Revista Mexicana de Ciencias Geológicas, v. 24, núm. 3, 2007, p. 510-523

Time dependence of the rate and direction of mineral weathering and clay mineral formation with special consideration to kaolinites

Arnt Bronger

*Geographisches Institut, Christian-Albrechts Universität zu Kiel,
Ludewig-Meyn-Str. 14, D-24098 Kiel, Germany.
bronger@uni-kiel.de*

ABSTRACT

This paper summarizes mineralogical results from temperate, subtropical and tropical soil chrono- and climosequences and aims to understand the role of the factor time for the formation of kaolinites. A mid-Pleistocene welded F6-paleosol at Stari Slankamen, Serbia, developed discontinuously in a time span of 140 ka, shows much greater pedochemical weathering and clay mineral formation than Holocene loess soils: more than 40% of the feldspars and almost 80% of the micas are decomposed; mainly smectites dominate the fine clay fraction followed by illites, but no kaolinites have formed. In the Atlantic coastal region of Morocco during the first 100 ka only weathering of calcarenites to Rendzinas (Typic Calcixerolls) has taken place with little formation of 2:1 clay minerals. In a time span of several 100 ka, however, the direction of weathering goes towards strong kaolinite formation. Six "red soils" (Typic Dystrudepts to a Typic Hapludalf) in hyperhermic SW Nepal with 1,500–1,750 mm annual rainfall (five humid months), and a "black soil" (Vertic Haplustoll) near Borada, Gujarat, India with 930 mm annual rainfall were studied. Two of the "Red Soils" have thermoluminescence (TL) ages between 10 and 30 ka, the "Black Soil" has an age of about 10 ka. Surprisingly, little pedogenic clay mineral formation could be identified. The illites and kaolinites are mostly of detrital (inherited) origin. Only in the Vertic Haplustoll, a small increase mainly of smectites but no kaolinites were found, although the content of weatherable minerals is large. In South India, nine soils in a climatic sequence derived from saprolite of weathered granitic gneiss were examined for recent and relict features. Above a threshold of about 2,000 mm in an Udic Rhodustalf deep weathering is a recent process leading to the formation of kaolinites; above 2,500 mm in a Typic Rhodudult it leads also to gibbsite. In Aridic Rhodustalfs (three to one humid months), besides the formation of 2:1 clay minerals, still strong formation of kaolinites has taken place. However, this process has now almost ceased; instead, secondary carbonate is accumulating in the lower part of the profile proving that these Alfisols are relict soils, formed in an earlier period of much moister climate. However, according to our recent results from Morocco and Nepal besides a climatic change, the soil forming factor time is very important: strong pedogenic formation of kaolinites also in the seasonal tropics needs a longer time, probably some 100 ka.

Key words: time, weathering efficiency, kaolinite formation, Morocco, Nepal, South India.

RESUMEN

En este estudio se resumen los resultados mineralógicos obtenidos a partir de crono y climosecuencias de suelos en zonas templadas, subtropicales y tropicales con el objetivo de dilucidar el papel del factor tiempo en la formación de caolinitas. En un paleosuelo F6 consolidado de mediados del Pleistoceno en Stari Slankamen, Serbia, que se formó en un periodo discontinuo de 140 ka, se observa un intemperismo pedogénico mucho más intenso y una neoformación de arcillas minerales mucho mayor que la registrada en suelos formados a partir de loess durante el Holoceno: más del 40% de los feldespatos y casi el 80% de las micas se descompusieron, y se formaron sobre todo esmectitas en la fracción de arcilla fina, que es la dominante, seguidas por illitas, pero no se neoformaron caolinitas. En la costa atlántica de Marruecos durante los primeros 100 ka sólo hay evidencias del intemperismo de calarenitas a partir de las cuales se formaron suelos de tipo Rendzinas (Calcixerolls típicos) con pequeñas cantidades de minerales de arcilla del tipo 2:1. Sin embrago, en periodos de varios cientos de ka, el intemperismo avanza hacia una fuerte neoformación de caolinitas. También se muestran datos de seis "suelos rojos" (Dystrudepts típicos a Hapludalfs típicos) en zonas hypertérmicas del SW de Nepal con una precipitación anual de 1,500 a 1,750 mm y cinco meses húmedos al año, y de un "suelo negro" (Haplustoll vértico) cerca de Baroda, Gujarat, La India con una precipitación anual de 930 mm. Dos de los "suelos rojos" tienen edades determinadas por termoluminiscencia entre 10 y 30 ka, mientras que el "suelo negro" tiene una edad de aproximadamente 10 ka. Sorpresivamente se identificó sólo una baja neoformación de minerales de arcilla. Las illitas y caolinitas presentes son dominantemente heredadas del material parental. Sólo el Haplustoll vértico muestra un ligero incremento dominantemente de esmectitas, pero no de caolinitas, a pesar de que el contenido de minerales intemperizables es alto. Al sur de La India se examinaron nueve suelos en una secuencia climática de suelos derivados a partir de saprolitas de gneiss granítico en búsqueda de rasgos relícticos y recientes. Más allá de un umbral de 2,000 mm de precipitación media anual en un Rhodustalf údico, el intemperismo profundo es un proceso reciente que conlleva a la formación de caolinita; y a más de 2,500 mm de precipitación en un Rhodudult típico incluso se forma gibsita. En Rhodustalfs arídicos con uno a tres meses de humedad aún se forman abundantes caolinitas, además de minerales 2:1. Sin embargo, este proceso en la actualidad se encuentra prácticamente detenido y en su lugar se observa una abundante acumulación de carbonatos de calcio secundarios en la parte inferior del perfil. Esto demuestra que los Alfisoles son suelos relícticos, que se formaron en un periodo anterior bajo climas mucho más húmedos. Sin embrago, de acuerdo con nuestros resultados más recientes obtenidos en Marruecos y Nepal, concluimos que el factor tiempo también es muy importante, además de un cambio climático: una formación abundante de caolinita necesita probablemente más de 100 ka también bajo climas estacionales del trópico.

Palabras clave: tiempo, eficiencia del intemperismo, formación de caolinita, Marruecos, Nepal, sur de la India.

INTRODUCTION

The efficiency of weathering under tropical climates has often been over-estimated, especially in the geomorphological literature. For instance, Thomas (1978, p. 33) estimated "30–50 m of weathering in crystalline rocks within $10^5 – 10^6$ years, perhaps even 10^7 years". Bremer (1981) assumed that the process of "deep and uniform weathering" in a tropical climate with more than 1,600 mm annual rainfall leads to weathering of rocks of different resistance at rates of 100 m in 2–3 Ma to 100 m in 3–6 Ma (Bremer 1986, p. 104). Later Bremer (1989, p. 371) postulated that the formation of a tropical "Rotlehm" (including Oxisols) is at least as quick as that of soils outside the tropics, assuming that a fully developed soil in the temperate climatic belt is formed in 2,000–5,000 years. According to Büdel's theory of the "mechanism of double planation surfaces" (Büdel 1965, 1982; see also Thomas 1994, p. 288), a "basal weathering surface" is separated from the "wash surface" by a 4–10 m thick "monogenetic Rotlehm" especially in the Madras-Bangalore area, South India. According to Büdel, this soil was formed mainly during the Holocene and later Pleistocene. The recent deep weathering of Peninsular Gneiss at the "basal weathering surface" is regarded by Büdel (1965) to occur faster than erosion of the surface soil even under semiarid conditions.

