

## Clay mineralogy of red clay deposits from the central Carpathian Basin (Hungary): implications for Plio-Pleistocene chemical weathering and palaeoclimate

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**Abstract:** Geochemical and mineralogical studies of palaeosols provide essential information for palaeoclimatic and palaeoenvironmental interpretation of continental deposits and can present a proxy for palaeoclimate. Red clays in the central Carpathian Basin (Hungary) (Tengelic Red Clay Formation; Kerecsend Red Clay Formation), overlain by loess–palaeosol sequences, were studied. Results from geochemical climofunctions applied to Upper Pliocene–Lower Pleistocene red clays and palaeosols located in the Carpathian Basin, and clay mineralogy, indicate that the palaeoclimate was considerably more humid and warmer during the Late Pliocene–Early Pleistocene in comparison to modern values.

**Key Words:** Palaeosol, red clay, loess, Pliocene, Pleistocene, palaeoclimate, East Central Europe

### 1. Introduction

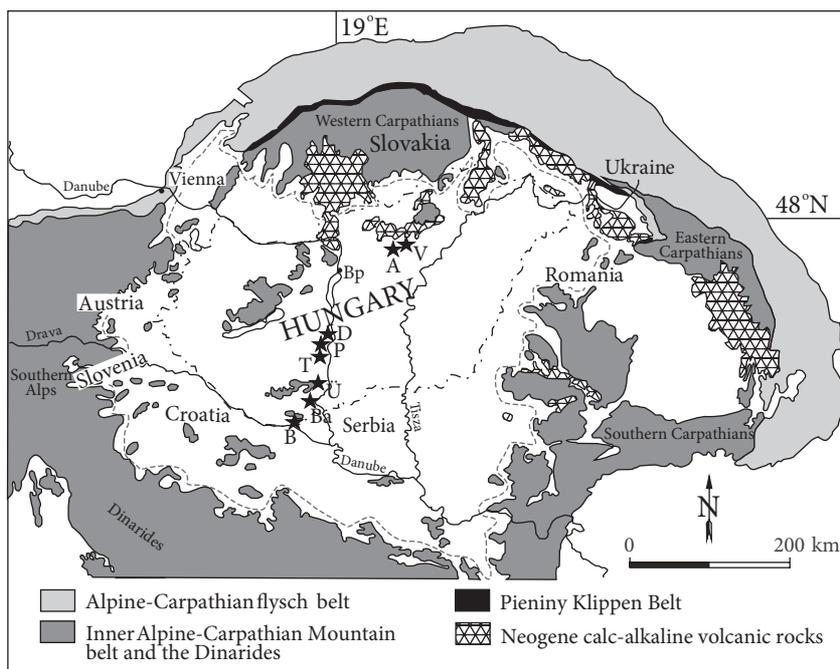
Representative parameters derived from the mineralogical and chemical composition of palaeosols are effective proxies for palaeoclimatic interpretation, and their use, particularly when there is a lack of other proxies, can provide quantitative and detailed palaeoclimatic information (Hamer *et al.* 2007). Palaeosols and Pleistocene loess–palaeosol sequences preserve important information on landscape stability, soil formation, and palaeoenvironment. Using both clay mineralogy and chemical composition, this article describes palaeoclimatic trends in the Late Pliocene–Early Pleistocene of the Carpathian Basin that are recorded in red clay deposits and palaeosols from outcrops and boreholes (Figure 1).

Because palaeoenvironmental and palaeoclimatic data for the Late Pliocene–Early Pleistocene of the Carpathian Basin are limited, the goals of this study are to determine the changes of clay minerals due to chemical weathering and age versus age/time. Moreover, this article provides a higher-resolution proxy that refines previous interpretations of the terrestrial palaeoclimate record of the Carpathian Basin.

#### 1.1. Clay minerals as palaeoclimatic indicators

Clay minerals are phyllosilicates, dominantly produced during chemical weathering processes (Chamley 1989). The nature of clay mineral assemblages (mineral composition of the clay fraction, <2 µm grain-size) is primarily a function of climate, essentially affected by the length of time of weathering, slope, water–rock ratio, and water chemistry (Chamley 1989; Nesbitt & Young 1989; Nesbitt *et al.* 1997; Fürsich *et al.* 2005). Therefore, clay mineralogy is considered to be a powerful tool for interpreting weathering conditions and palaeoclimate (Chamley 1989; Ruffell *et al.* 2002; Sheldon & Tabor 2009), and clay mineral assemblages may provide integrated records of overall climatic impacts (Thiry 2000). In general, illite and chlorite are formed during initial stages of chemical weathering (Nesbitt *et al.* 1980; Nesbitt & Young 1989). Their dominance in a sample indicates relatively fast erosion of the source area (Fürsich *et al.* 2005) and also cold and/or dry conditions. Illite and Al-rich chlorite have been considered to be less sensitive to chemical weathering (e.g., Ruffell *et al.* 2002). During advanced stages of chemical weathering, smectite and

