Chapter 7
Clay and Other Minerals in Soils and Sediments as Evidence of Climate Change

Abstract Identification of paleoclimatic signatures in paleosols is a major challenge to soil scientists. It is realized that the clay minerals of the paleosols are potential promising materials for documenting and resolving a wide spectrum of different genetic environments and reactions. It is often difficult to determine which soil minerals are characteristic of different climatic zones as the environment itself changes over time with consequent further modification of mineral assemblage and this is particularly true for clay minerals. Clay minerals such as kaolinite often remain unaltered through subsequent changes in climate, and therefore, may preserve a paleoclimatic record. 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. However, Indian soil scientists, clay mineralogists and earth scientists indicate that minerals of intermediate weathering stage can act as potential indicators of paleoclimatic changes in parts of central India and Gangetic Plains. They have demonstrated how secondary minerals like di- and trioctahedral smectites (DSm and TSm), smectite-kaolinite interstratified minerals (Sm/K), hydroxy-interlayered smectite (HIS), hydroxy-interlayered vermiculite (HIV), pseudo-chlorite (PCh) of intermediate weathering stage, and $CaCO_3$ of pedogenic (PC) and non-pedogenic (NPC) origin can be regarded as potential indicators of paleoclimatic changes in major soil types of India and also in paleosols of the alluvial sediments of the Himalayan river systems and Cratonic source from Peninsular India.

Keywords Clay minerals · Soils · Sediments · Climate change · Polygenetic soils

7.1 Introduction

Perhaps majority of soils contain elements, which formed as a result of environmental conditions that have now altered. Paleosols have become standard tools in the study of Quaternary sequences and such studies are well documented. Paleosols are also abundant in the pre-Quaternary geological record and even Precambrian

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types are now widely recognized. Paleopedological research unravels the signatures of climate change that generally remain stored in soils and sediments of the past (Pal et al. 2000a) and such soils are known as paleosols, formed on a landscape of the past (Valentine and Dalrymple 1976). The study of paleosols is still in infancy but studies on paleosols have caught the attention of the pedologists, sedimentologists and soil mineralogists in India and abroad (Singer 1980; Beckmann 1984; Jenkins 1985; Fenwick 1985; Wright 1986; Pal et al. 1989, 2001, 2009, 2012a; Srivastava et al. 1998, 2007, 2009, 2010, 2013).

Identification of paleoclimatic signatures in paleosols is a major challenge to soil scientists. Yaalon (1971) points to the fact that the products of the self-terminating, irreversible reactions such as calcareous or siliceous incrustations are among the most permanent and best indicators of paleo-environmental conditions. Whenever reliable paleoclimatic indicators, such as paleontological remains, pollen or isotope chemistry were absent or have failed, paleoclimatologists have turned to paleosols for clues as to the nature of climates of the past (Singer 1980). It is realized that the clay minerals of the paleosols are potential promising materials for documenting and resolving a wide spectrum of different genetic environments and reactions (Keller 1970). The use of clay minerals in paleosols and saprolites (weathering profiles) for the purpose of paleoclimatic interpretation is based on five assumptions (Singer 1980) and they are (i) clay minerals and climatic parameters (rainfall and temperature) are quantitatively related, (ii) clay minerals are stable and do not change as long as the climate remains stable and tectonic rejuvenation does not take place, (iii) clay mineral assemblages are uniform throughout the weathering profile, (iv) clay minerals are stable (post-burial stability) and (v) clay minerals have a uniform sensitivity towards environmental change. Singer (1980) further stated that since only soils are in direct contact with atmospheric agencies, the impact of climatic variables on soil clay formation is also most direct.

It is well known that equilibria are only apparent and in any case ephemeral in nature. Therefore, it is often difficult to determine which soil minerals are characteristic of different climatic zones. However, those clay minerals which occur most frequently can be considered to have climatic significance (Tardy et al. 1973). Environment itself changes over time with consequent further modification of mineral assemblage and this is particularly true for clay minerals (Jenkins 1985). Clay minerals such as kaolinite often remain unaltered through subsequent changes in climate, and therefore, may preserve a paleoclimatic record. 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 (Singer 1980). However, recent research records of Indian researchers indicate that minerals of intermediate weathering stage can act as potential indicators of paleoclimatic changes in parts of central India and Gangetic Plains (Pal et al. 1989, 2009, 2012a; Srivastava et al. 1998, 2013). These authors have demonstrated how secondary minerals like di- and trioctahedral smectites (DSm and TSm), smectite-kaolinite interstratified minerals (Sm/K), hydroxy-interlayered smectite (HIS), hydroxy-interlayered vermiculite (HIV), pseudo-chlorite (PCh) of intermediate weathering stage, and $CaCO_3$ of pedogenic (PC) and non-pedogenic

(NPC) origin can be regarded as potential indicators of paleoclimatic changes in major soil types of India and also in paleosols of the alluvial sediments of the Himalayan river systems and Cratonic source from Peninsular India.