This paper summarizes the findings of former studies (Bruhn, 1990; Bronger and Bruhn, 1989; Bronger *et al.*, 1998a, 1998b; Bronger and Sedov, 2003) on particle size and mineralogy of surface soil climo- and chronosequences from subtropical and tropical regions. In several cases it was possible to provide the instrumental age control through the thermoluminescence (TL) dating of the sedimentary parent material. These results are compared with Holocene soils and a Pleistocene welded paleosol from loess formed discontinuously in several interglacials (Bronger and Heinkele, a)

b)

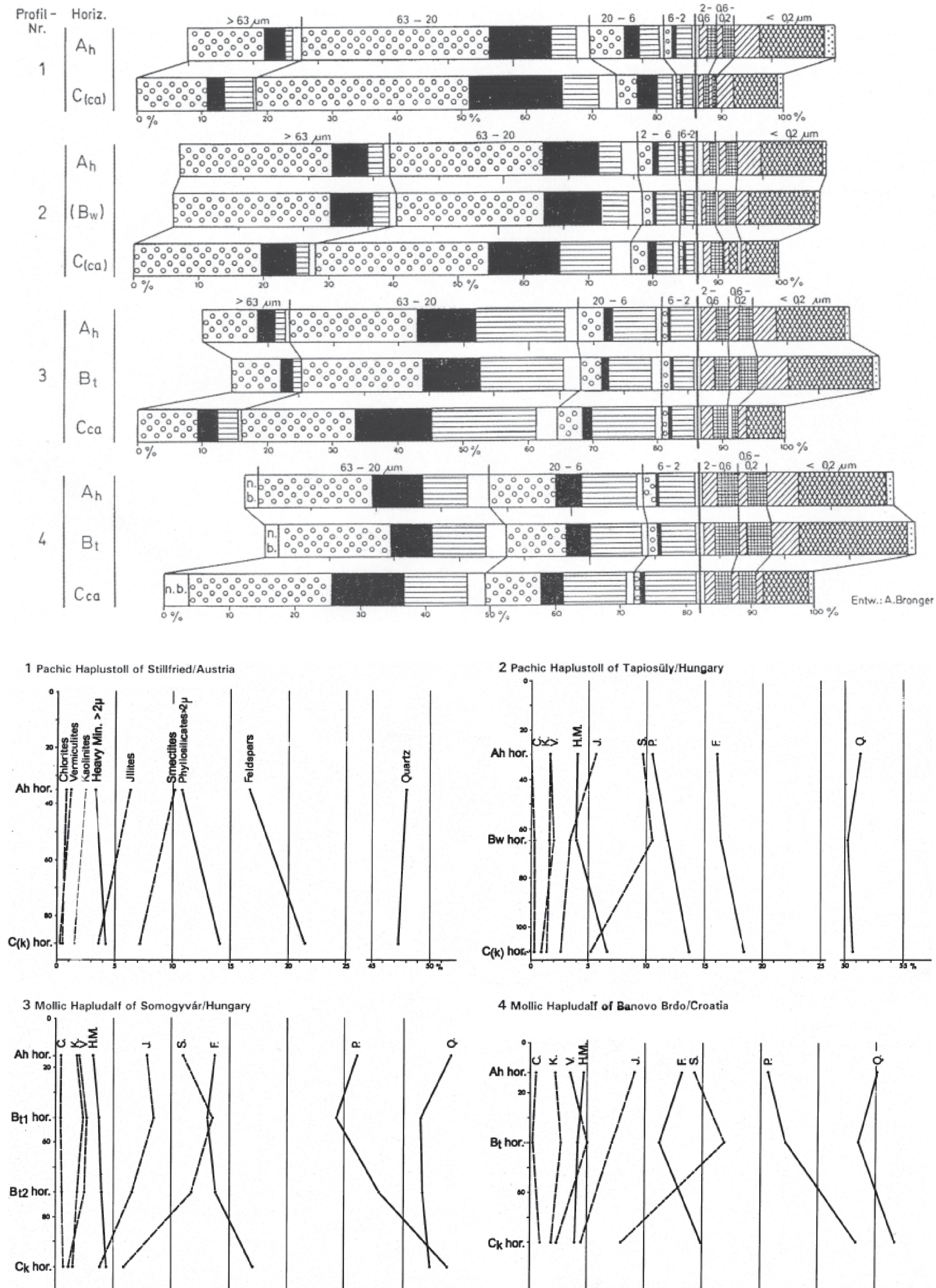

Figure 1. Particle size, mineralogical and clay mineralogical composition of four Holocene loess soils in the Carpathian basin. a) Mineralogical composition of each fraction (legend see Figure 9). b) Cumulative curves of the primary minerals >2 µm and the clay minerals.

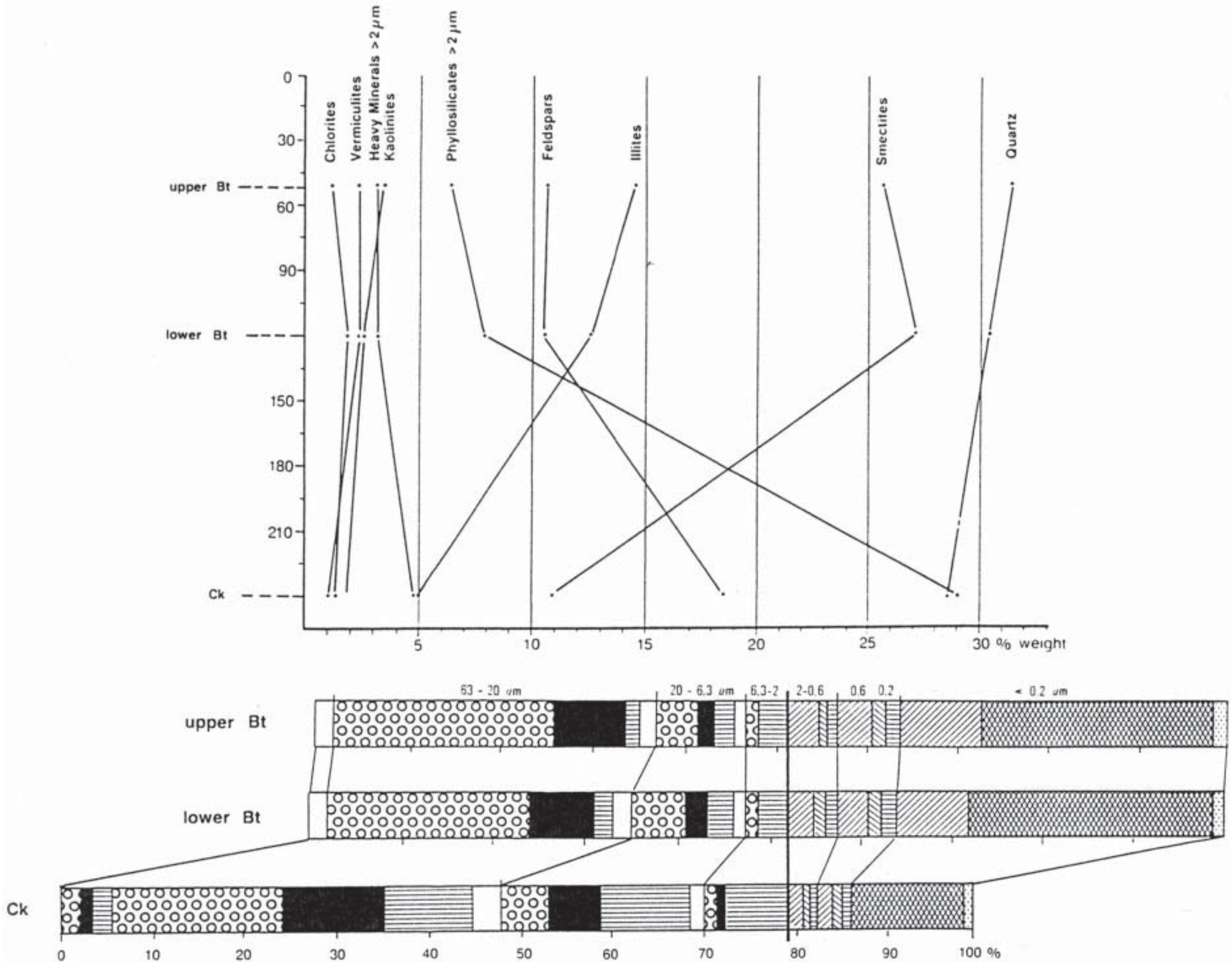

Figure 2. Particle size, mineralogical and clay mineralogical composition of the strongly rubefied mid-Pleistocene F6 complex in the loess profile of Stari Slankamen, Serbia.

1989; Bronger *et al.*, 1998), where the time frame of its formation is well known. These results were re-interpreted with focus on the comparison of time aspects (trends, rates and efficiency) of weathering and mineral neoformation under different climates. The main aim is to get an idea of the minimum age of kaolinite formation under semiarid conditions.

METHODS

The main methodical principle of the work is to study the mineralogical composition of all particle size fractions in all profiles in a uniform way, and to evaluate (semi) quantitatively both the loss of primary minerals due to weathering and the accumulation of neoformed clay minerals in the fraction 2–0.2 µm and <0.2 µm.

For particle size and mineralogical analyses, the following methods were employed:

1) Particle size analysis (sieving and pipette method) after H_20_2 and HCl (pH 4) pretreatment and dispersion with $Na_4P_2O_7$.

