the Himalayan foreland basin sediments provides an insight into the time-controlled variability in the source terrain through time. Rapid uplift of the proto-Himalayan orogen and accompanying exhumation imparted significant changes in the chemical signature of the sediments deposited in front of the rising mountain chain. The provenance of this Himalayan foreland basin-fill was inferred with the help of the geochemistry of these sediments. Such studies have shown a marked shift in the source terrain

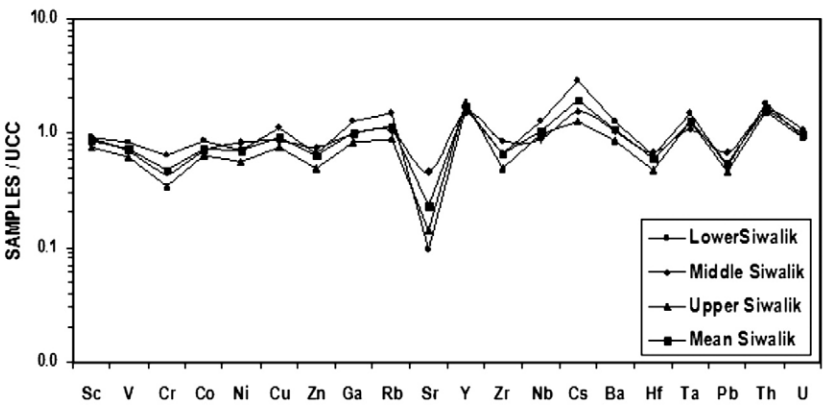

Fig. 8. Trace element concentration in the sediments of the Siwalik Group normalized to Upper Continental Crust revised estimate (McLennan, 2001).

providing sediments to the Subathu, Dagshai, Kasauli and Siwalik basins. For the syn-collisional marginal marine sediments of the Subathu Formation (Paleocene to Middle Eocene), the provenance could be traced to the Indus Suture Zone represented by multicycled sedimentary and distinctly volcanic and ophiolitic lithologies (Najman, 2006). Subsequent continental deposits of the Dagshai and Kasauli Formations reflect an evolution of the growing orogen with minimal volcanic inputs. By the time the Siwalik depression started receiving sediments, the provenance shifted to the Higher Himalayan areas. During formation of the upper parts of the Middle and Upper Siwalik formations, the Higher Himalayan contribution diminished with a switch over to a more prominent Lesser Himalayan source terrain. As recorded in the published literature, a significant increase in the high-grade metamorphic components in the pre-Siwalik Kasauli Formation suggests an initiation of rapid tectonic exhumation of the Higher Himalayan crystalline sequence in early Miocene times. This activity led to the activation of the Main Central Thrust (MCT) resulting in progressive uplift of the deeper crust from the core of a growing orogen (Najman and Garzanti, 2000). On the other hand, subsequent compositional shifts from a quartzolithic source terrain for the Siwaliks to a quartzofeldspathic provenance for the Bengal and Indus Fan (Critelli and Ingersoll, 1994) provide support to the hypothesis of progressive exhumation of the Higher Himalayan crystalline sequence.

Precambrian and Paleozoic granitoids and their metamorphic equivalents were mobilized in different tectono-stratigraphic units of the Higher and Lesser Himalayas possessing distinct chemical composition. These rocks have left their indelible geochemical imprints on the entire Siwalik succession. A substantial part of such granitoids, having the imprints of crustal protoliths was believed to have suffered deformation and was reworked during the collisional stage. On being eroded and deposited in the foreland basin, they imparted well defined chemical signatures to the Siwalik sedimentary package.

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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.gr.2009.07.005.

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