7.2 Di- and Tri-Octahedral Smectite as Evidence for Paleoclimatic Changes

Pal et al. (1989) reported the presence of fairly well crystallized dioctahedral smectites as the first weathering product of Peninsular Gneiss, which partly transformed to kaolin (interstratified hydroxy interlayered smectite and kaolinite, KI-HIS) in ferruginous soils (Alfisols) formed in a pre-Pliocene tropical humid climate. Careful examination confirms that such kaolin is not a discrete kaolinite as XRD diagrams of its Ca-saturated and glycolated sample indicates the broad base of 0.72 nm peak and tails towards the low angle. On heating the K-saturated sample at 550 °C, the 0.72 nm peak disappears, confirming the presence of kaolin and simultaneously reinforces the 1.0 nm region at much higher degree even in presence of 1.4 nm minerals. Thus it confirms the presence of KI-HIV/HIS (kaolinite interstratified with hydroxy-interlayered vermiculite, HIV or smectite, HIS) (please refer to Fig. 3.4b). When the termination of humid climate during the Plio-Pleistocene transition did occur, both these clay minerals were preserved to the present. Therefore, the ferruginous Alfisols overlying the saprolites dominated either by dioctahedral smectite or kaolin qualify to be relict paleosols (Pal et al. 1989; Chandran et al. 2000). During the Plio-Pleistocene transition period, such paleosols have been affected by the climatic change from humid to drier conditions. Effect of such climate shift is evidenced by the formation of trioctahedral smectite in the present dry climate from the sand and silt size biotite (please refer to Fig. 3.6a), which survived weathering during the earlier humid climate. This trioctahedral smectite is truly high charge smectite or low charge vermiculite that expands to 1.7 nm on glycolation of Ca-saturated sample but contracts readily to 1.0 nm on K-saturation and heating to 110 °C. The presence of both di- and trioctahedral smectite in relict paleosols is common as reported from ferruginous Alfisols of southern India (Pal et al. 1989; Chandran et al. 2000). This is a unique combination in soils, which helps in identifying the climate change in the geological past. The present day warm semi-aridic climatic conditions also favoured the formation of pedogenic calcium carbonate (PC) (please refer to Fig. 3.6b) by inducing the precipitation of $CaCO_3$ with a concomitant development of subsoil sodicity (Pal et al. 2000b, 2012a). These relict paleosols did preserve the dioctahedral smectite of earlier humid climate, and trioctahedral smectite and PC of the present semi-arid climate. Therefore, these relict paleosols qualify to be polygenetic soils with strong paleoclimatic potential (Pal et al. 1989).

7.3 Red and Black Soils in Semi-arid Climatic Environments

In semi-arid region of southern Peninsular India, the occurrence of spatially associated red ferruginous (Alfisols) and black (Vertisols) soils on gneiss under similar topographical conditions is very common (Pal and Deshpande 1987). Ferruginous soil clays consist mainly of kaolin and smectite whereas the low charge dioctahedral smectite is dominant in black soil clays. The inverse relation between kaolin and smectite with pedon depth of ferruginous soil clays (please refer to Fig. 3.2b) (Pal et al. 1989) indicated the transformation of smectite to kaolin but such transformation is not feasible in slightly acid to moderately alkaline soil reaction, which is a characteristic of the prevailing semi-arid climate. Moreover, the arid climate cannot yield the huge amount of smectite required for the formation of black soils (Vertisols) (Bhattacharyya et al. 1993). Earlier studies in southern Peninsular India (Murali et al. 1978; Rengasamy et al. 1978) suggested that kaolinite was formed in an earlier geological period with more rainfall and great fluctuations in temperature, as evidenced by the presence of granitic tors all around such area (Pal and Deshpande 1987). Therefore, the smectite of Vertisols formed in the earlier humid climate, which was detached from the weathering gneissic rock and transported downstream and deposited in low-lying areas following the landscape reduction process (Bhattacharyya et al. 1993). Due to this process the typical Vertisols were developed in the micro depressions (please refer to Fig. 2.4). After the peneplanation, red ferruginous soils on stable surface continued to weather to form kaolin mineral as the stability of the smectite was ephemeral in tropical humid climate (Bhattacharyya et al. 1993). Termination of the humid climate did occur during the Plio-Pleistocene transition, and due to the termination smectite and kaolin could be preserved to the present day (Pal et al. 2000a).