2) For the analyses of minerals >2 µm, the three sand and silt fractions (in some soils only 63–2 µm because the samples were too small) were separated quantitatively. The fractions >20 µm were analysed on slides by identifying ≥ 300 particles per slide with a polarizing microscope. The proportions of quartz, feldspar, phyllosilicates, and heavy minerals in the fractions 20–6 and 6–2 µm were determined by identifying 700–1,000 particles per slide by phase contrast microscopy. The percentages of the minerals or mineral groups in each fraction were then multiplied by the weight percentages of each fraction to obtain the weight percentages of each mineral, as shown in Figures 1-5, and 6-11 (left side).

3) For the analysis of silicate clay minerals, the fractions of coarse and medium clay (2–0.2 µm) were also separated quantitatively after removal of carbonates and

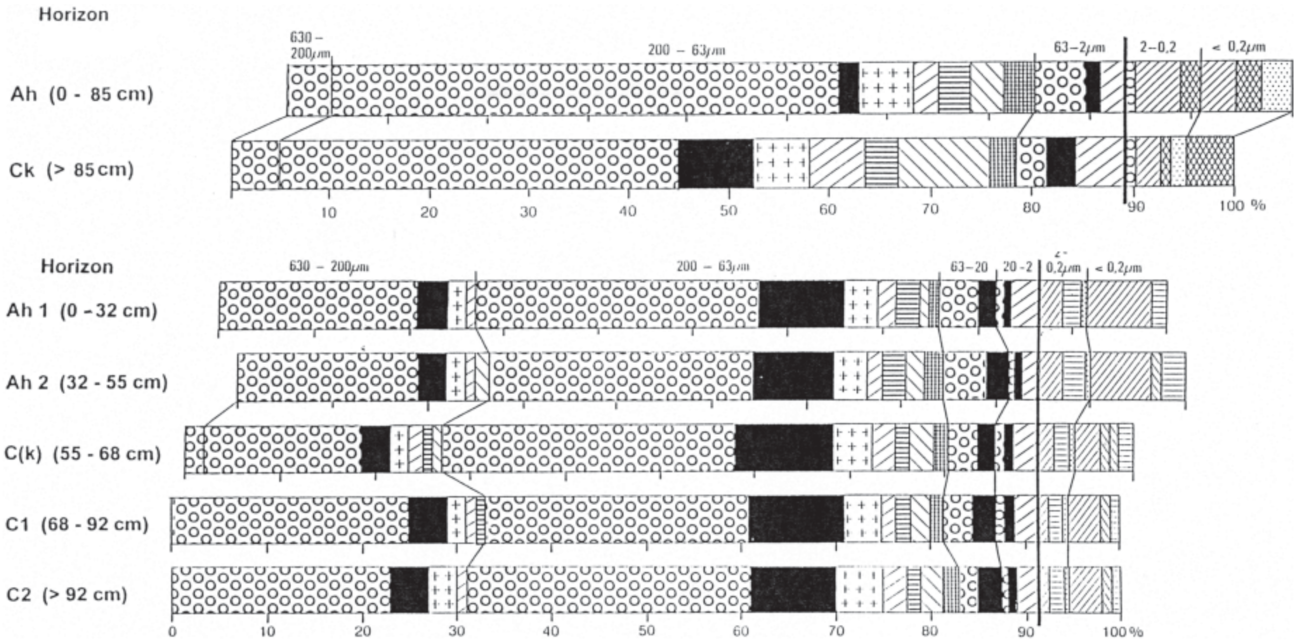

Figure 3. Particle size, mineralogical and clay mineralogical composition of a Typic Calcixeroll in Temara (C horizon >100 ka) near Rabat and a Typic Calcixeroll (C horizon 117 ± 24 ka) near Mohammedia, Morocco. Legend see Figure 5.

free iron oxides. Samples were saturated with Mg, ethylene glycol and K^+, and heated at 25 °C, 125 °C, 400 °C, and 560 °C. The composition of the clay subfractions was estimated semiquantitatively on the basis of the areas under selected X-ray diffraction (XRD) peaks, using the weighting factors recommended by Laves and Jähn (1972). Illites, vermiculites and quartz (4.26 Å) were given a weighting factor of 1, mixed layer minerals and gibbsites (4.81Å) of 0.5, and kaolinites and smectites of 0.25. The results were compared with the CEC of each fraction to correct gross deviations. The weight percentages of the minerals, calculated by multiplying the estimated percentages of clay minerals in each fractions by the weight of each fraction, shown in Figures 1-11, right side, are consequently only estimates, in contrast to the weight percentages of the fractions >2 μm. Figures 1-11 show also

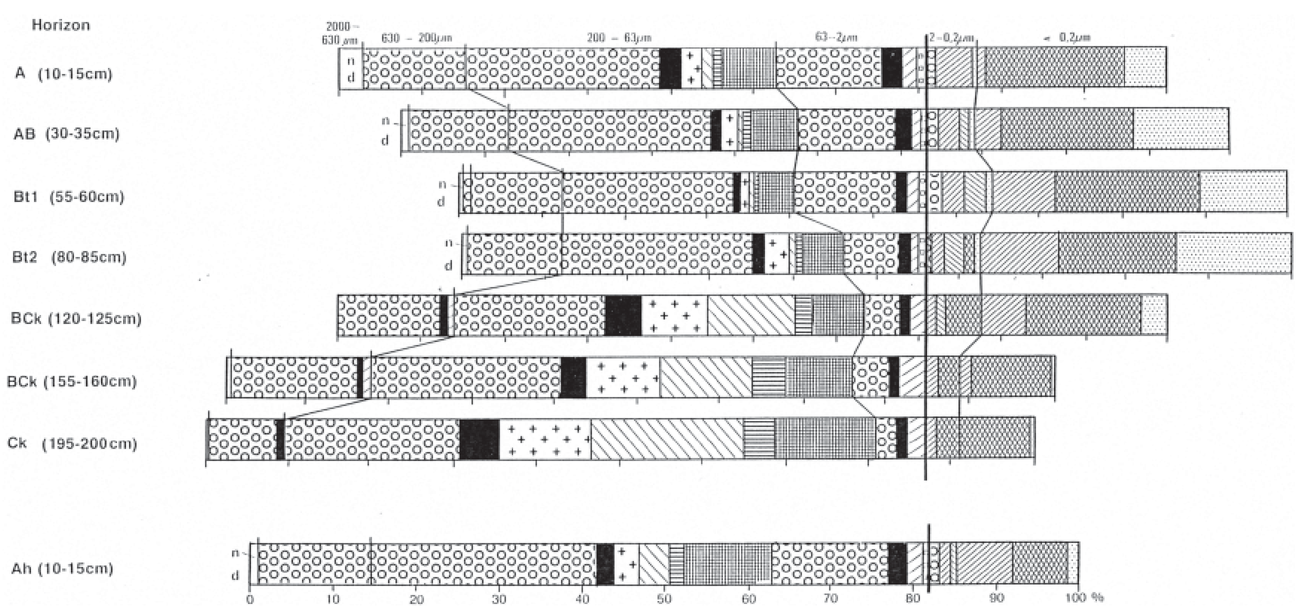

Figure 4. Particle size, mineralogical and clay mineralogical composition of a Typic Rhodoxeralf and a Typic Calcixeroll (Ah horizon) of Oued Ykem near the coast in the vicinity of Rabat. Legend see Figure 5.

