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Clay minerals record from Late Quaternary drill cores of the Ganga Plains and their implications for provenance and climate change in the Himalayan foreland

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ABSTRACT

This study documents the coupling of provenance and climate change over the last 100 ka manifested in clay mineralogy of sediments from two cores (~50 m deep) in the Ganga-Yamuna interfluve in the Himalayan Foreland Basin, India. Depth distribution of the texture and clay mineral assemblage in the two cores show notable differences on account of pedogenesis and sediment supply over the last 100 ka. Core sediments from the northern part of the interfluve (IITK core) are micaceous and dominated by hydroxyl-interlayered dioctahedral low-charge smectitea (LCS) in fine clay fraction but by trioctahedral high-charge smectite (HCS) in silt and coarse clay fractions. In contrast, core sediments from the southern part of the interfluve (Bhognipur core) are poor in mica and both LCS and HCS are recorded in the upper 28 m of the core while the lower part is dominantly LCS in all size fractions. The paleosols in the two cores formed in the sub-humid to semi-arid climatic conditions resulting in clay minerals such as 1.0-1.4 nm minerals, vermiculite, HCS and also preserved the LCS, hydroxyl-interlayered vermiculite (HIV) and pseudo-chlorite (PCh), and kaolin that formed earlier in a humid climate. The preservation of LCS, HIV, kaolin and PCh is a clear indicator of climate shift from humid to semi-arid in the Ganga Plains as their formation does not represent contemporary pedogenesis in the alkaline chemical environment induced by the semi-arid climate. As the simultaneous formation of both HCS and LCS is not possible at the expense of mica, the abundance of LCS sediments from both the cores suggests the role of plagioclase weathering in the formation of LCS. In the upper 28 m of the Bhognipur core, the presence of both HCS and LCS in the fine clays suggests a change in sediment provenance from cratonic to a dominantly Himalayan source during Holocene. The climatic records inferred from the typical clay mineral assemblages of the two interfluve cores are consistent with the Marine Isotope Stages (MIS). The humid interglacial stages (MIS 5, 3, and 1) are marked by dominance of HIV, PCh, and LCS whereas the dominance of HCS together with pedogenic carbonate (PC) is noted in semi-arid stages (MIS 4 and 2). © 2011 Published by Elsevier B.V.

1. Introduction

The Ganga Plains of the northern India constitute one of the world's most extensive alluvial tracts traversed by large rivers such as the Ganga and the Yamuna that are sourced in the Himalayan orogen, as well as rivers such as the Betwa, the Chambal, the Ken and the Son that are sourced in the central Indian Craton and many smaller rivers sourced within the plains. These rivers are distinctive in terms of their source area which in turn translates into distinctive hydrological and sediment transport characteristics (Sinha and Friend, 1994). The Ganga Plains are of great significance as they hold important clues regarding the tectonic and climatic factors that governed the interaction between the Himalayan orogen and the foreland. It is important to track changes in the alluvial landscape on different time

scales as well as their spatial variability which is a function of rainfall variability and hinterland characteristics including tectonic regime (Sinha et al., 2005a). For a comprehensive understanding of the plains, multiple approaches need to be adopted that combine the studies of modern process as well as paleo-landscape development and sedimentation history as recorded in the alluvial stratigraphy. Evolutionary history of most landforms (mega- and meso-scale) in the Ganga Plains remains inadequately understood because of the methodological difficulties associated with the study of subsurface deposits. Such studies require a multi-disciplinary approach including geological, geophysical, geochemical, hydrological, atmospheric, soil and agricultural, ecological and microbiological database and knowledge.

The chemical equilibria of clay minerals are only apparent and in any case ephemeral in nature. It is often difficult to determine as to which minerals are diagnostic of different climatic zones. However, those clay minerals which occur most frequently can be considered to have climatic significance (Tardy et al., 1973). For example, minerals

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such as kaolinite often remain unaltered through subsequent changes in climate, and therefore, may preserve a paleoclimatic record. Singer (1980) indicated that other layered silicates at a less advanced stage of weathering may adjust to subsequent environmental changes and thus may lose their interpretative value for paleoclimatic signatures. There are various processes that are capable of producing distinctive types of clay minerals (Birkland, 1984). Several workers have also considered minerals of intermediate weathering stage as potential indicators of paleoclimatic changes in parts of central India and Gangetic Plains (Pal et al., 1989; Srivastava et al., 1998; Pal et al., 2009).

The study has attempted to reconstruct the Late Quaternary climatic changes and source area variability in terms of sediment supply in the Ganga–Yamuna interfluve using high resolution data on clay mineralogy of sediment samples from shallow cores (~50 m deep) covering a time span of ~100 ka.