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**Figure 1.** The Carpathian Basin with the locations of sampling sites (map is modified from Varga *et al.* 2011). Grey, dashed line outlines the area of the Carpathian Basin, black dot-dashed line indicates the border of Hungary, black stars are sampling sites, V – Visonta; A – Atkár; D – Dunaföldvár; P – Paks; T – Tengelic; Ü – Üveghuta; Ba – Bár; B – Beremend.

kaolinite are formed (Chamley 1989; Nesbitt & Young 1989; Bronger 2007). Smectite is generally thought to form during weathering in seasonally wet and dry climates with low water-rock ratio and low relief (Ruffell *et al.* 2002; Fürsich *et al.* 2005). The abundance of kaolinite is an especially good indicator of landmasses with hot and humid (subtropical to tropical) climate supported by high water-rock ratio and well-drained, steep slopes (Chamley 1989; Ruffell *et al.* 2002; Fürsich *et al.* 2005; Bronger 2007; Sheldon & Tabor, 2009). In general, dominance of smectite and kaolinite indicates slow erosion rates or erosion of soil horizons formed over long periods of time (Fürsich *et al.* 2005).

## 2. Geological settings

The red clay sediments in the Carpathian Basin are known from both exposures and boreholes. Sections selected for this study are located mainly in the foothills of the Hungarian mountains, except for those in the central part (Figure 1). The red clays (Tengelic Red Clay Formation: TRCF; Kerecsend Red Clay Formation: KRCF) are widespread in the hilly and mountainous areas of the Carpathian Basin underlying the Pleistocene Paks Loess Formation (PLF) (Jámbor 1997; Schweitzer & Szöör 1997; Viczián 2002, 2007; Kovács 2003, 2008). The thickness of red clay ranges from 4 to 90 m (Jámbor 1997; Viczián

2002; Kovács *et al.* 2008, 2011). The age of the Tengelic Red Clay Formation is ca. 3.5–1.0 Ma (Gyalog & Budai 2004; Kolozsár 2004, 2010), and that of the Kerecsend Red Clay Formation ca. 1.5–0.5 Ma (Jámbor 2001).

The red clays and palaeosols have a reddish (7.5YR 7/4) or reddish-brown (5YR 5/6) colour, and vertic features are most prominent in the older, reddest palaeosols. The reddish colour of soils and palaeosols is attributed to hematite, goethite, maghemite, and/or “amorphous” Fe oxides, formed pedogenically or during early diagenesis as the result of dehydration or oxidation of Fe oxyhydroxides, or they may be inherited from the parent material. Generally, red clay displays a prismatic structure with slickensides, stress surfaces, and brown and yellowish spots. Calcretes, 3–5 cm in diameter, occur in the lower part of the red clay. Usually, the lower part (Bk horizon with carbonate nodules) is paler than the upper part (Bt horizon). Black Fe–Mn stains are generally abundant throughout the entire red clay unit. According to Fekete (2002), the kaolinite-rich red clays (TRCF, Beremend Member) are ferralsols (oxisols); the younger red clays and palaeosols are vertisol-type palaeosols.

In extensive areas of Central and SE Transdanubia and in certain occurrences east of the Danube River, the lower part of the Pleistocene is represented by the TRCF, consisting of red and variegated clay, sand, and silt of

fluvial facies. It is underlain unconformably by Upper Pannonian (Zanclean) or older deposits and generally covered by loess. Loess is widespread in alluvial plains and, to a lesser extent, on slopes of the mountainous areas as well. However, it reaches its widest extent and greatest thickness in hilly regions of Central and SE Transdanubia (Jámbor 2001; Koloszár 2010). Loess is a wind-blown sediment of silt-size formed in periglacial areas during cold and dry periods of the Pleistocene. It may have transitions to, or may alternate with, aeolian sand deposits. Loess sequences can be interrupted by palaeosol horizons formed during milder interglacial periods. In hilly and mountainous regions the loess complex is called the Paks Loess Formation (PLF), which is underlain in SE Transdanubia by the TRCF. According to Jámbor (1997), loess was formed in hilly regions between 1.2 Ma and 12 ka ago. Stratigraphic relations of these terrestrial sediments are described in classical studies of the vertebrate fauna by Kretzoi (1956, 1969) and Jánosy (1986) and in more recent summaries by Koloszár (2004), Koloszár and Marsi (2005), and Kovács *et al.* (2008). Schweitzer and Szöör (1997) distinguished and characterised the subsequent periods set up by the former authors as Ruscinian (4.5–3.0 Ma), Villanyian (3.0–1.8 Ma), and Biharian (younger than 1.8 Ma).