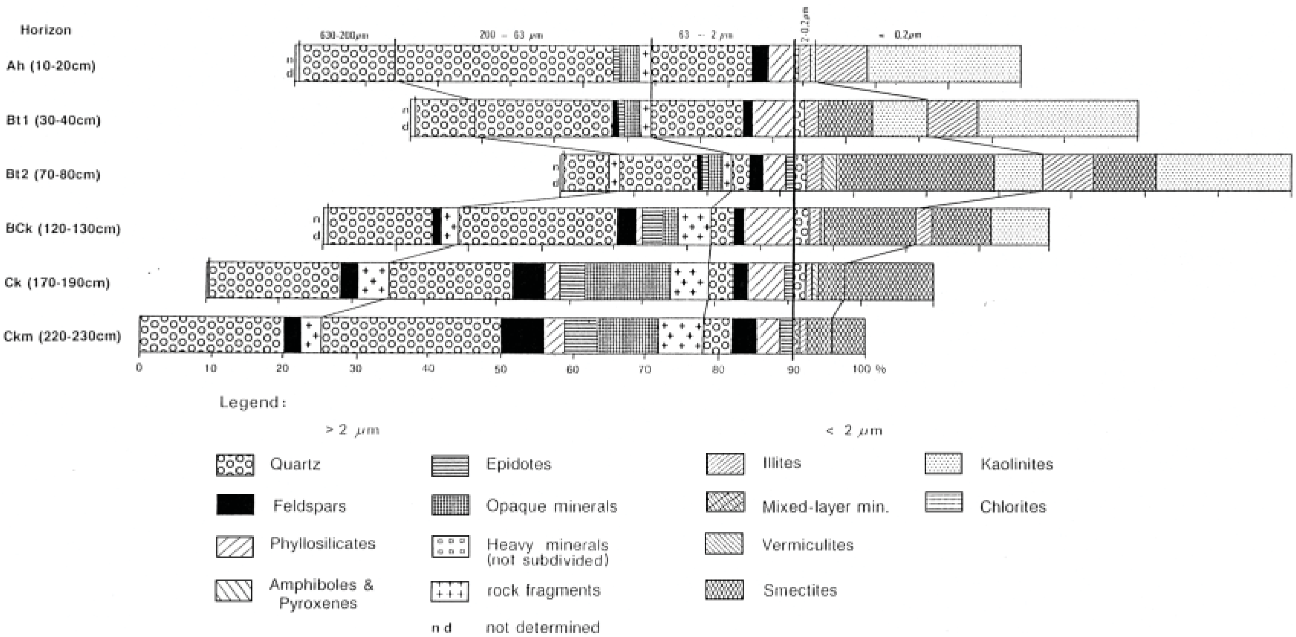

Figure 5. Particle size, mineralogical and clay mineralogical composition of a Typic Rhodoxeralf of Tal'at Ach Chwar near Rabat, 7 km inland 130 m a.s.l. Below the soil, the Brunhes/Matuyama boundary was found.

the particle size distribution of the soil horizons including their C, Ck, Ckm or Cr horizons.

4) The micromorphology of all soils and their parent material was studied in thin sections.

RESULTS AND INTERPRETATION

In the selected Holocene soils (Figure 1b) and the welded F6 soil Stari Slankamen (Figure 2), developed in OIS (Oxygen Isotope Stage) 15 to 13 (Figure 12; see section 3.2), the percentage of quartz by weight in the different soil horizons is approximately constant. Using quartz as an index mineral (Barshad, 1967), there is good evidence that the soil parent material was originally homogenous. This is also the case in the Typic Dystrudept from SW Nepal (Figure 6). The data shown in Figures 3-5 and 7-11, on the other side, do not indicate mineral weathering balances; instead, they should be interpreted as pedogenic mineral weathering tendencies.

The results are summarized in Figures 1-11. The main points worth emphasizing are the following:

1) In Holocene loess soils, *e.g.*, in southeast central Europe, the rate of pedogenic clay formation is up to 6% in Chernozems (Pachic Haplustolls) and up to 15% in Haplic to Luvic Phaeozoms (ISSS – ISRIC – FAO, 1998) or Mollic Hapludalfs (Soil Survey Staff, 2003, see Figures 1a and 1b). In this area, smectites in the fine clay fraction <0.2 µm and illites in the coarse clay (2–0.2 µm) are the dominant pedogenetically formed clay minerals. However, in Tadjikistan and the Chinese Loess Plateau, the dominant pedogenic clay minerals are illites and vermiculites even in the fine clay fraction (Bronger *et al.*, 1998b, Bronger and Heinkele, 1989). Main sources of pedogenic clay are biotites and feldspars in the silt fractions.

2) For the Brunhes epoch, the loess-paleosol sequence at Karamaydan/Tadjikistan seems to be very complete and allows a very good correlation with the deep-sea oxygen isotope record, which includes the development of an accurate astronomical time scale (Bassinot *et al.*, 1994). Therefore, the approximate duration of the development of a paleosol, which corresponds with the length of an interglacial, can be estimated at about 10–15 ka, the duration of the development of the frequent pedocomplexes (two or more pedomembers as parts of different soils), which correspond each with the length of an interglacial complex, is about 50 ka (Bronger *et al.*, 1998a). The rate and direction of pedogenic clay mineral formation is very similar to those of Holocene soils in the respective area (Bronger *et al.*, 1998b). The mid-Pleistocene welded F6 soil at Stari Slankamen, Serbia, shows much more pedogenic weathering: more than 40% of the feldspars and almost 80% of the micas are decomposed; in thin sections, only muscovites are found and no biotites remain. The rate of pedogenic clay formation is nearly 30%, mainly smectites in the dominating fine clay fraction followed by illites, but no kaolinites were formed (Figure 2). The F6 soil is correlated chronostratigraphically with pedocomplexes VI and V at Karamaydan, consisting of 5–6 paleosols that were formed over a period of 140 ka, although pedogenesis was interrupted several times by

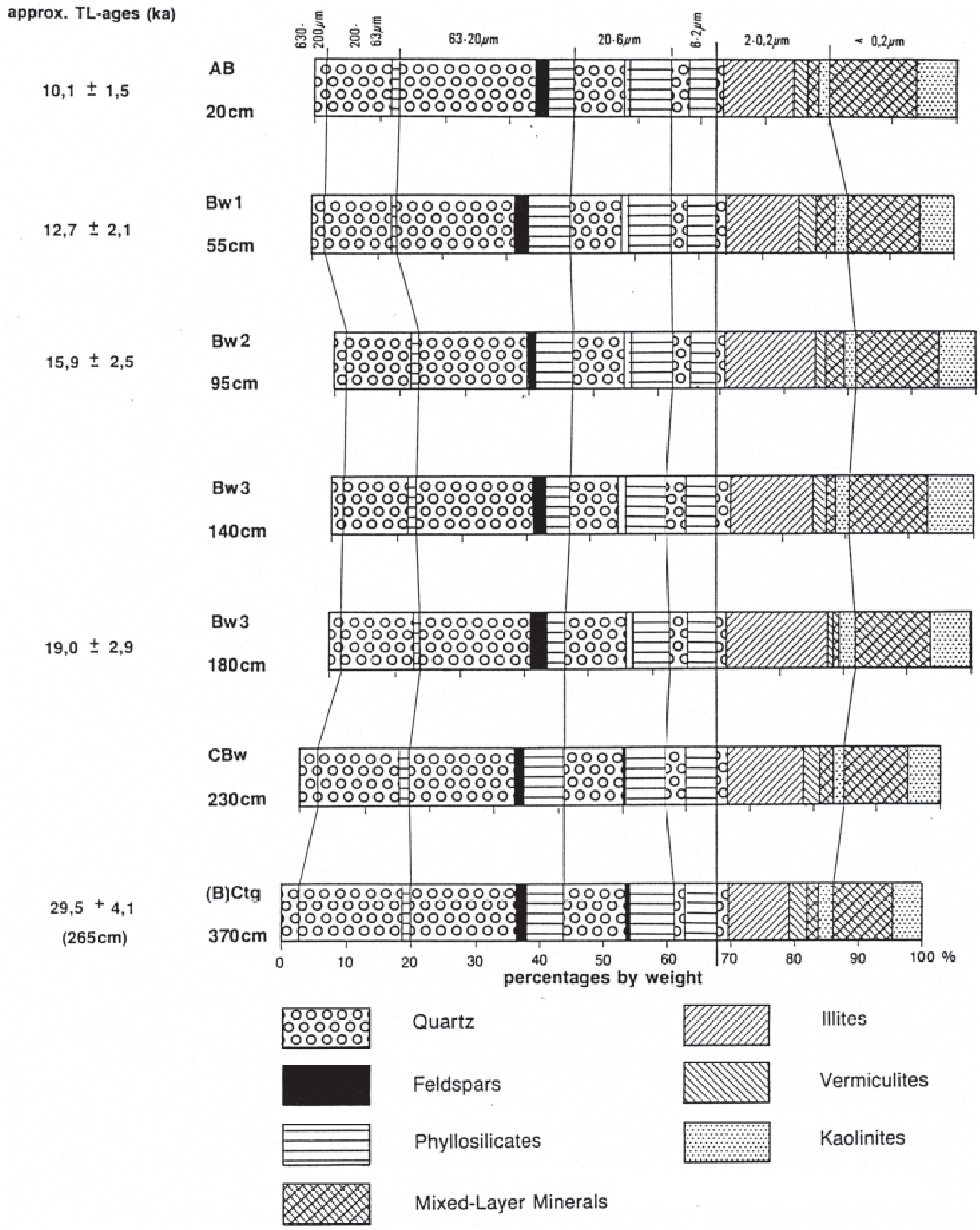

Figure 6. Particle size, mineralogical and clay mineralogical composition of a Typic Dystrudept (Lalmatya soil, Deokhuri Valley, SW-Nepal). Thermoluminescence dates from the adjacent Argun Khola soil: Zöller (2000).

loess formation (Figure 12). Therefore, the much greater pedochemical weathering and clay mineral formation of the welded F6 soil in comparison with the Holocene soils results from a much longer period of pedogenesis and does not indicate a warmer or wetter climate.