7.4 Clay Minerals in Soils of the Indo-Gangetic Plains (IGP)

Srivastava et al. (1998) reported the transformation of clay minerals in a soil chrono-association comprising 5 fluvial surfaces (QGH1 to QGH5) of the IGP between Ramganga and Rapti rivers. They demonstrated that pedogenic smectite-kaolinite (Sm/K) can be considered as a potential indicator for Holocene climate changes from arid to humid conditions. The ages of QGH1 to QGH5 are <500 year BP, >500 year BP, >2500 year BP, 8000 Cal year BP and 13,500 Cal year BP, respectively. During soil formation, two major regional climatic cycles are recorded, and they are relatively arid to semi-arid cycles between 10,000–6500 year BP and 4000 year BP till present and were punctuated by a warm and humid climate. During arid climates biotite weathered to trioctahedral vermiculite and smectite in the soils that was unstable and transformed to Sm/K (please refer to

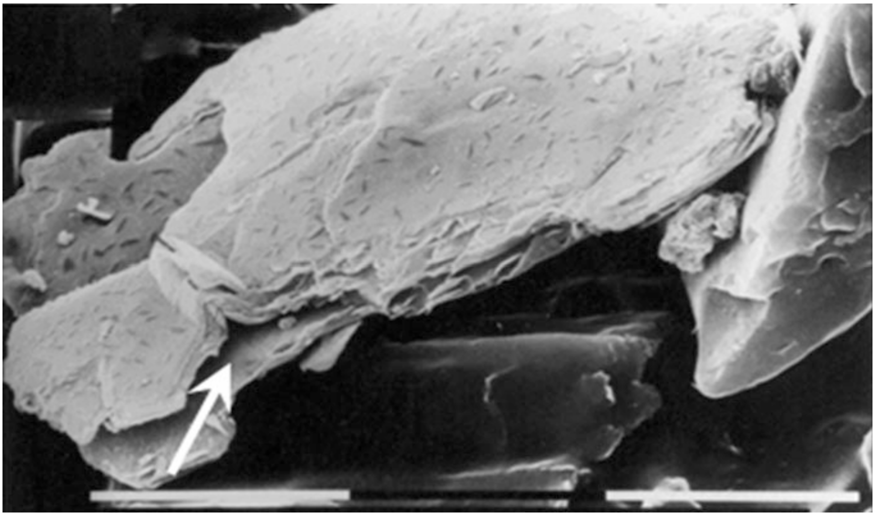

Fig. 7.1 SEM photograph of sand size biotite in soils of the IGP showing the formation of vermiculite (Adapted from Srivastava et al. 2013)

Fig. 4.3) during the following warm and humid climate phase (7400–4150 Cal year BP). When the humid climate terminated, vermiculite, smectite and Sm/K were preserved to the present. During the hot semi-arid climate that followed the humid climate, transformation of biotite into its weathering products like trioctahedral vermiculite and smectite did continue (Fig. 7.1). In arid to semi-arid climate weathering of plagioclase feldspar yielded $CaCO_3$ in soils, which is designated as pedogenic $CaCO_3$ (PC). Initiated by the formation of PC, fine clay vermiculite and smectite translocated downward in the profile as Na-clay, to make soils calcareous and sodic (Pal et al. 2003). This pedogenetic process with time becomes an example of self-terminating process (Yaalon 1971). Presence of Sm/K, trioctahedral smectite and PC in the IGP soils older than 2500 year. BP exhibit their polygenetic features (Srivastava et al. 1998).