1.1. Study area description and methods

The study area constitutes a part of the Ganga–Yamuna interfluve (GYI) in the Ganga Plains (Fig. 1) formed by continuous filling of the Himalayan Foreland Basin through most of the Late Quaternary. The Himalayan foreland is the largest foreland basin in the world consisting of folded sedimentary sequences of the Siwaliks in the north and the vast Ganga Plains in the south (Raiverman et al., 1983; Burbank et al., 1993). The Ganga Basin has been filled with sediments derived from both Himalayan as well as cratonic sources forming several kilometres thick alluvial strata. While a predominance of the Himalayan source has been widely suggested through the Quaternary (Burbank, 1992), Sinha et al. (2009) showed that the contribution from the cratonic source has also been significant during the Late Quaternary. This paper examines the clay mineralogy of core sediments in the northern and southern interfluves in various size fractions and attempts to establish the pathways of the formation of these clay minerals as a function of climatic and source area variability.

The stratigraphic framework of the study window is primarily based on the cliff sections exposed along the river banks of the major rivers (Sinha et al., 2002; Gibling et al., 2005; Sinha et al., 2005b) which was later complemented by drill core studies (Sinha et al., 2005c, 2007a, 2009). This study has utilized two available cores from the G–Y interfluve, ~75 km wide as the crow flies. The core from the northern part of the interfluve was located in the campus of the Indian Institute of Technology Kanpur (IITK) (Fig. 1). Another core at Bhognipur (BHOG) is located in the southern part of the interfluve (Fig. 1). Most parts of the interfluves are monotonously flat as reflected from the relief of only a few metres between the IITK and



Fig. 1. Study area in the Ganga-Yamuna interfluve (GYI) showing drill core at Indian Institute of Technology, Kanpur (IIT, K) and Bhognipur, Kalpi (BHOG, K).

BHOG separated by ~55 km. Modern climatic condition in the study area is marked by monsoonal regime; average annual rainfall is 550–750 mm in IITK region and 450–700 mm in Bhognipur region. Most of the rainfall is received in summer monsoon during June to September (IMD, 2009).

A total of 92 sediment samples from IITK core and 62 samples from the Bhognipur core were collected from different depths and analysed for their texture, pH, electrical conductivity (EC), calcium carbonate (CaCO₃), organic carbon (OC), and the mineralogy of the silt, and clay fractions. Detailed procedures for these analyses are given in the supplementary material.

2. Core lithostratigraphy

The IITK core can be divided into four lithostratigraphic units (Fig. 2a). The lowermost Unit 1 is primarily made up of deposits of floodplain facies with occasional sand and silt beds/patches. This unit represents a distal floodplain environment in the lower parts



Fig. 2. (a) Stratigraphy and paleosol distribution of the IITK drill core. A total of 4 major stratigraphic units and 13 paleosols were identified in this core covering a time span of ~100 ka. The entire core is dominated by muddy sediments with thin silt layers at regular intervals. (b) Stratigraphy and paleosol distribution of the Bhognipur drill core. A total of 6 major stratigraphic units and 10 paleosols were identified in this core. This entire core is distinctly coarser in the lower parts with >10 m sand body representing a major channel.

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(Unit 1A) which grades to a relatively less distal environment in the upper half (Unit 1B). Unit-2 starts with a ~2 m thick bed of yellowish brown micaceous fine sand with some intervening thin beds of floodplain facies. Unit-3 is made up of repeated alternation of thin laminae of brownish fine sand (fluvial) and yellow silt (eolian) within floodplain facies. This unit is interpreted as a channel margin/levee indicating further proximity to the channel than the underlying units. The uppermost Unit-4 starts with a ~70 cm thick silty clay bed containing distinct convolute laminations made of yellow silt and very fine sand. The rest of the unit is made up of floodplain facies along with some intervening silt beds of eolian facies. The unit has a significantly high content of medium to large (1-3.5 cm) kankars throughout the unit, especially near the lower boundary with Unit-3. The IITK core represents a floodplain sequence typical of an interfluve setting, with indications of a gradual change from distal to proximal condition. The distal floodplain condition in the lower half of the core led to intermittent exposure and pedogenesis of the sediments. In the upper part of the core, proximal floodplain conditions prevailed, as characterized by the presence of intermittent sand beds, including a 2 m thick crevasse splay deposit.

Four luminescence dates for the IITK core were reported by Sinha et al. (2007a) and are shown in Fig. 2a. Two dates of 86 ± 7.39 ka and $\sim 63 \pm 4.0$ from Unit 1 correspond to MIS-5 and 4 respectively. Two samples from the upper part of Unit 2 yielded ages of 38.7 ± 3.7 ka and 30.3 ± 3.4 ka both spanning MIS-3.

Based on varying depth functions of macro and micromorphic features and maturity of the pedofeatures, Srivastava et al. (2010) recognised 13 paleosols in the IITK core (Fig. 2a). Except for one *Alfisol* (It12), most of the paleosols (It1–It11 and It13) are *Inceptisol* marked by poorly developed pedogenic features and one by strongly developed pedofeatures. Key micromorphic features of these paleosols are marked by weak pedality, undifferentiated to stipple speckled b-fabric, few thin patchy clay pedofeatures, pedogenic carbonates, and rhizocretes (Srivastava et al., 2010).