3) In the Atlantic coastal region of Morocco, with a thermic soil temperature and a xeric soil moisture since the Pliocene, the Moroccan Meseta has gradually emerged from the sea. Marine sediments are covered by polycyclic coastal dune complexes, transformed to calcarenites, which are younger near the coast and older farther inland. Below two of the selected Terrae Rossae (Typic Rhodoxeralfs), all on level surfaces, the Brunhes/Matuyama boundary was found (Bronger and Sedov, 2003). In the first about 100 ka, only weathering of calcarenites to Rendzinas (Typic Calcixerolls) has taken place with small formation of 2:1 clay minerals. In one example, kaolinite formation may just have started (Figure 3). In a time span of several

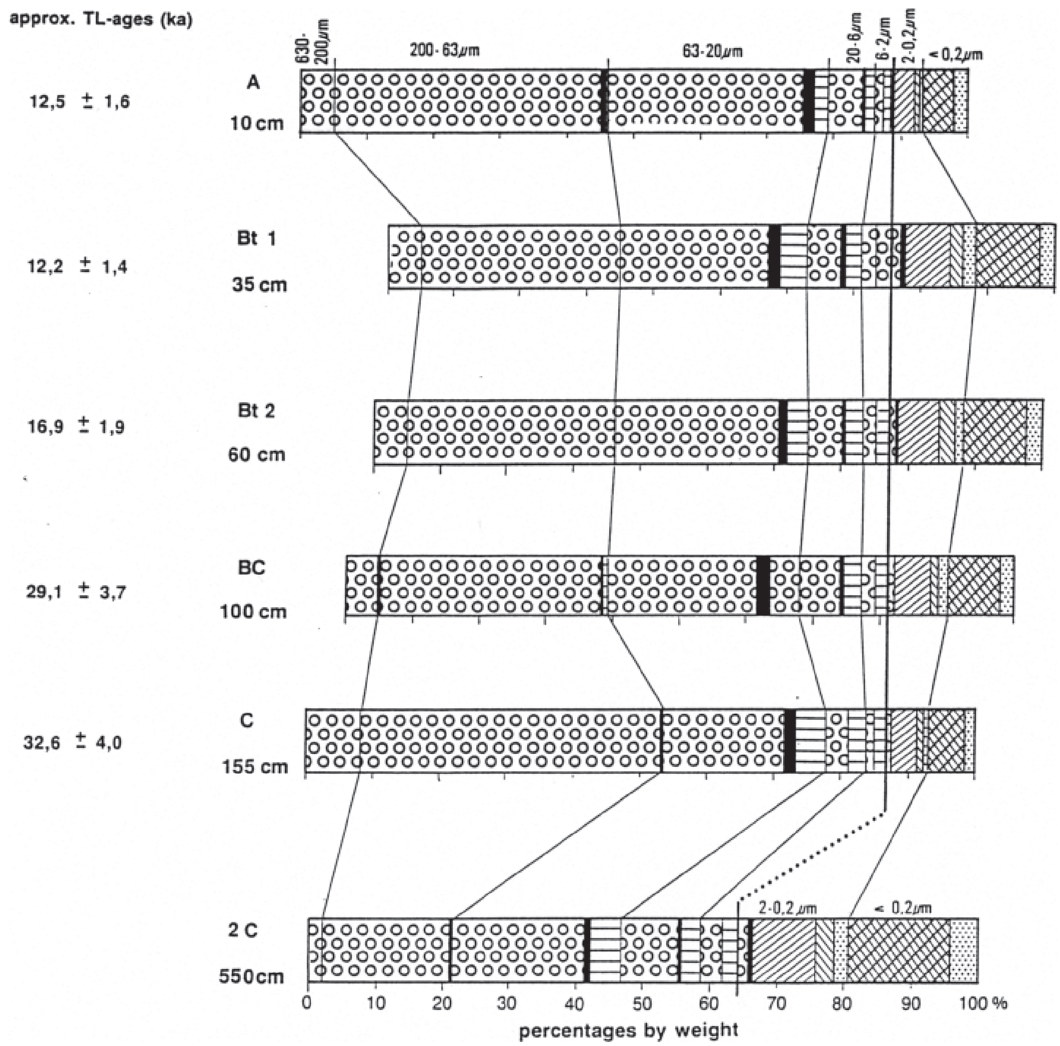

Figure 7. Particle size, mineralogical and clay mineralogical composition of a Typic Hapludalf (Gidhniya soil, Tui (Dang) Valley, SW-Nepal). Thermoluminescence dates: Zöller (2000).

100 ka, however, the direction of weathering goes towards strong kaolinite formation. For the most part, the pedogenic kaolinites show poor crystallinity of the fireclay type. They are mainly formed from feldspars, in same Terrae Rossae (Rhodoxeralfs) also from phyllosilicates, amphiboles and pyroxenes in the fraction >2 µm, and from smectites, inherited from the calcarenite-residual loam in the coarse (2–0.2 µm) and fine clay fraction (<0.2 µm; Figures 4 and 5).

4) The efficiency of weathering in the seasonal semi-arid to subhumid tropics has often been greatly overestimated. Six selected “red soils” (Typic Dystrudepts and one Typic Hapludalf) in two intramontane basins of hyperthermic SW Nepal, near the border with India, with 1,500–1,750 mm annual rainfall (five humid months) and a “black soil” (vertic Haplustoll) near Baroda, Gujarat, India with 930 mm annual rainfall (three to five humid months; Figure 13) were studied mineralogically. Two of the “red soils” have TL ages between 10 and 30 ka (Zőller, 2000), the “black soil” has one of about 10 ka, the yellowish silty parent material of the “red soils” is a preweathered soil sediment; it contains only small amounts of more easily weatherable primary minerals: around 5% feldspars and 10–15% phyllosilicates, dominantly relative stable muscovites (Figures 6 and 7). Surprisingly, little pedogenic clay mineral formation could be identified. The illites and kaolinites are mostly of detrital (inherited) origin. The few non-regular mixed layer minerals in the fine clay fraction can be interpreted as resulting from the initial stage of silicate weathering. In the dated Vertic Haplustoll, only a small increase mainly of smectites but no kaolinites could be found, although the content of weatherable minerals is high (Figure 8).

5) Nine soils from South India, when possible Benchmark soils (Murthy *et al.*, 1982), in a climatic sequence from ten to one humid months per year and derived from saprolite of weathered granitic gneiss were examined