7.5 Vertisols, Carbonate Minerals and Climate Change

In the Deccan basalt area, smectitic Vertisols occur in humid tropical (HT), sub-humid moist (SHM), sub-humid dry (SHD), semi-arid moist (SAM), semi-arid dry (SAD) and arid dry (AD) climatic environments (Pal et al. 2009). Smectites are ephemeral in HT climate as they readily transform to kaolinite (Pal et al. 1989; Bhattacharyya et al. 1993). The formation and persistence of Vertisols in HT climate has been possible because the soils are endowed with Sm/K and Ca-zeolites. These two minerals created a favourable chemical environment necessary for the formation and persistence of Vertisols in lower topographic situation

(Bhattacharyya et al. 1993, 1999). As for the formation of Vertisols huge amount clay smectite as parent material is a mandatory requirement and such amount clay smectite cannot be generated in dry climates. Therefore, it is difficult to understand the formation of Vertisols in SHM, SHD, SAM, SAD and AD climates especially when the weathering of primary minerals contributes very little towards the formation of smectites in these climatic environments (Srivastava et al. 2002; Pal et al. 2009). XRD analysis of fine clays (Fig. 7.2) indicates that smectites of Vertisols from sub-humid to arid climates are fairly well crystallized as evident from a regular series of higher order reflections and do not show any sign of transformation except for hydroxy-interlayering (HI) in the smectite interlayers (Pal et al. 2000a; Srivastava et al. 2002). Such interlayering did also occur in vermiculite of the silt and coarse clay fractions (please refer to Fig. 2.9) that further transformed to the formation of pseudo- chlorite (PCh).The hydroxy-interlayering in smectite interlayers is identified from the broadening of the low angle side of the collapsed 1.0 nm peak of K-saturated smectite heated to 550 °C (Fig. 7.2). PCh is not a true chlorite as it shows a broad peak around 1.4 nm when K-saturated sample is heated to 550 °C (please refer to Fig. 2.9). Thus the presence of hydroxy-interlayered dioctahedral smectite (HIS), hydroxy-interlayered vermiculite (HIV, as an alteration product of biotite mica) and PCh (as a transformation product of HIV) is a unique combination of secondary minerals in size fractions of Vertisols (Pillai et al. 1996; Pacharne et al. 1996; Vaidya and Pal 2003). The hydroxy-interlayering in the vermiculite and smectite occurs when positively charged hydroxy-interlayer materials such as $[Fe_3(OH)_6]^{3+}$, $[Al_6(OH)_{15}]^{3+}$, $[Mg_2Al(OH)_6]^{+}$, and $[Al_3(OH)_4]^{5+}$ (Barnhisel and Bertsch 1989) enter into the inter-layer spaces at pH much below 8.3 (Jackson 1964). Moderately acidic conditions are optimal for hydroxyl 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). It is to be noted that the pH of the majority of Vertisols of sub humid to arid climates is either near to neutral or well above 8.0 throughout the profile. In mildly to moderately alkaline conditions of soils, 2:1 layer silicates suffer congruent dissolution (Pal 1985) and thus discounts the hydroxy-interlayering of smectites and vermiculites during the post depositional period of the basaltic alluvium (Pal et al. 2012a), likewise the subsequent transformation of vermiculite to PCh. The mechanism of the formation of HIS, HIV and PCh, described here, does not represent contemporary pedogenesis of Vertisols in the prevailing dry climatic conditions (Pal et al. 2012a). Vertisols of sub humid to arid climates have both NPC (relict Fe–Mn coated carbonate nodules) and PC (pedogenic $CaCO_3$) (Pal et al. 2000b, 2009). Based on ^{14}C dates of carbonate nodules, Mermut and Dasog (1986) concluded that Vertisols with Fe–Mn coated $CaCO_3$ are older soils than those with PCs that are formed in soils of dry climate (Pal et al. 2000b). NPCs were therefore, formed in a climate much wetter than the present, which ensured adequate soil water for reduction and oxidation of iron and manganese to form Fe–Mn coatings. The formation of Fe–Mn coatings indicate that Vertisols areas did experience a humid climate in the past and thus, the large amount of DSm formed in an earlier humid climate in the source area as an alteration product of plagioclase in tropical humid climates (Pal et al. 1989, 2012a;