### 2.1. Tengelic Red Clay Formation (TRCF)

Red clays are widespread in the hilly and mountainous areas of Hungary underlying the Pleistocene PLF. According to Schweitzer and Szöör (1997), red clays can be subdivided into 2 compositional groups: the first rich in kaolinite, and the other rich in illite and smectite. The kaolinite-rich variety seems to be the older one (“Beremend Member”, Koloszár 2004), while the illite–smectite rich variety is generally younger, more widespread, and occurs in hilly areas (“Tengelic Member”, Koloszár 2004). Red clays (Beremend site, Figure 2; e.g., Kovács *et al.* 2011, p. 38, figure 4B) of Late Pliocene–Early Pleistocene age (3.3–2.4 Ma, MN16 mammal biozone) were dated using vertebrate mammals by Jánosy (1986) and Kretzoi (1987). The red clay (in the Tengelic-2 borehole, Figure 2) is the uppermost bed of a 25–60 m thick sequence consisting from the bottom upward of alluvial sand, occasional bentonite derived from basalt tuff, eluvial-deluvial variegated clay and clayey silt, and finally the red clay, which is of residual facies. The bentonite layer, derived from basalt, is important for age determination. These potassic volcanic rocks dated at  $2.17 \pm 0.17$  Ma (K–Ar method) were recovered from boreholes at Bár (Balogh *et al.* 1986). At Bár, in Bá-4 borehole (Figure 2), K-rich basalt and basalt pyroclastite intercalations can be found between red clay layers. The whole sequence was deposited after a considerable hiatus on the eroded surface of Upper Pannonian sediments. Its age is supposed to be Early Pleistocene. The thickness of the red clay varies

from a few metres up to nearly 20 m. The red clay beds are overlain by other red clay strata forming the lower members of the PLF (Figure 2). The colour is actually less deep red and has been called “reddish” by Schweitzer and Szöör (1997).

### 2.2. Kerecsend Red Clay Formation (KRCF)

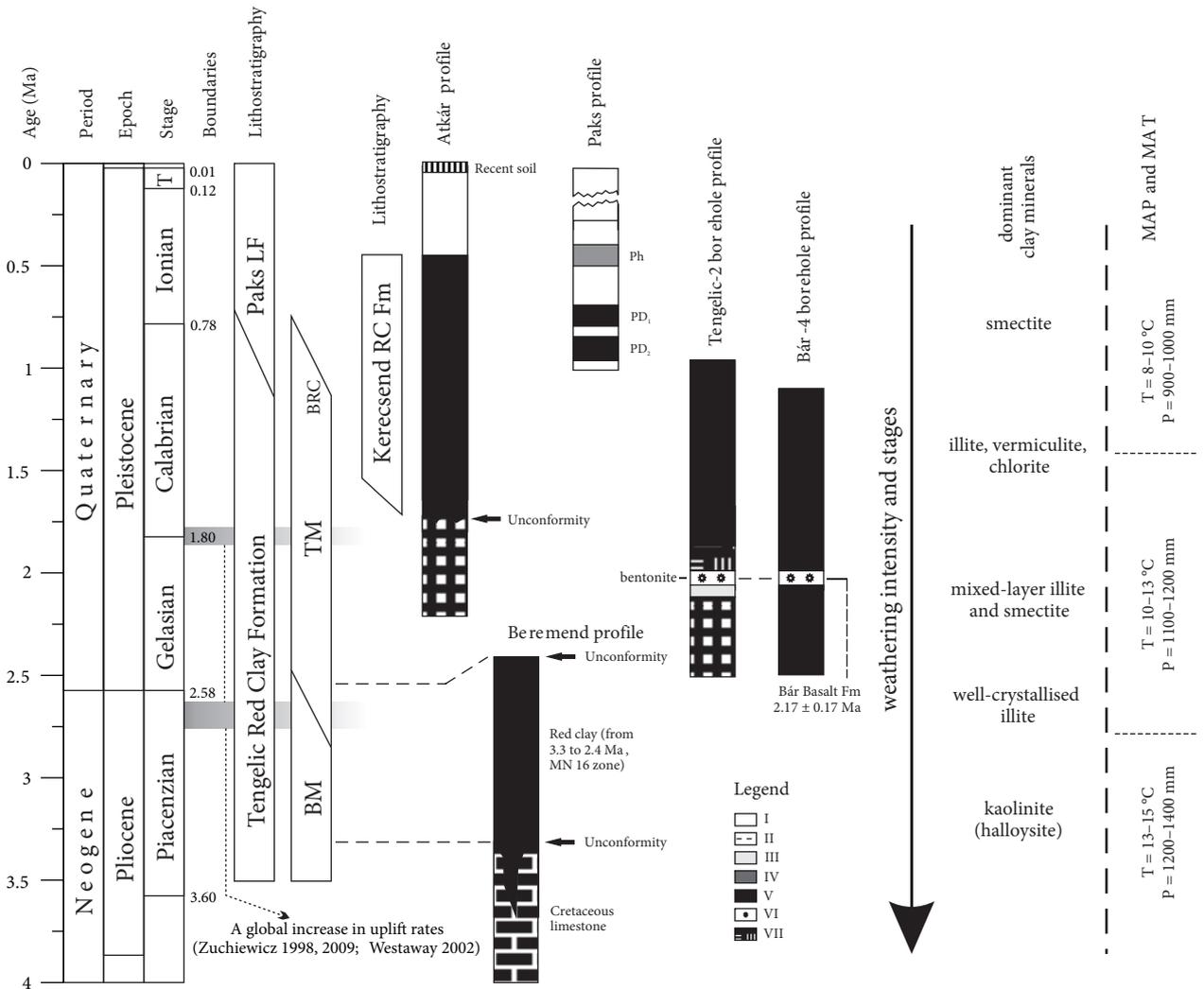
In the mountain areas of the northern part of the country, red clay occurs in karstic limestone areas as depression and cave fills. Jámbor (2001) supposed that this clay was formed during the Middle Pleistocene, or possibly even prior to the Quaternary. Red clay fillings, containing bone fossils, have been described in several places in the caves of the northeastern mountains. Age determination was based on stratigraphical position and vertebrate fauna (Jánosy 1986; Kretzoi 1987). The oldest vertebrate fossils are 700,000 years old, although they mark only the age of the accumulation, while the red clays could be significantly older. In the northern part of the country, in the southern foothills of the mountains, cross-bedded sand or sandy clay is overlain by a 3–20 m thick red clay horizon (KRCF, Figure 2), about 3–6 m thick (Atkár site, Figure 2; e.g., Kovács *et al.* 2011, p. 38, figure 4A).