The Bhognipur core is located approx. 6 km north of the present day Yamuna river, and is divided into six lithostratigraphic units based on their facies association (Fig. 2b). The lowermost Unit 1 is essentially composed of fine intercalations of floodplain and lacustrine facies with abundant kankars all through. Unit-2 is dominated by sandy facies, and a kankar zone occurs near the bottom of this unit, probably representing basal carbonate gravel lag. The sandy facies in the lower part is made up of medium to coarse-grained, pink sand devoid of any mica and resembles the modern day channel deposit of the cratonic river Betwa, which presently flows ~50 km south of this site. The upper part is micaceous fine sand devoid of any pink grains (feldspar), and is similar to the present day channel deposit of Yamuna and/or Chambal rivers. In-between the sandy beds, thin layers of lacustrine (backswamp) facies are also present suggesting brief interruptions in channel deposition. Unit-3 is made up of intercalations of floodplain, lacustrine and channel facies. Unit-4 is made up mainly of reddish brown silty clay and grades to Unit 5 with a gradual change of colour from reddish brown below to yellowish brown above. Unit-5 is made up of thin laminae of silty clay intervened by clayey beds. The uppermost Unit-6 is made up of a combination of floodplain, eolian and lacustrine facies. The Bhognipur core thus represents an interesting depositional history; this site was initially an active channel belt of a major river which subsequently migrated away from the site leading to deposition of proximal and distal floodplain sediments in the upper parts. The river sand at the bottom of the core has a strong imprint of a cratonic source (pink coloured coarse 'Betwa sand'), whereas the upper part has micaceous sands typical of Himalayan-derived sediments.

Paleopedological investigations of the Bhognipur core have shown 10 paleosols in the ~50 m sequence of the core (Fig. 2b) and they show different pedosedimentary characters in comparison to the paleosols in the IITK core (Srivastava et al., 2010). The transition from the lower feldspar rich sand to quartz and mica dominated sediments is marked by a prominent erosional surface with extensive pedorelicts and papules and very weak pedogenesis at 25–28 m depth. It is overlain by several thick-cumulic paleosols with weakly developed pedofeatures and one mature paleosol with strongly developed pedofeatures in the upper half of the core (Srivastava et al., 2010).

3. Physical and chemical characteristics of the core sediments

The IITK core sediments along with 13 paleosols are generally finegrained, slightly to moderately alkaline, calcareous (2-24%) and nonsaline and also impoverished in organic carbon (<0.5%) (Fig. 3a, b, and c). Texturally, core sediments are predominantly silty clay. An increase in the clay fraction was observed at certain depth intervals (e.g. 0.4–3.4 m, 12.0–15.0 m, 17.5–20.8 m, 29–30.5 m, 40.0–46.2 m) that correspond to lt1, lt2, lt6, lt7, lt10 and lt12 paleosols with weakly to strongly developed pedofeatures. However, an increase in clay fraction at a couple of depth intervals (e.g. 20–25 m, 35–40 m) does not seem to be related to pedogenesis.

The depth distribution of physical and chemical characteristics of the Bhognipur core along with a log of paleosol intervals is presented in Fig. 4 (a, b and c). Core samples down to the depth of about 1.5 m are neutral to slightly alkaline in reaction representing Bh1 paleosol with Inceptisol like characters. However, the remaining part of the core with 9 paleosols (Bh2-Bh9) having Inceptisol, Vertisol and Alfisol like characters (Soil Survey Staff, 2003) is moderately to highly alkaline with a very low concentration of soluble salts. The entire core with 10 paleosols is calcareous and the maximum concentration of CaCO₃ (~20%) is noted in sediments between Bh1 and Bh2 paleosols at about 3.5 m, but organic carbon content is considerably low (<0.5%). The texture of the core varies from sandy loam to silty clay with moderate to high content of clay fraction in some of the paleosols. There is an appreciable increase in clay fraction at depth intervals 1.55-1.86 m, 3.54-8.27 m, 8.37-10.90 m and 12.13-13.84 m that correspond to the occurrences of Bh1, Bh2 and Bh3 paleosols (Srivastava et al., 2010). The Bh3 paleosol occurring between 10 and 14 m depth is marked by strongly developed argillic and vertic horizons (Soil Survey Staff, 2003) whereas the others (Bh1 and Bh2) are marked by weakly developed pedofeatures (Srivastava et al., 2010). The paleosols below 15 m depth (Bh4-Bh10) are marked by a large amount (>30%) of clay that does not appear to have any relationship with pedogenic development. These paleosols are marked by weakly developed syn-depositional pedofeatures (Srivastava et al., 2010).

4. Mineralogical attributes of core sediments

Table 1 shows the major clay minerals identified in core sediments and the diagnostic criteria used for their identification. The following sections describe the results of semi-quantitative analysis of clay mineralogy for both the cores highlighting the spatial as well as temporal variability of clay mineralogy.

4.1. IITK core

The XRD analysis of core sediments from IITK core shows dominance of mica in the silt ($50-2 \mu m$) and coarse clay ($2-0.2 \mu m$) fractions that also contain mixed-layer minerals, smectite, vermiculite, HIV, PCh, kaolin, feldspar and quartz (see Figs. 1–3 in the supplementary material). The fine clay fractions ($<0.2 \mu m$) are dominated by mica and smectite along with vermiculite, HIV, PCh and kaolin. The smectite is predominantly LCS and dioctahedral in nature in fine clay fraction but silt and coarse clay fractions are dominantly HCS. The collapsing characteristics of K-saturated fine clay on heating from 110 °C to 550 °C indicate that most of the LCS has hydroxy-interlayering (Harward et al., 1969).