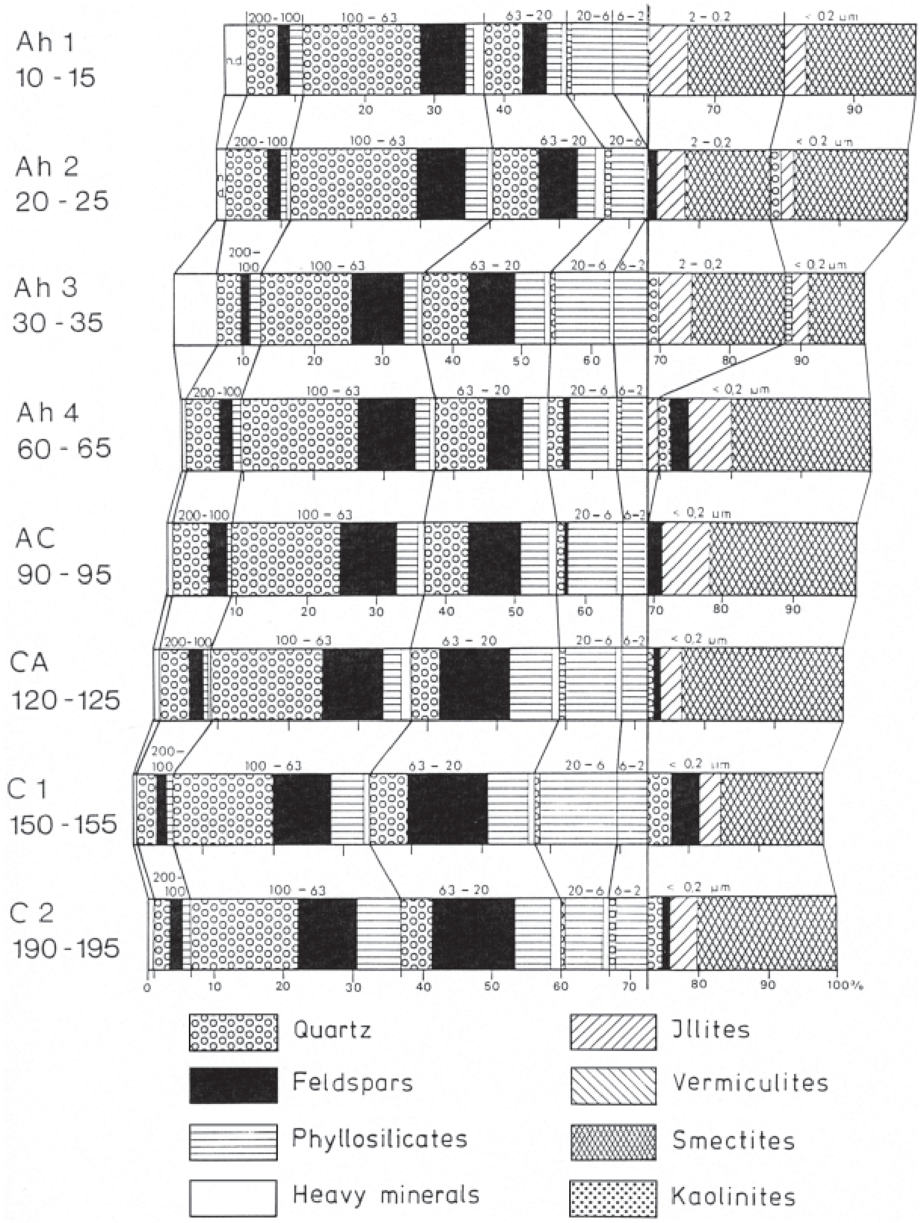

Figure 8. Particle size, mineralogical and clay mineralogical composition of a Vertic Haplustoll (Purohit soil near Baroda, Gujarat, India).

for recent and relict features (Bronger and Bruhn, 1989; Bruhn 1990). Above a threshold of about 2,000 mm (six humid months) in an Udic Rhodustalf, deep weathering is a recent process leading to the formation of kaolinites (Figure 10); above 2,500 mm (ten humid months) on the windward side of the Western Ghats at 900 m a.s.l. in a Typic Rhodudult, it leads also to the formation of gibbsite (Figure 9). In Aridic Rhodustalfs (three to one humid months), besides the formation of 2:1 clay minerals, still strong formation of kaolinites has taken place (Figure 11). However, this process has now almost ceased because of the desiccation of peninsular India east of the Western Ghats; instead secondary carbonate is accumulating in the saprolite (Cr) and lower part of the Bt horizons surrounding weathered biotite, hornblende (Figure 14) and plagioclase.

CONCLUSIONS AND DISCUSSION

Earlier we concluded that Alfisols (close to Lixisols) with kaolinites as the dominant pedogenic clay mineral in now semiarid India are relict soils or un-buried paleosols, formed in an earlier period of much moister climate (Bronger and Bruhn, 1989; Bruhn, 1990). However, according to our recent results from Morocco and Nepal, besides a climatic change, the soil forming factor time is very impor-

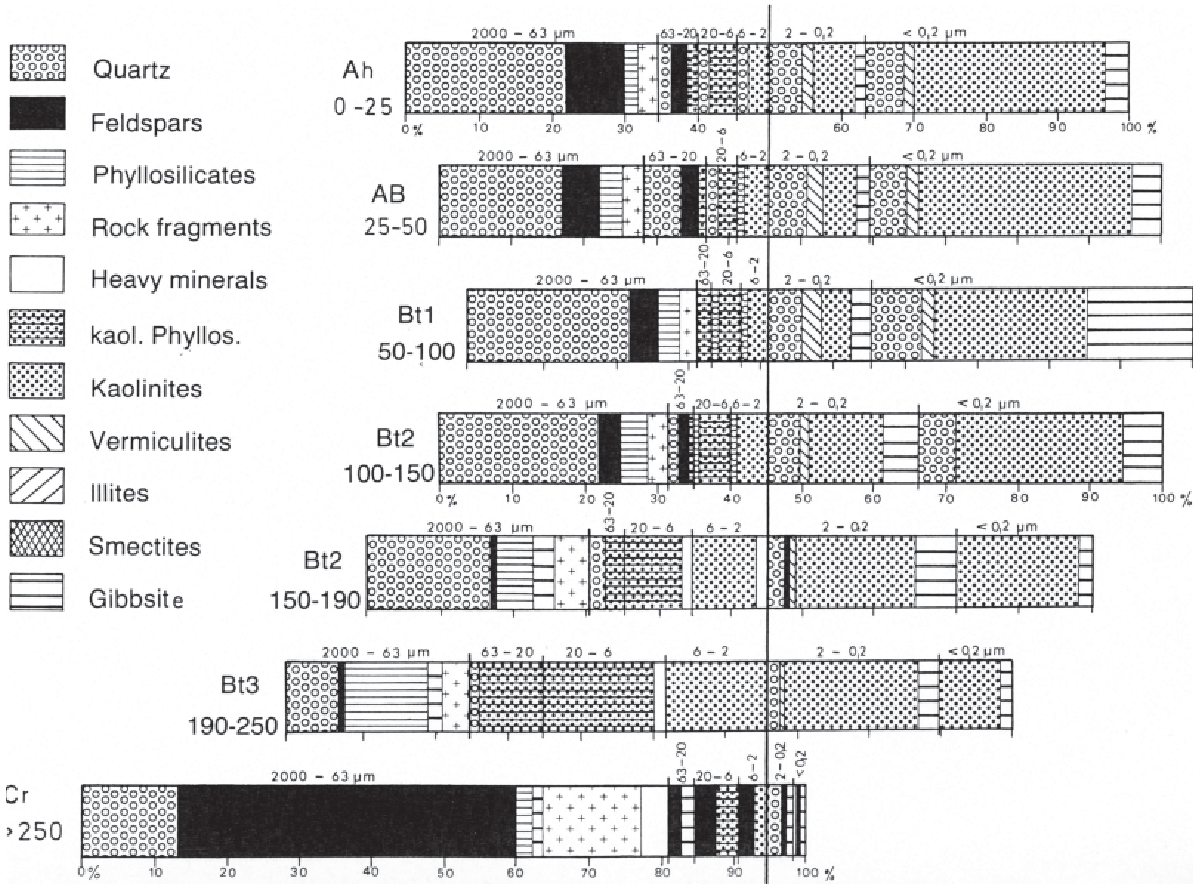

Figure 9. Particle size, mineralogical and clay mineralogical composition of a Typic Rhodudult (Vandiperiyar soil).

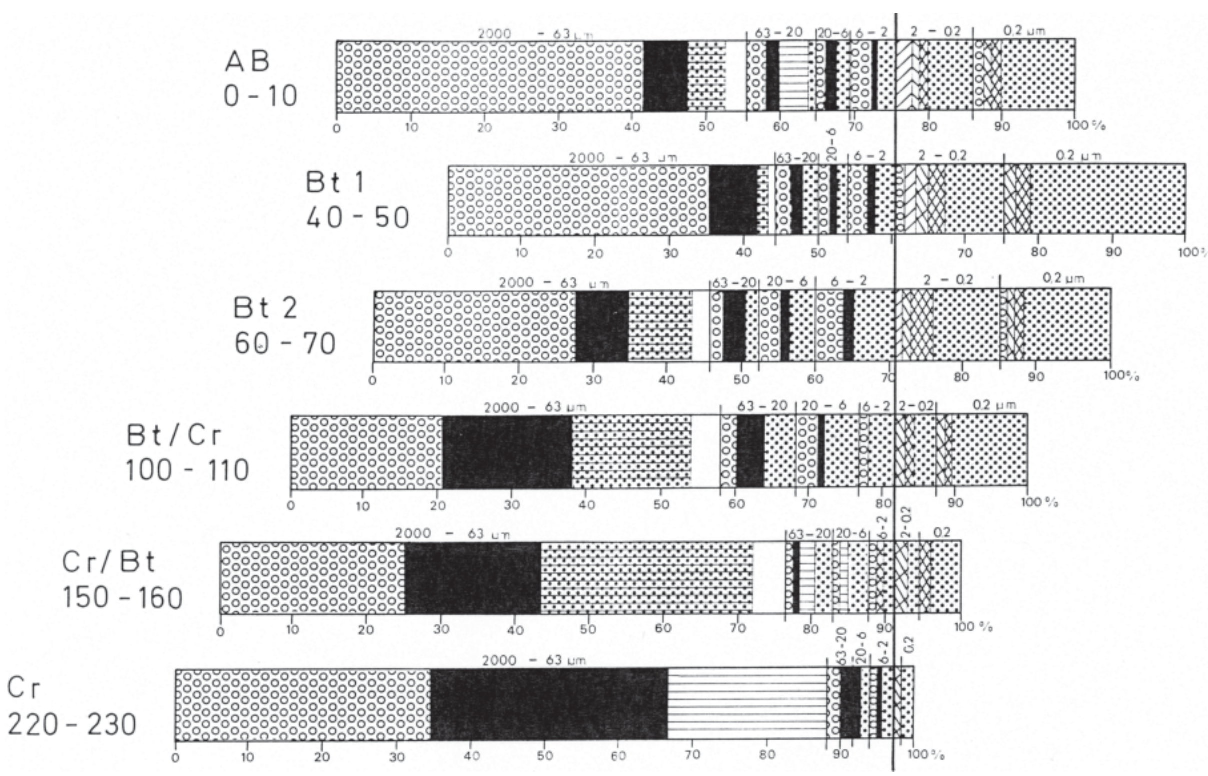

Figure 10. Particle size, mineralogical and clay mineralogical composition of a Udic Rhodustalf (Palghat soil).