### 2.3. Paks Loess Formation (PLF), lower palaeosols

The Paks loess profile is located in the mid-Carpathian Basin on the right bank of the River Danube (Figures 1 and 2). Boreholes reveal that the whole loess–palaeosol series is underlain by a clay, silt, and red clay sequence called the TRCF (Koloszár 2004; Kovács *et al.* 2008, 2011), which is ca. 60 m thick and represents approximately the last 1 Ma (Pécsi 1979). Two lithological units have been distinguished within the Paks Loess Formation: 1) the Young Loess Series (YLS; MIS 2–10) and 2) the Old Loess Series (OLS; MIS 11–22) (Pécsi 1995; Gábris 2007). Three loess layers and 3 Mediterranean (terra rossa) type palaeosols (in Figure 2, PD<sub>2</sub>, PD<sub>1</sub>) constitute the lower part of the OLS, while the upper part of the OLS consists of 3 loess layers, 2 brown forest soils, and a pseudogley soil (Újvári *et al.* in press). As shown in Figure 2, the PD<sub>2</sub> fossil soil was the lowest palaeosol studied in the exposure (e.g., Kovács *et al.* 2011, p. 38, figure 4C). The stratigraphic position of red clays (palaeosols), on the basis of the Middle Pleistocene PLF, is Lower Pleistocene to lowest Middle Pleistocene (0.8 to ~1.2 million years). The stratigraphic position is given by the scheme of Koloszár and Marsi (2002).

## 3. Methods

Red clay, palaeosol, and loess samples from the Carpathian Basin were collected during fieldwork. A total of 80 samples of red clay and palaeosol (and loess for comparison) were taken from the northern, southern, and the central part of Hungary. The sequences were continuously sampled for analysis at 10–20 cm intervals.



**Figure 2.** Geochronological and stratigraphical framework of the Hungarian red clays with the stratigraphic position of the studied profiles. Global chronostratigraphy is from Gibbard and Cohen (2008). T – Tarantian, Paks LF – Paks Loess Formation, BM – Beremend Member, TM – Tengelic Member, BRC – Basal Red Clays of the Paks Loess Formation (after Kretzoi 1987; Jámboor 1997; Schweitzer & Szöör 1997; Kolozsár 2004; Marsi & Kolozsár 2004; Kovács *et al.* 2008). Ph – Paks sandy soil complex, PD<sub>1,2</sub> – Paks Double, MN 16 zone – European Land Mammal Mega Zone MN 16 (roughly coeval with the Piacenzian between 3.600 and 2.588 Ma). Legend: I – loess, II – sand, III – sandy-loamy marl, IV – palaeosol, V – (terra rossa)/red clays, VI – basalt/bentonite, VII – sandy clay.