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Fig. 3. Depth distribution of pH, EC, CaCO₃, OC, particle size distribution and textural class of the IITK core.

The depth distribution of these minerals (Fig. 5) allows us to distinguish four zones in the IITK core. The lowermost *Zone A* (base to 40 m) is characterized by relatively low mica content in all size fractions compared to the upper zones. Both silt and coarse clay fractions show variable but significant amount of vermiculite and <10% kaolin whereas the fine clay fraction is high in LCS. Srivastava et al. (2010) also reported a mature paleosol with pedogenic carbonates from this zone. *Zone B*

(40–25 m) is marked by a gradual decrease in LCS in fine fraction although HCS remains high particularly in the coarse clay fraction. Mica is low in fine clay fraction, variable in coarse clay and high in silt fraction in this zone. This zone also marks the highest but variable contents of vermiculite in all size fractions but kaolin remains low.

Zone C (25–15 m) marks a sharp decrease in LCS in fine clay and also in HCS in coarse clay and silt fractions. In contrast, mica is high in



Fig. 4. Depth distribution of pH, EC, CaCO₃, OC, particle size distribution and textural class of the Bhognipur core.

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Table 1

Identification key for the clay minerals in IITK and Bhognipur cores, Ganga-Yamuna interfluve, India.

Mineral	Diagnostic criteria	Climatic and provenance implications
Mica (biotite/muscovite)	Characteristic peak at 1.0 nm, not affected by heating or glycolation; both biotite and muscovite present if the ratio of 001/002 reflection of 1.0 nm of mica is more than 1.	Himalayan sediments rich in biotite mica derived from granitic rocks
Vermiculite (Vm)	Characteristic peak at 1.40 nm on glycolation which decreases on heating the K-saturated samples with concurrent increase in 1.0 nm peak of mica (Schultz et al., 1971).	Weathering product of biotite mica and their content is more in granitic rocks
Pedogenic chlorite (PCh)	Shows a peak at ~1.38-1.36 nm instead of 1.42 nm (true chlorite) on heating the K-saturated samples at 550 °C.	Continued weathering of vermiculite under humid conditions (through formation of HIV)
Kaolin (Sm/K and Vm/K)	Characteristic peak at 0.7 nm, no shifting on glycolation and heating up to 300 °C and but after heating the K saturated samples to 550 °C, 1.0 nm peak of mica gets reinforced.	Forms from weathering of vermiculite and smectite through hydroxy-interlayering in acidic conditions induced by humid climate. It remains as interstratified smectite/vermiculite-kaolinite stage, mostly as remnant in arid and semi-arid environments.
Low charge smectite (LCS)	Characteristic peak at 1.7 nm on glycolation, shifts to 1.1–1.2 nm on K-saturation and heating to 110 °C; expansion of 1.4 nm peak on glycolation of K-saturated and heated (300 °C) samples; expansion of Ca-saturated samples to 1.8 nm region with glycerol vapour (Harward et al., 1969).	First weathering product from plagioclase feldspar under humid conditions; can be derived from both Himalayan and cratonic sources but a larger amount may suggest a dominant cratonic source
High charge smectite (HCS)	Characteristic peak at 1.7 nm on glycolation, collapses readily to 1.0 nm on K-saturation and heating to 110 °C and gets destroyed in HCl treatment	Derived from weathering of biotite under arid and semi-arid climate; indicates a dominantly Himalayan source
Hydroxyl-interlayered vermiculite (HIV)	Characteristic peak at 1.4 nm on glycolation and collapses not readily to 1.0 nm peak on heating to 110 °C but shows tailings on the low angle side of 1.0 nm peak after heating the K-saturated samples at 550 °C.	Generally occurs in acidic soil conditions induced by humid climate
Hydroxyl-interlayered smectite (HIS)	Characteristic peak at 1.7 nm on glycolation but collapses with tailings to 1.0 nm peak of mica on its low angle side on heating the K-saturated samples at 550 °C.	-do-
Mixed layer minerals (10-14 nm)	Generally found in region between 1.0 and 1.4 nm peak areas on glycolation and also on beating the K-saturated samples	Forms when micas start weathering in ambient conditions
Quartz	Characteristic peaks at 0.334 and 0.426 nm	Mainly in detrital fraction; Himalayan source rich in quartz compared to cratonic source
Feldspar	Characteristic peaks at 0.318, 0.403 and 0.32 nm	Mainly in detrital fraction; cratonic source rich in plagioclase feldspar compared to Himalayan source.

all the size fractions and particularly in the silt fraction. Vermiculite content decreases significantly in coarse clay and silt fractions but remains high in the fine clay fraction. Kaolin is present in this zone but does not exceed 10% in any size fraction. The uppermost *Zone D* (15 m-top) shows high mica content in all size fractions but very significant drop in HCS in both silt and coarse clay fractions. The fine clay fraction shows significant amount of LCS in this zone. This zone also marks a distinct zone of high vermiculite in silt fraction but a corresponding decrease in coarse clay fraction. The fine clay fraction shows an extremely variable content of vermiculite throughout this zone. This is also the zone in which kaolin content frequently exceeds 10% particularly in silt fraction in the upper parts of the core.