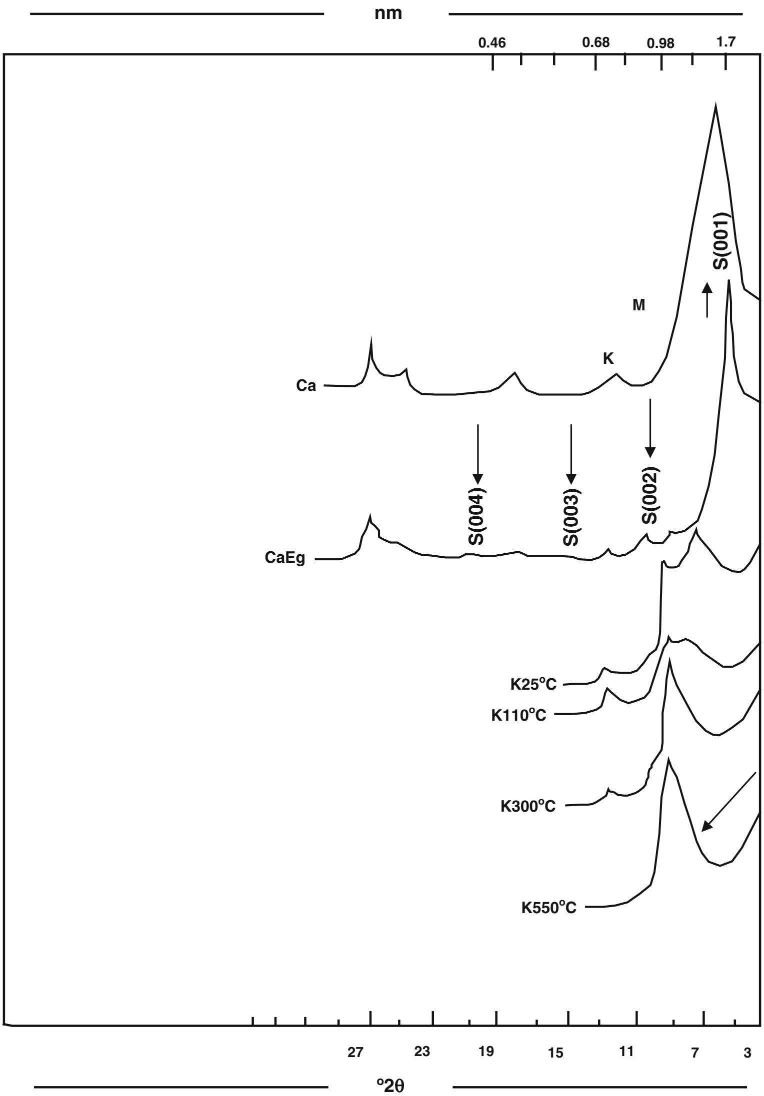

Fig. 7.2 Representative X-ray diffractograms of fairly well crystalline fine clay smectite of Vertisols despite having partial hydroxy-interlayering (Adapted from Pal 2003)

Srivastava et al. 1998), and during this weathering vermiculite transformed to HIV, which further transformed to PCh because hydroxy-interlayering in vermiculite would occur in acidic soil conditions. However, the formation of HIS in humid tropical climate did not continue as evidenced from the presence of very small amount of kaolin (KI-HIS) in the fine clay fractions. In the event of prolonged formation of HIS, the content of kaolin should have been dominant as reported by

Bhattacharyya et al. (1993) for the ferruginous Alfisols of the humid Western Ghats. The smectite of Vertisols thus formed in an earlier and more humid climate. Its crystallinity, and also the PCh were preserved in the non-leaching environment of the latter dry climates (Pal et al. 2009, 2012a, b). The ^{14}C age of soil organic carbon of Vertisols indicate soil age between 3390 to 10,187 year BP (Pal et al. 2006). This suggests that the climate did change from humid to drier climate in Peninsular India during the late Holocene (Pal et al. 2001, 2003, 2006; Deotare 2006). As a result, Vertisols in SAM, SAD and AD climates became more calcareous and sodic (Pal et al. 2006, 2009) than those of SHM and SHD climates. Therefore, they qualify to be polygenetic (Pal et al. 2001).