### 3.1. Grain-size analyses

The grain-size distribution of all samples was measured by laser diffraction (Fritsch Analysette 22) methods using the approach described by Konert and Vandenberghe (1997) and Kovács (2008). After processing the samples with 10 mL of 30% H<sub>2</sub>O<sub>2</sub> and 10 mL of 10% HCl to remove organic matter and carbonate, respectively, 10 mL of 0.05 N (NaPO<sub>3</sub>)<sub>6</sub> was added to the sample, which was then ultrasonicated for about 15 min. Subsequently, the sample was transferred to the laser grain-size analyser.

### 3.2. Mineralogical analyses

Clay mineralogy in this paper is based on X-ray powder diffraction (XRD) analyses of red clay, loess, and palaeosol

samples gathered from published papers and technical reports from the last 2 decades. Most of the measurements were performed at the Geological Institute of Hungary, Budapest (Földvári & Kovács-Pálffy 2002; Dezső *et al.* 2007; Viczián 2007 and references therein) and at the Department of Earth and Environmental Sciences, University of Pannonia, Veszprém, Hungary (Dezső *et al.* 2007; Viczián 2007; Varga *et al.* 2011; Újvári *et al.* in press). Additionally, results of Berényi Üveges *et al.* 2003 and Vincze *et al.* 2005 were also used and interpreted. Complete descriptions of the methods including instruments used for XRD analyses are available in the aforementioned papers.

### 3.3. Geochemical analyses

Loess and palaeosol samples were analysed for major and trace element abundances with X-ray fluorescence spectrometry (XRF) using a Thermo ARL Advant'XP+ sequential XRF spectrometer in the GeoAnalytical Laboratory of Washington State University, Pullman, WA, USA. After drying, samples were prepared for analysis by grinding to a very fine powder, weighing with di-lithium tetraborate flux (2:1 flux:sample), fusing at 1000 °C in muffle oven, and cooling. The bead is then reground, refused, and polished on diamond laps to provide a smooth flat analysis surface. The major element concentrations are expressed as wt%, volatile-free, with all the iron expressed as FeO<sub>tot</sub>. Loss on ignition (LOI) was obtained by weighing after 16 h of calcination at 900 °C. Analytical uncertainties are ±2% for the major elements (except Na<sub>2</sub>O).

Individual data were published by Újvári *et al.* (in press) for loess and palaeosol samples. Red clay data, with a complete description of the method used for chemical analyses, come from Kovács (2007).

### 3.4. Palaeoproxy Indicators

A variety of semiquantitative and quantitative tools, including mineralogical and geochemical proxies, have been developed to examine past weathering and pedogenesis, and to reconstruct both palaeoenvironmental and palaeoclimatic conditions at the time of palaeosol formation (e.g., Bokhorst *et al.* 2009; Sheldon & Tabor 2009; Buggle *et al.* 2011; Gulbranson *et al.* 2011). The concept of geochemical proxies of mineral alteration (i.e. weathering indices) relies on the selective removal of soluble and mobile elements from a weathering profile compared to the relative enrichment of rather immobile and nonsoluble elements (Nesbitt & Young 1982; Buggle *et al.* 2011). Based on this principle, simple ratios of bulk element composition, together with chemical weathering indices, have successfully been used for the reconstruction of palaeoenvironmental conditions of palaeosols and loess–palaeosol successions (e.g., Retallack 2001; Sheldon 2006; Kovács 2007; Bokhorst *et al.* 2009; Buggle *et al.* 2011; Muhs *et al.* 2011).

Major element concentrations of red clays and palaeosols have been used to reconstruct patterns in the long-term chemical weathering of the land surface through the use of the chemical index of alteration (CIA; Nesbitt & Young 1982), the chemical index of weathering (CIW; Harnois 1988), the chemical index of alteration minus potassium (CIA–K; Maynard 1992; Fedo *et al.* 1995), and the chemical proxy of alteration (CPA; Buggle *et al.* 2011). As weathering progresses, the value of the CIA, CIW, or CIA–K of soil B horizons will increase relative to the unaltered parent material. Considering element behaviour during weathering or diagenesis, the chemical proxy of alteration (CPA) is proposed as the most appropriate index

for silicate weathering (Buggle *et al.* 2011). However, the CIA is used in this study, because all the former calculations were based on this index. Moreover, the chemical index of alteration minus potassium (CIA–K) is used in the geochemical climofunctions (see in the next part).