4.2. Bhognipur core

In the Bhognipur core, the mineral assemblage of the silt (50– 2 μ m) fraction is marked by mica, mixed-layer minerals (1.0–1.4 nm), vermiculite, PCh, HIV and smectite, kaolin, quartz and feldspars (see Fig. 1 in the supplementary material). Semi-quantitative estimates of these minerals indicate that none of the layer-silicate mineral was dominant (>50%) in this fraction. The mica consists of both muscovite and biotite as evident from the ratio of 001/002 reflection of 1.0 nm peak, which is more than unity (Pal, 2003). In the X-ray diffractograms, both trioctahedral HCS and dioctahedral LCS are recorded (see Table 1 for the criteria used for their identification). The LCS also shows partial hydroxy interlayering because on heating to 550 °C the K-saturated smectites collapsed to 1.0 nm peak but with a broadening at the low angle side of 1.0 nm peak.

The pseudo-chlorite (PCh) (Table 1) described here is not a true chlorite showing a peak at \sim 1.38–1.36 nm instead of 1.42 nm, on heating K-saturated samples at 550 °C. The mineral assemblage in the

coarse clay (2–0.2 $\mu m)$ fraction is similar to those in the silt fraction. Both HCS and LCS are present along with variable contents of mica and kaolin which is stratified with hydroxyl-interlayed minerals (Sm/K and Vm/K, see Table 1). This situation is similar to that of kaolin in the silt fraction, suggesting this to be a part of the sediment. The fine clay fractions (<0.2 μm) contain mica, vermiculite, HIV, PCh, smectite and kaolin.

The depth distribution of mica and smectites allows us to divide the Bhognipur core into five distinct zones (Fig. 6). The lowermost *Zone A* (base to 35 m) has rather limited data points but this is generally low in mica and LCS constitutes the most dominant clay mineral (>60%) in all the size fractions. Coarse clay fraction has some HCS as well. Significant amount of vermiculite (up to 20%) is noted in this zone but kaolin content is low (<10%) in all the size fractions. The overlying *Zone B* (35–28 m) is marked by a sharp increase in LCS content constituting >90% of the total clay mineral assemblage in the fine clay fraction. Mica increases slightly in this zone in all the size fractions. Only silt fraction has some vermiculite (<10%) in this zone and kaolin remains low in all the size fractions.

Zone C (28–19 m) marks a significant change in the type of smectite from LCS to a mixture of LCS and HCS in silt and coarse clay fractions (>20% and >60% respectively) but the fine clay fraction is still dominated by LCS (>90%) in this zone. Mica follows an increasing trend particularly in silt fraction. Vermiculite content picks up again in this zone particularly in the coarse clay fraction (>20%). Kaolin starts increasing in the silt and the coarse clay fraction but the fine clay fraction still has <5% kaolin. *Zone D* (19–2.8 m) is characterized by a sharp decrease in smectite in all the size fractions. In silt fractions, LCS is less than 10% and the coarse clay fraction is still dominated by LCS but a drop from 90% to ~50% is noted towards the top. In contrast,

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Fig. 5. Depth distribution of mica, vermiculite (Vm), pseudo-chlorite (PCh), smectite (LCS and HCS), and kaolin in the silt, coarse clay (CC) and fine clay (FC) fraction of the IITK core.

mica content remains unchanged from the lower zone in all size fractions except a few isolated peaks. Vermiculite content is variable in this zone in all size fractions. Kaolin rises gradually in silt and coarse clay fractions (~15–20%). Srivastava et al. (2010) reported a mature paleosol with pedogenic carbonates from this zone. The uppermost *Zone E* (2.8 m–top) shows a sharp rise in mica content particularly in the coarse clay fraction (up to 50%). Smectites in silt and coarse clay fractions are of HCS type whereas those in the fine fractions are both HCS and LCS. Vermiculite is present in significant amount in this zone and kaolin shows a distinct enrichment in the fine clay fraction but decreases in the coarse clay and silt fractions.

5. Discussion

5.1. Systematic changes in physical and chemical properties in the two cores

The paleosols in both cores are slightly to moderately alkaline, calcareous, non-saline and impoverished in organic carbon but their textural classes are different. The Bhognipur core is primarily sandy in lower parts and silty clay loam in upper parts whereas the IITK core is dominantly silty loam. At certain depths, an increase in the clay content is indicative of an argillic horizon formed by clay illuviation (Soil Survey Staff, 2003), suggesting a phase of landscape stabilization during which pedogenic processes operated. Some of these intervals correspond to mature paleosol layers in the cores inferred from the micromorphological investigations (Srivastava et al., 2010). These mature paleosols of the cores are similar to modern Alfisols of the Ganga Plains with a soil-forming interval of 8000–13,500 years

(Srivastava et al., 2010). The absence of argillic horizon along with increase in clay content suggests the occurrence of detrital clays or paleosols with low maturity developed during short pedogenic intervals. These paleosols of low maturity are similar to the modern Inceptisols of the Ganga Plains with soil forming intervals of 500–2500 years (Srivastava et al., 2010).