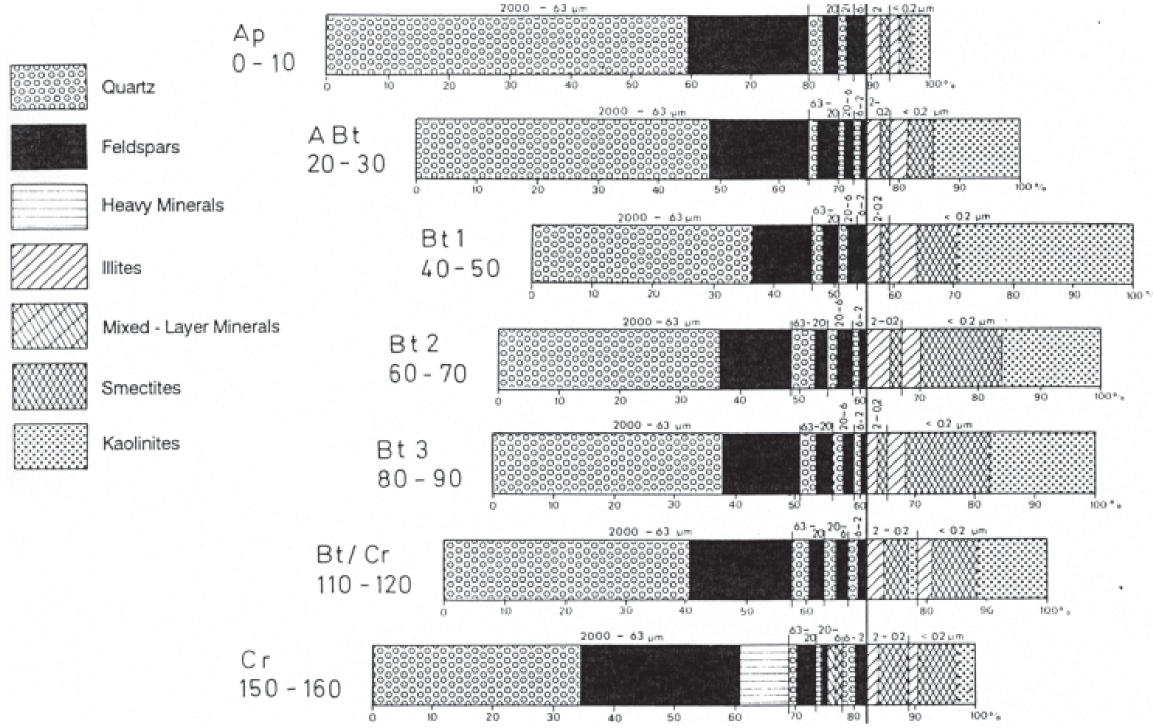

Figure 11. Particle size, mineralogical and clay mineralogical composition of an Aridic Rhodustalf (Patancheru II soil).

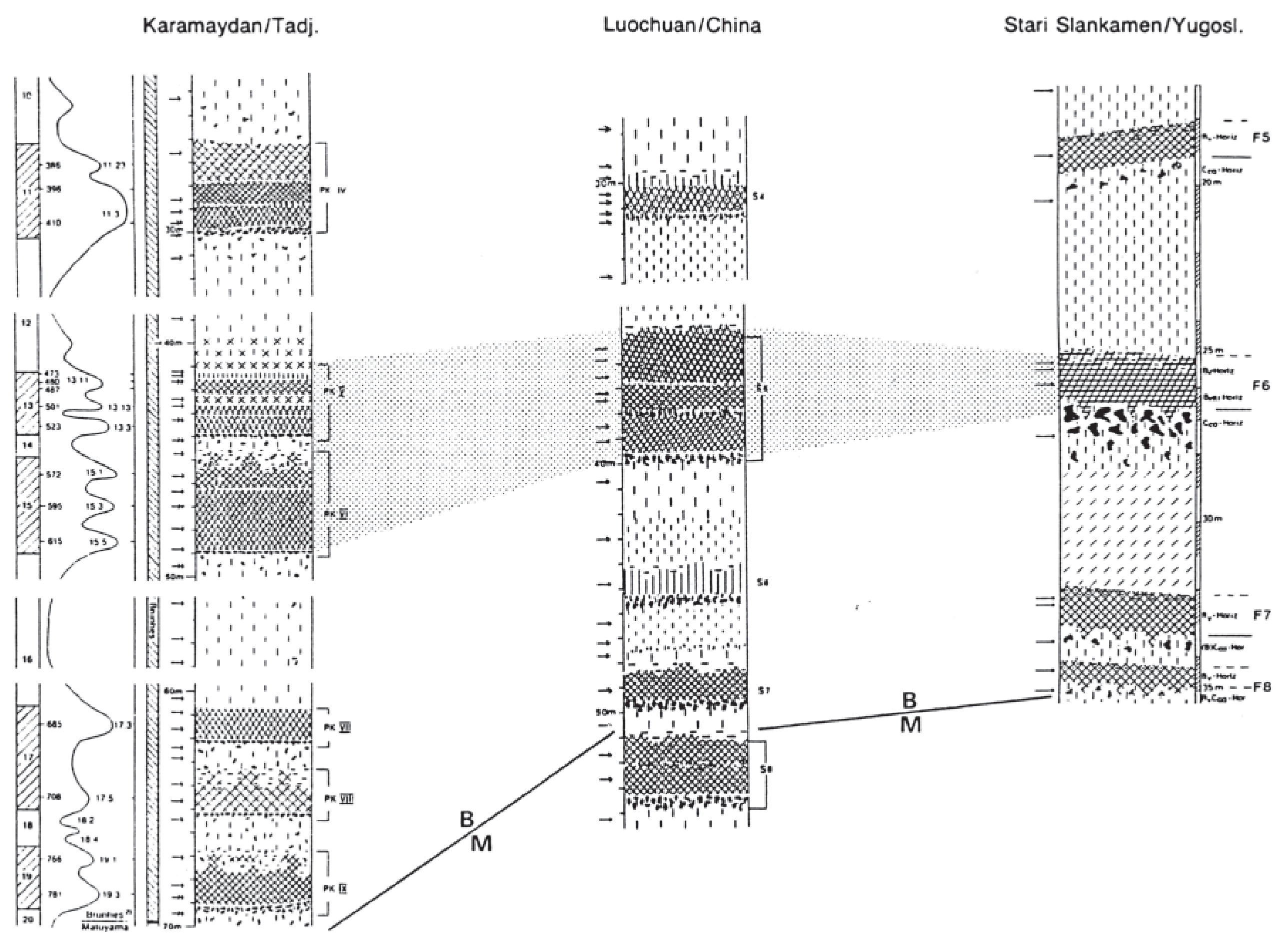

Figure 12. Pedostratigraphic correlation of loess-paleosol sequences of Karamaydan, Central Asia, Louchuan, East Asia and Stari Slankamen, Serbia with stages 15 to 13 of the deep-sea oxygen record of Bassinot et al. (1994).