Earlier reviews suggest generally that clay mineralogy follows a weathering pattern, from hot and humid to cool and dry, in the order of kaolinite → smectite → vermiculite → chlorite and mixed-layer phyllosilicates → illite and mica (e.g., Retallack 2001; Sheldon & Tabor 2009). In the context of this study, the data collected from the bulk as well as the clay mineral analysis are supposed to serve as a proper basis for an estimate of the weathering intensity of the individual samples (Terhorst *et al.* 2012). The most sensitive minerals, such as carbonates and chlorite, will probably be dissolved or transformed first, and, with progressive weathering, the more stable minerals also, such as mica and feldspars (Terhorst *et al.* 2012).

### 3.5. Geochemical climofunctions

The degree of chemical weathering in soils increases with mean annual precipitation (P; mm) and mean annual temperature (T; °C). These relationships were quantified by Sheldon *et al.* (2002) and Nordt and Driese (2010) using a database of major-element chemical analyses of modern soils, which were selected from the compilation of Marbut (1935). This is based on the spatial extent and continuity of coverage on a continental scale to ensure representation of a large range of climate regimes.

According to Sheldon *et al.* (2002), mean annual precipitation (MAP) can be related to the chemical index of alteration without potassium (CIA–K) and is calibrated for precipitation values between 200 and 1600 mm/year:

$$\text{MAP (mm/year)} = 14.265(\text{CIA-K}) - 37.632, \quad (1)$$

where CIA–K =  $100 \times [\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO} + \text{Na}_2\text{O})]$  and  $R^2 = 0.73$  ( $R^2$  is the coefficient of determination in linear regression), with an error of ±182 mm/year. Results obtained with this method are consistent with independent estimates from other proxies, such as plant fossils (Sheldon & Retallack 2004).

A function by Sheldon (2006) for use with inceptisols allows mean annual temperature (MAT) to be calculated as follows:

$$\text{MAT (°C)} = 46.94C + 3.99, \quad (2)$$

where  $C = m\text{Al}/m\text{Si}$  and  $R^2 = 0.96$ , with an error of ±0.6 °C ( $m$  is the molar ratio).

Another climofunction was used for MAP estimation, as well established by Nordt and Driese (2010):

$$\text{MAP (mm/year)} = 22.69(\text{CALMAG}) - 435.8, \quad (3)$$

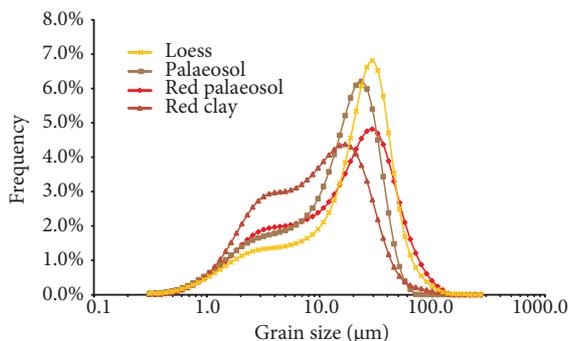
where CALMAG (calcium-magnesium index) =  $100 \times [\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO} + \text{MgO})]$ . This function is restricted to vertisols; oxides are in units of moles.

The palaeoclimatological results are shown in the Table.