Further, the impoverishment of organic carbon, calcareousness of the sediments, and alkaline pH suggest that both the cores experienced dry sub-humid climate during the post-depositional period (Pal et al., 2009). Such climatic conditions induce the precipitation of $CaCO_3$ causing depletion of Ca^{+2} ions in sediment solution and the rise in pH (Pal et al., 2003a, 2009) facilitates the illuviation of clay, thus creating the most conducive conditions for the formation of argillic horizon (Eswaran and Sys, 1979; Pal et al., 2003b). Presence of pedogenic CaCO₃ (PC) along with rare occurrence of non-pedogenic CaCO₃ (NPC), which is generally considered a pedorelict feature (Brewer, 1976), provides evidence of the prevalence of the dry subhumid climate during the post-depositional period (Pal et al., 2009). The mature paleosols with argillic horizons at 10 to 14 m depth of the Bhognipur core and also at 40 to 45 m depth of IITK core show higher amount of CaCO₃ than the overlying and the underlying layers (Fig. 2a and b), suggesting a drier phase within the dry sub-humid climatic conditions.

5.2. Clay minerals as evidence of provenance changes

Depth distribution of clay minerals can be used to infer the sediment provenance and its variability through time. As discussed earlier, the parts of the interfluve under investigations have been filled

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Fig. 6. Depth distribution of vermiculite (Vm), pseudo-chlorite (PCh), smectite (LCS and HCS), and kaolin in the silt, coarse clay (CC) and fine clay (FC) fraction of the Bhognipur core.

by sediments from both Himalayan as well as cratonic sources, which are quite distinctive in terms of their clay mineralogy. While the Himalayan source is dominantly micaceous and consists of metamorphic and granitic rocks, the cratonic source, represented by Deccan Basalt and Vindhyans is characterized by dioctahedral smectites (LCS) which form as the first weathering product of plagioclase (Pal and Deshpande, 1987). In contrast, the smectites in the Himalayan source, if present, are generally trioctahedral (HCS) formed from the weathering of biotites (Srivastava et al., 1998).

The IITK core is highly micaceous throughout in contrast to the Bhognipur core, which is poor in mica particularly in the lower parts of the core (>10 m depth). The LCS dominates in fine clay fraction of the IITK core and HCS in silt and coarse clay fractions throughout the core. In the Bhognipur core, the presence of both HCS and LCS in the upper 28 m depth and almost exclusive presence of LCS beyond this depth suggests that the sediment supply at Bhognipur changed from a dominantly cratonic source (characterized by low mica and high LCS) in the lower part to a Himalayan source (characterized by higher mica and HCS) in the upper part. This interpretation is based on the fact that the formation of such a large amount of smectite at the expense of mica in semi-arid climate is improbable (Pal et al., 1989; Bhattacharyya et al., 1993; Ray et al., 2006; Pal et al., 2009).

Further, as the biotite mica cannot yield both trioctahedral HCS and dioctahedral LCS simultaneously, the genesis of LCS and its accumulation in these two cores needs to be viewed in terms of the influence of weathering products of the plagioclase present in both Deccan basalt of Central Indian Craton (Pal and Deshpande, 1987) and micaceous Himalayan river sediment. This smectite must have been carried through the cratonic (Betwa, Chambal, Ken and Son) as well as the Himalayan (Ganga–Yamuna) rivers to Bhognipur and Kanpur areas in the past. Low amount of kaolin in the fine clay fraction in the Bhognipur core suggests that it may not have formed during the post-depositional period but may have resulted due to physical communition of larger kaolin particles present in the coarse silt fractions derived from the Deccan basalt of the Craton. Unlike large amount of interstratified (Sm/K) minerals reported from the Holocene soils of the Ganga Plains (Srivastava et al., 1998), the Bhognipur core sediments and the paleosols show only a small amount of Sm/K. It is likely that the Sm/K formed along with the LCS during the weathering of the Deccan basalt under humid climatic condition (Pal and Deshpande, 1987) and was transported to the Ganga Plains over a short period of time.

5.3. Pathways of clay mineral transformations and implications for climate changes

Although the chronological data for these cores is very limited, an attempt has been made to associate transitions in clay mineralogical assemblages through the cores to major climatic events during the last ~100 ka. The mineralogical composition of core sediments shows significant variability in different size fractions and in depth distribution. For example, the contents of mica and vermiculite are generally higher in silt fraction than in fine clay fractions whereas that of smectite is higher in fine clay fraction. Further, the Bhognipur core shows the presence of trioctahedral HCS in the silt and coarse clay fractions in the upper 28 m (Fig. 6) below which the dominance of LCS is reported. On the other hand, the IITK core has predominantly trioctahedral HCS in both silt and coarse clay fractions all through the core (Fig. 5). The fine clay fractions of both the cores contain predominantly LCS with low amount of HCS (Figs. 5 and 6).