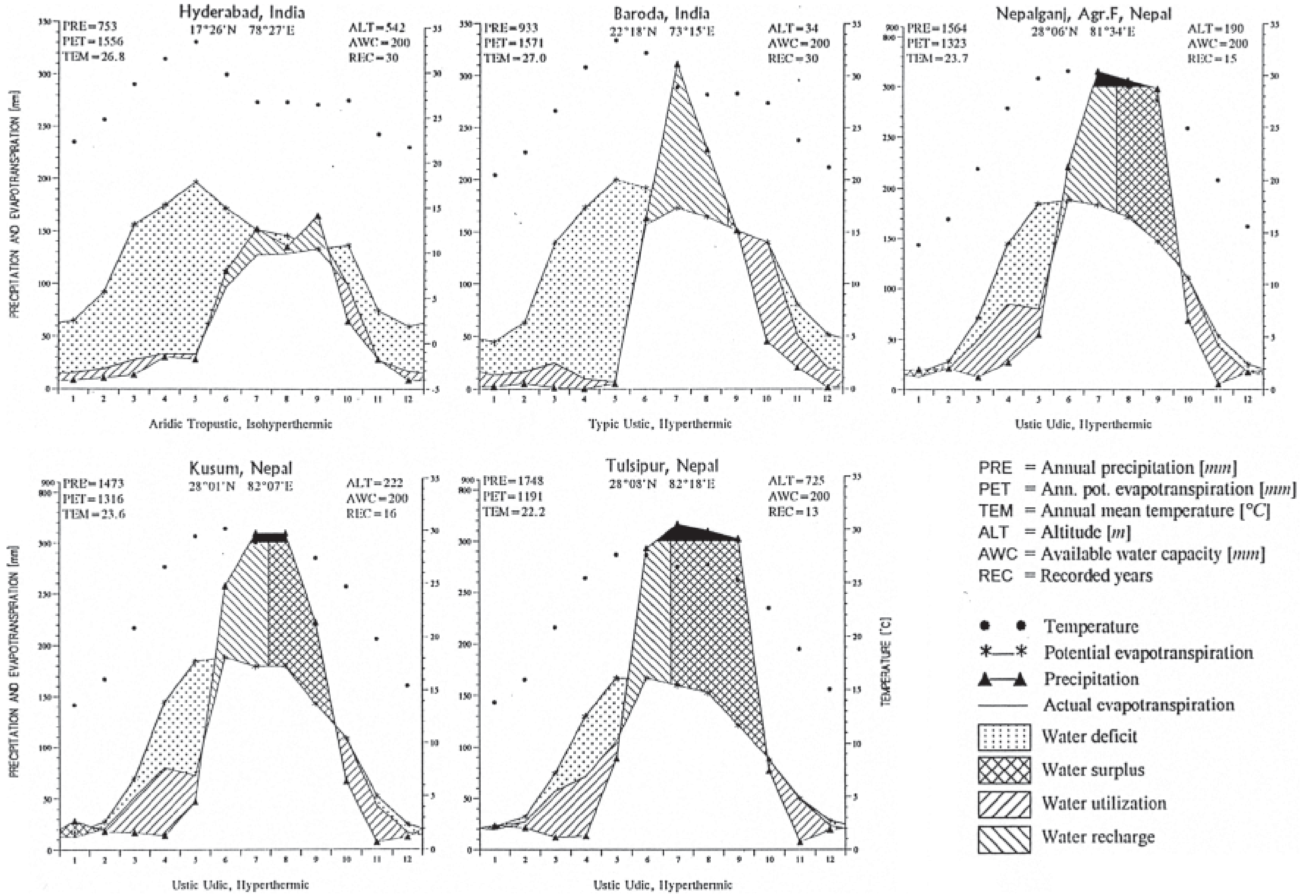

Figure 13. Climate data and soil water balances for selected stations from South India and SW-Nepal.

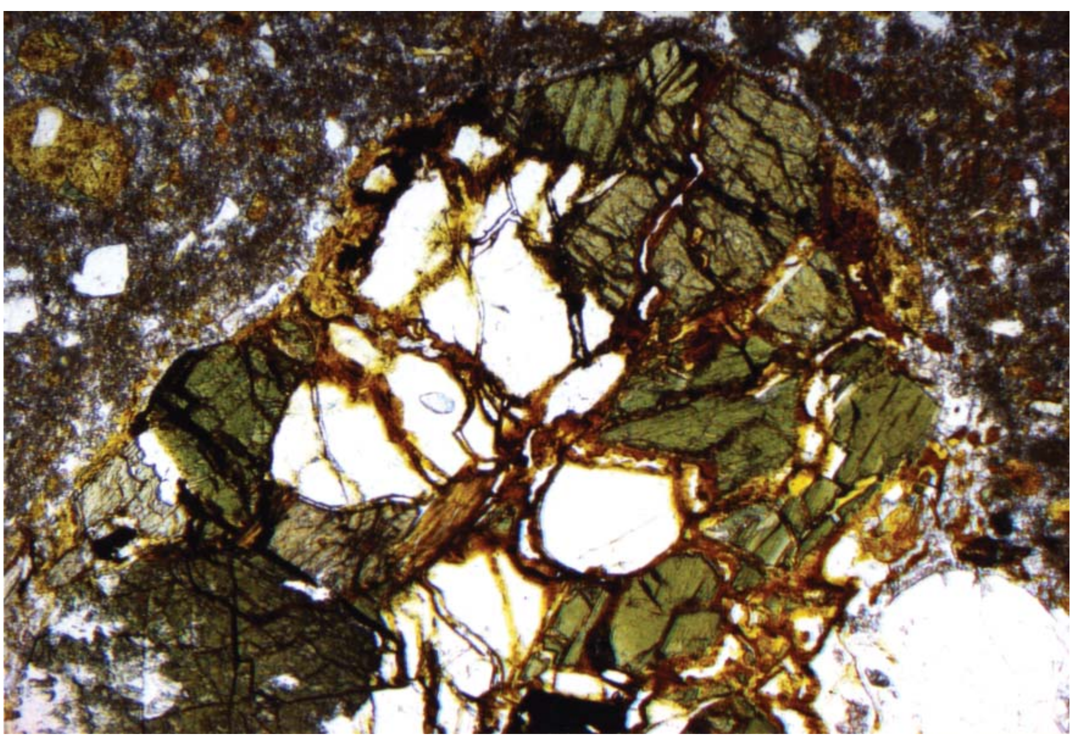

Figure 14. Weathered hornblende in secondary CaCO3 accumulation. Irugur soil near Coimbatore, Crk horizon. Plain polarized light, length of picture 2 mm.

Figure 15. Deforested, then strongly eroded inselberg landscape northwest of Hyderabad (Andhra Pradesh).

tant: pedogenic formation of kaolinites also in the seasonal tropics need a longer time, probably some 100 ka.

Pal *et al.* (1989) even assume that kaolinite formation mainly from smectites as the first weathering product in sarpolites of the Precambrian peninsular gneiss took place in now semiarid India in a pre-Pliocene tropical climate. When the climate becomes drier with the rising of the Western Ghats during the Pliocene-Pleistocene (Brunner, 1970; Kale, 1983), the kaolinites were preserved to the present (also Pal, 2003). However, the question remains whether kaolinite formation was possible also in the post-Pliocene (Pleistocene) period in semiarid India, as it is suggested by our results from Morocco.

Historically, the whole of India was under forest cover except the driest parts of the Rann of Kutch and the Thar Desert (Gaussen *et al.*, 1961-1978; Puri *et al.*, 1978; Bronger, 1998: Figure 1). According to official Indian sources, 22–23% is forest (Das Gupta, 1976; Agarwala, 1985; Tata, 1992); however, the area of true forests is now scarcely 10% of the total area (Misra, 1980; Meher-Homji, 1989), even if the “open forest” areas are included (Puri *et al.*, 1990; Bronger, 1998; Figure 2). The extensive deforestation together with the widely used dryland farming system, in which the land is fallowed for 5–7 months, favors soil erosion, especially under monsoon rainfall patterns. As a result, the depth of the soil cover in large parts of India south of the Ganga plain is only 50–100 cm. In an even larger area between Madhya Pradesh, Orissa and Andhra Pradesh, the depth of the soil cover is only 20–50 cm (Das Gupta, 1980). Because the underlying saprolite is even more susceptible to soil erosion (Scholten, 1997), it is often totally eroded. Therefore, stripped inselbergs are coming to the surface (Figure 15) in many parts of South India. This is a serious situation in view of the high age of the Rhodustalfs.

ACKNOWLEDGEMENTS

We thank the Deutsche Forschungsgemeinschaft for supporting field, micromorphologic and laboratory work (Grants Br 303/19 (1-4), 21 (1-2) and 26 (1-2). My thanks go to two unknown reviewers for their constructive criticism of the manuscript.

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Manuscript received: October 15, 2005
Corrected manuscript received: April 10, 2006
Manuscript accepted: February 5, 2007