7.6 Implications for Climate Change from Clay Minerals Record in Drill Cores of the Ganga Plains

High resolution clay mineralogical study by Pal et al. (2012a) documents the climate change over the last 100 ka manifested in clay mineralogy of paleosols and sediments from two cores ($\sim$50 m deep) in the Ganga–Yamuna interfluve in the Himalayan Foreland Basin, India. Core sediments from the northern part of the interfluve (Indian Institute of Technology, Kanpur, IITK core) are micaceous and dominated by hydroxy-interlayered dioctahedral low-charge smectite (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 interfluves (Bhognipur core) are poor in mica and both LCS and HCS are present in the upper 28 m of the core and the lower part is dominantly LCS in all size fractions. In the two cores paleosols are formed in the sub-humid to semi-arid climatic conditions and the formation of paleosols has resulted clay minerals such as 1.0–1.4 nm minerals, vermiculite, HCS and also preserved the LCS, hydroxy-interlayered vermiculite (HIV) and pseudo-chlorite (PCh), and kaolin that formed earlier in a humid climate. The preservation of LCS, HIV, kaolin and PCh bears the signature of climate shift from humid to semi-arid conditions in the Ganga Plains as their formation does not represent contemporary pedogenesis in the present alkaline chemical environment induced by the semi-arid climate. The abundance of LCS sediments in both the cores suggests the role of plagioclase weathering in the formation of LCS. The climatic records inferred from the typical clay mineral assemblages of the two interfluve cores are consistent with the Marine Isotope Stages (MIS) over the last 100 ka. Typical clay mineral assemblage in humid interglacial stages (e.g. MIS 5, 3 and 1) (Fig. 7.3) 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 $CaCO_3$ were the dominant processes (Fig. 7.3). The formation of pedogenic $CaCO_3$ created conducive environment for illuviation of clays forming argillic (Bt) horizon in the paleosols of the interfluve (Srivastava et al. 2010).

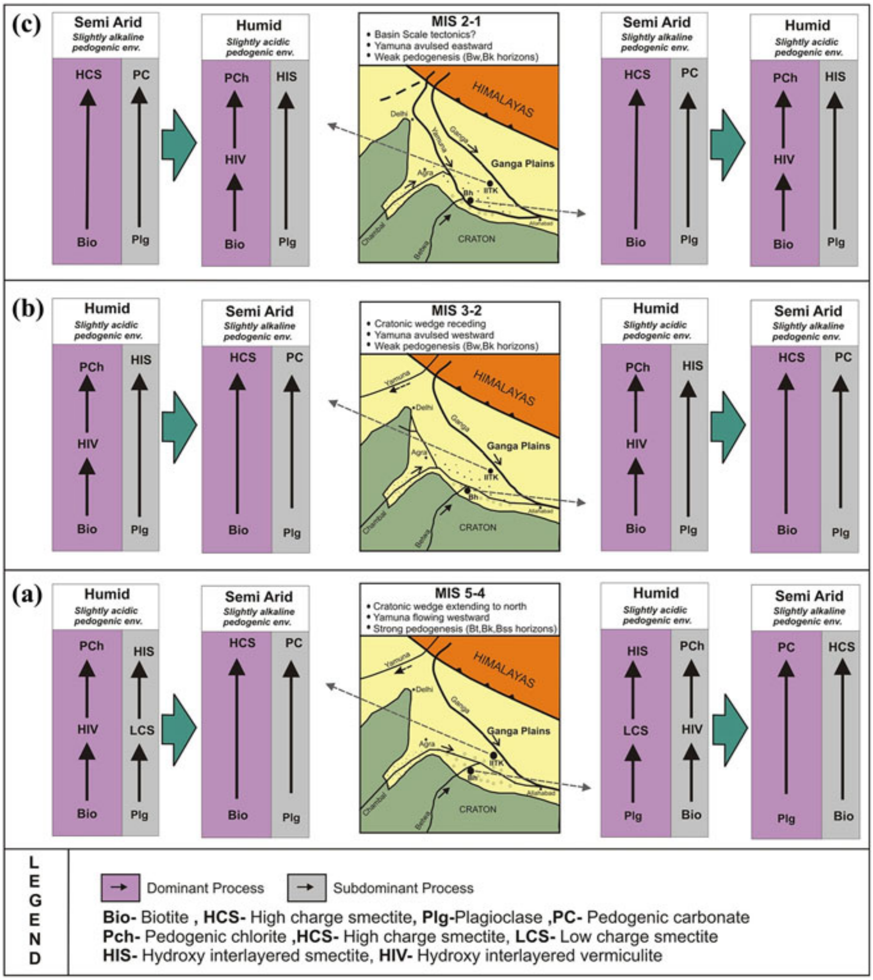

Fig. 7.3 A schematic climate-driven model for clay mineral alteration for the last 100 ka in northern and southern parts of the Ganga–Yamuna interfluve represented by IITK and Bhognipur cores (Adapted from Pal et al. 2012a)

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