A common feature in both the IITK and Bhognipur cores is therefore the increasing amount of HCS from the silt to the clay

fractions at the expense of mica and vermiculite. This suggests early stages of weathering of biotite to mixed-layered minerals containing vermiculite layers. As more interlayer regions were affected by weathering there was a progressive formation of vermiculite and HCS. Previous studies (Pal, 2003) indicated that the HCS of trioctahedral type is generally an alteration product of biotite mica even in presence of muscovite and can be regarded as an alteration product in semiarid and arid climate. Micromorphological studies on pedality and formation of argillans can complement this observation. In some core samples where clay illuviation is observed, major alteration of biotite to vermiculite and smectite presumably occurred during the postdepositional period.

In the Bhognipur core, the dominance of LCS along with some HCS below 28 m depth (Zones A and B) indicates that the source of LCS may not be the dioctahedral muscovite mica because the weathering of muscovite is very sensitive to potassium in soil solution. Biotite releases considerable amount of K in soil solution, and as a result, weathering of muscovite is inhibited (Pal, 2003). Thus, the formation of HCS and LCS simultaneously from mica is very unlikely (Pal et al., 1989; Ray et al., 2006). Moreover, in dry environments of sub-humid and semi-arid climate that facilitates the formation of CaCO₃ from plagioclase (Srivastava et al., 2002), micas may not yield so much LCS as observed in both the cores (Pal et al., 2009). We therefore suggest that the large amount of LCS formed in an earlier humid climate in the source area as an alteration product of plagioclase (Pal et al., 1989). Plagioclase feldspars are present in both the Deccan basalt of the Central Indian Craton and also in the Himalayan hinterland, which contain sufficient silica to form smectite in tropical humid climates (Tardy et al., 1973; Pal et al., 1989; Srivastava et al., 1998).

Further, the LCS also dominates in the fine clay fraction in the IITK core but its content (25-75%) is less than that in the Bhognipur core (30–95%). This is more explicit when the LCS content in the fine clay fraction is calculated on fine earth basis (<2 mm) as Bhognipur core sediments contain more amount of clay than the IITK core. This is due to the impoverishment of plagioclase in the Himalayan rocks as compared to the Deccan Basalt. On the other hand, the formation of smectite from biotite is quite unlikely in humid climate because in this climate silica and cations are removed thereby inducing separation of layers precluding the formation of clay minerals more siliceous than biotite itself (Tardy et al., 1973). However, during this weathering some amount of vermiculite could have transformed to HIV, which may have further weathered to form PCh because hydroxy-interlayering in vermiculite and smectite generally occurs in acidic soil conditions (Jackson, 1964) under humid climate. Thus, the HCS existing in arid climate is the alteration product of biotite, which survived earlier weathering. The aridity appears to have reduced leaching and helped concentrate silica in solution for the formation of HCS. However, the formation of smectite did not continue in humid tropical climate as evidenced from the presence of very small amount of kaolin (Sm/K) in the fine clay fractions of Bhognipur core (Fig. 6), and also from the absence of kaolin (Sm/K) in the fine clay fractions of IIT K core (Fig. 5). In the event of prolonged weathering of smectite in humid climate, the content of kaolin (Sm/K) should have been dominant (Bhattacharyya et al., 1993).

The presence of HIS, HIV and PCh in the fine clay fractions, and HIV and PCh in the silt and coarse clay fractions indicates that the hydroxyinterlayering in the vermiculite and smectite interlayers did occur when positively charged hydroxy-interlayer materials such as [Fe₃ $(OH)_{6}]^{3+}$, $[Al_{6}(OH)_{15}]^{3+}$, $[Mg_{2}Al(OH)_{6}]^{+}$, $[Al_{3}(OH)_{4}]^{5+}$, etc. (Barnhisel and Bertsch, 1989) entered into the inter-layer spaces at pH much below 8.3 (Jackson, 1964). Moderately acidic conditions are optimal for hydroxy-Al interlayering of vermiculite and smectite and the optimum pH for interlayering in smectite and vermiculite is 5.0-6.0 and 4.5-5.0 respectively (Rich, 1968) as small hydroxyl ions are most likely to be produced at low pH (Rich, 1960). The pH of the sediments from both the cores is well above 7.6 throughout suggesting mildly to moderately alkaline conditions under which 2:1 layer silicates suffer congruent dissolution (Pal, 1985) discounting hydroxy-interlayering of smectites during the post depositional period. The hydroxy-interlayers in vermiculite and smectite and the subsequent transformation of vermiculite to PCh therefore do not represent contemporary pedogenesis in the prevailing dry climatic conditions. However, the crystallinity of LCS is being preserved in the non-leaching environment of the arid climatic conditions (Pal et al., 2009). This suggests that the presence of HIS and also HIV, and PCh in arid and semi-arid climatic environments could be used as an indicator of climate change from humid to arid (Pal et al., 2009). The alkaline chemical reaction, formation of CaCO₃, formation of trioctahedral HCS, preservation of HIS, HIV and PCh, indicate the role of climate change from humid to arid during the development of soils within the cores.