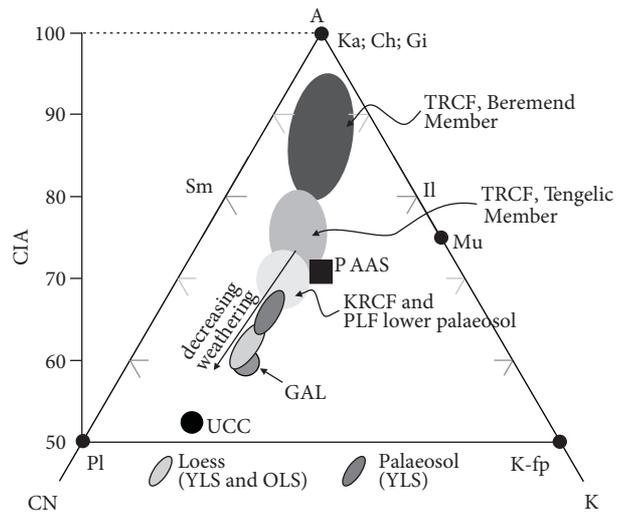
## 4. Results

### 4.1. Granulometry

Grain-size distributions of detrital sediments are usually regarded as useful parameters in characterising sedimentary environments and dynamics. Grain-size distributions of red clay were analysed and compared with typical aeolian loess and palaeosols developed on loess (Kovács 2003, 2008; Kovács *et al.* 2008; Varga 2011). The grain-size distribution curves of loess deposits closely resemble the red clays and palaeosols (Figure 3). The bimodal pattern could also be identified, indicating that 2 sediment populations have been involved in the loess formation. The pronounced peak in the coarse silt fraction and the secondary maximum in the clay-, fine silt fraction is a common characteristic of loess deposits. The possible factors that could be the cause of secondary maxima are a second dust source area; background dust-load; and postdepositional weathering or dispersion of silt- and sand-sized clay-aggregates. As demonstrated by Yang and Ding (2004) and Varga (2011), pedogenic processes and aggregation have restricted effect on the grain-size of loess. The fine-grained component was mainly transported by upper level air-flow, and was deposited far from the source areas. Detailed discussion on the effect of background dust-load can be found in Varga *et al.* (2012). The fine-grained populations in the grain-size distribution curves of loess deposits have a lower percentage than in the red clay samples. This suggests that for loess the proximal mineral material may have played a much larger role in the



**Figure 3.** Grain-size distribution curves of the red clay (TRCF), red palaeosol (KRCF), and Quaternary loess and palaeosol (PLF) samples from Hungary.



**Figure 4.** Ternary A-CN-K diagram (Nesbitt & Young 1982) of the red clay, palaeosol, and loess samples (in molar proportions). The samples plot subparallel to the A-CN join, suggesting an ideal weathering of a slightly more felsic source than the UCC (Rudnick & Gao 2003). Abbreviations are as follows: Sm – Smectite, Il – Illite, Mu – Muscovite, Ka – Kaolinite, Ch – Chlorite, Gi – Gibbsite, Pl – Plagioclase, K-fp – K-feldspar, UCC – upper continental crust, GAL – global average loess (Újvári *et al.* 2008), PAAS – post-Archaean Australian Shale (Taylor & McLennan 1985). Note that only the top 50% of the triangle is shown.

sedimentation than did the background dust. However, this does not mean that the amount of the distal dust material was reduced, but the increased quantity of the local material caused a decrease in the relative proportion of the fine-grained particles. Particle-size characteristics and micromorphological investigations suggest that most of the red clay is wind-blown in origin (Kovács 2008; Kovács *et al.* 2008; Varga 2011). Detailed granulometric analyses of red clays show similar bimodal grain-size distribution patterns to loess horizons, as in the Chinese Loess Plateau (Yang & Ding 2004; Kovács *et al.* 2008). More detailed grain-size properties of red clays and palaeosols can be found in Kovács (2008) and Varga (2011).

### 4.2. Mineralogy

**TRCF, Beremend Member.** According to the unpublished report by Marsi *et al.* (2001 in Viczián 2007), bulk red clay samples are dominated by smectite with additional disordered kaolinite. Hematite, Ti-oxides, and some quartz and illite are present as well. In the separated <2 µm fraction, dominance of highly disordered kaolinite is the most obvious (60%–80%), while smectite (20%–40%) and illite + illite/smectite (<10%) are low, and gibbsite was identified only as traces in some samples (Marsi *et al.* 2001 in Viczián 2007). Dezső *et al.* (2007) reported red clay samples with very similar mineralogy from the same

locality, emphasising a more significant role of gibbsite in the clay fraction (up to 27%). Viczián (2007) found that kaolinite is disordered, with a mixed-layer kaolinite/smectite character (Figure 2).

*TRCF, Tengelic Member.* A well-studied example of the upper part of the TRCF occurs in the fossil vertebrate locality Somssich Hill Nr. 2. The sequence was dated by Jánossy (1986) as the end of Lower Pleistocene. Based on Viczián (2007), bulk composition of a fissure-filling yellow silt can be characterised by the dominance of calcite and quartz with some feldspar. Discrete and well-crystallised illite was found as the main clay mineral accompanied by minor chlorite, kaolinite with goethite, and amorphous iron hydroxide. Discrete and well-crystallised illite phases were found as the main clay components, accompanied by less abundant disordered smectite, chlorite, and kaolinite in this monotonous silty clay sequence. The only systematic variation is that kaolinite is at the bottom and chlorite at the top of the sequence; in the middle both minerals occur.

In particular, at Dunaföldvár (near Paks) mixed-layer illite/smectites were identified as the main clay minerals, accompanied by illite and kaolinite (Figures 1 and 2). In borehole Tengelic-2, which is the type section of the formation, brownish-reddish clay contains poorly crystallised but discrete smectite and illite, and minor chlorite. Recently detailed geological and palaeopedological studies were carried out on occurrences of the TRCF (e.g. Üveghuta; Figure 1). This clay is poor in carbonates and contains predominantly smectites. Bulk composition of the 'reddish' clay samples examined by Földvári and Kovács-Pálffy (2002) shows dominance of quartz, feldspars, and smectite, which is replaced by vermiculite in some samples. Illite, minor chlorite, and very scarce kaolinite are present as well. As for the separated clay fraction, illite and 14 Å phases (smectite and/or vermiculite) are the major clay minerals. A variety of different mixed-layer clay minerals between vermiculite, smectite, chlorite, and illite were identified. Kaolinite was detected in traces.