5.4. A model for clay mineral formation and transformation in interfluve sequences of the Ganga Plains in response to climate change over the last 100 ka

In view of clay mineral distribution in the two interfluve cores that is related to sedimentation and pedogenesis in Ganga Plains over the last 100 ka, a summarized account of clay mineral formation and their transformation in response to the climatic changes during this period is proposed for three major climatic transitions namely, Marine Isotope Stage (MIS) 5-4, 3-2 and 2-1 (Fig. 7).

(a) MIS 5-4 transition:

The available climatic reconstructions (Prell and Kutzbach, 1987; Clemens and Prell, 2003) and earlier studies in this region (Gibling et al., 2005; Sinha et al., 2007b; Gibling et al., 2008) suggest that the MIS 5-4 transition is marked by weakening of the SW monsoon after a prolonged humid phase during MIS 5. The lower part of the IITK core is marked by the transformation [*Biotite* \rightarrow *HIV* \rightarrow *PCh*] as the dominant process and [*Plagioclase* \rightarrow *LCS*] as the sub-dominant process in a relatively humid climate and slightly acidic pedogenic environment (Fig. 7a). Later, the semi-arid climate in MIS 4 was dominated by transformations [Biotite \rightarrow HCS] and [Plagioclase \rightarrow Pedogenic carbonate (PC)] in moderate to slightly alkaline pedogenic environment (Pal et al., 1989; Srivastava et al., 2002).

In the Bhognipur core, the formation of the mature paleosol (Bh3 paleosol, Zone D) shows [Plagioclase $\rightarrow LCS$] as the dominant and [*Biotite* \rightarrow *HIV* \rightarrow *PCh*] as the subdominant clay mineral transformations indicative of humid climate and acidic pedogenic environment. This is attributed to the dominance of plagioclase in the Bhognipur core in contrast to that of mica in the IITK core. In the upper part of Zone D in the Bhognipur core, the dominant clay mineral transformations are recorded as [*Plagioclase* \rightarrow *PC*] and [*Biotite* \rightarrow *HCS*] which suggest semi-arid climate and alkaline pedogenic environment (Fig. 7a).

It is important to note however that the formation of LCS, HIV, and PCh is not a part of contemporary pedogenesis in the prevailing semi-arid climatic conditions. Instead, they formed in the source area when the climate was relatively more humid and were transported and preserved as such in the Ganga Plains when the climate changed to semi-arid.

(b) MIS 3–2 transition:

Further up in both the interfluve cores, a dominance of the Himalayan source and frequent development of immature paleosols are noted that are related to MIS 3-2 transition. The clay mineral transformations during the initial phase of this transition are characterized by [Biotite \rightarrow HIV \rightarrow PCh] and [*Plagioclase* \rightarrow *LCS*] in both the cores in a relatively humid climate and slightly acidic pedogenic environment (Fig. 7b).

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Fig. 7. A schematic climate driven model for clay mineral alteration for the last 100 ka in northern and southern parts of the G-Y interfluve represented by IITK and Bhognipur cores.

The change to semi-arid climate during MIS 2 is marked by the transformations of [*Plagioclase* \rightarrow *PC*] and [*Biotite* \rightarrow *HCS*] in a moderately alkaline pedogenic environment in both the cores (Fig. 7b).

(c) MIS 2–1 transition In the uppermost parts of both the cores, the formation of weakly developed cumulative paleosols suggests that sedimentation reached to a steady state. A climatic shift from semi-arid to humid conditions related to MIS 2–1 transition is inferred on the basis of the shift from the dominance of the [*Biotite* \rightarrow *HCS*] and [*Plagioclase* \rightarrow *PC*] transformations to [*Biotite* \rightarrow *HIV* \rightarrow *PCh*] and [*Plagioclase* \rightarrow *LCS*] (Fig. 7c). While the former indicates

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slightly alkaline pedogenic environment during semi-arid conditions of MIS 2, the latter suggests a relatively humid climatic condition during Holocene time.

6. Conclusions

We conclude that the subtle spatial differences in clay mineralogy and their depth distribution in cores from northern and southern parts of the Ganga–Yamuna interfluve are manifestations of coupling of sediment source and climatic transitions. The influence of micaceous sediments of the Himalayan orogen in the upper part of both the cores is reflected in dominance of HCS whereas the lower part of the core from the southern part of the interfluve is characteristically LCS-rich. Since the formation of large amounts of LCS at the expense of biotite from either Himalayan or cratonic sources is not possible and also the hydroxy-interlayering in smectite is improbable in the alkaline soil environments of the present-day semi-arid climate, this reflects a major change in sediment supply in the southern interfluve from cratonic to Himalayan source.

Distinct clay mineral transformations are recorded in both the cores under humid and arid climatic conditions during the last ~100 ka. Typical clay mineral assemblage in a humid climate (e.g. MIS 5, 3 and 1) is marked by HIV, LCS and PCh formed under acidic soil conditions. In a drier climate (MIS 4 and 2), formation of trioctahedral HCS from biotite weathering and precipitation of pedogenic CaCO₃ were the dominant processes that created conducive conditions for illuviation of clays forming argillic (Bt) horizon. Thus, the presence of minerals such as LCS, HIV and PCh in soils of MIS 4 and 2 indicates that these minerals were preserved in the subsequent drier climates.

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