*KRCF.* Vincze *et al.* (2005) showed that in the bulk red clay samples collected from the NE Hungarian region, the most abundant minerals are quartz and phyllosilicates, while feldspars are minor constituents. Goethite, hematite, and dolomite are accessory phases but the systematic presence of the amorphous material is significant. Among the clay minerals smectite (montmorillonite) and illite are dominant, while kaolinite and chlorite are subordinate. It should be noted that lithostratigraphy of these red clays is debated, while they can represent either the TRCF or the KRCF (Vincze *et al.* 2005). Based on the XRD investigations of Berényi Üveges *et al.* (2003), in a representative palaeosol profile near Visonta (Figure 1), the dominant clay mineral is smectite in all layers and

horizons. Kaolinite, illite, vermiculite, chlorite, illite/smectite, and chlorite/vermiculite were identified in most of the samples. Smectites in the red palaeosol have primarily high layer charge; both montmorillonitic and beidellitic character is present.

*PLF, Lower Palaeosol.* The bulk mineralogical composition of the sediments, estimated from XRD data, indicates that quartz, smectite (up to 30% in loess and up to 40% in palaeosol), and carbonates are the dominant minerals (Újvári *et al.* in press). Loess samples contain higher proportions of calcite and dolomite compared to palaeosols, which can be characterised by smectite dominance. Illitic material (illite ± muscovite), together with chlorite, is present in all samples but usually in small proportions (<10%); YLS sediments, however, have a relatively higher bulk illite ± muscovite and chlorite content compared to the OLS loess samples. Goethite is present in 3 samples in the lower part of the MB palaeosol (OLS), whereas hematite occurs only in a single YLS palaeosol sample.

Other authors have also noted large amounts of illite, chlorite, and smectite (especially in lower palaeosols) with heterogeneous distribution in the Paks section (Pécsi-Donáth 1979; Nemezc *et al.* 2000). Pécsi (1993) demonstrated that bulk samples of the PLF older palaeosols are composed of quartz, feldspar, calcite, dolomite, abundant 'hydromica', and chlorite, with minor montmorillonite and kaolinite. Kaolinite-bearing samples also contain traces of Al-hydroxide phases. At the Beremend site, Dezsó *et al.* (2007) showed that 'reddish' lower palaeosol of the PLF is dominated by quartz and well-crystallised illite (probably 2M polytype), with minor kaolinite and smectite. Goethite is present as well. As for the clay fraction, the above-mentioned clay minerals are identified, but chlorite also occurs. Interestingly, the smectite described from the whole rock samples shows a highly expandable mixed-layer illite/smectite character.

#### 4.3. Geochemical properties

The chemical composition of the red clay deposits in Hungary is dominated by SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, CaO, MgO, and K<sub>2</sub>O (Kovács 2007; Kovács *et al.* 2008). The chemical index of alteration (CIA) for the samples varies from 64 to 93 (Figure 4). As shown in the A-CN-K diagram (Figure 4), samples are distributed along the A-CN line and tend to approach the A-pole, reflecting a process in which K<sub>2</sub>O is leached out and Al<sub>2</sub>O<sub>3</sub> is increased in the samples, i.e. the dissolution of feldspar minerals and the production of new clay minerals (smectite, illite, and kaolinite). This pattern suggests that the chemical weathering of the sediments resulted in removal of Ca and Na (primarily plagioclase) from the source rocks and less intense leaching of K, whereas the stronger chemical weathering in the sediments caused considerable dissolution of Ca-

Na-bearing host minerals and even K-bearing minerals (mainly K-feldspar) as well.

In the OLS palaeosol samples (PLF) from the Paks section, a heterogeneous chemical composition is apparent.  $\text{SiO}_2$  content and CaO content both vary widely (Újvári *et al.* in press). Nevertheless, other major elements have a less pronounced variation. CIA values are in the range of 61–71 (average:  $68 \pm 1$ ), which are higher than the UCC (upper continental crust) and GAL (global average loess) values of 53 and 60 (Rudnick & Gao 2003; Újvári *et al.* 2008) and slightly lower than the PAAS (post-Archaean Australian Shale) value of 70 (Taylor & McLennan 1985). Palaeosol samples show higher CIA values than intervening loess (OLS loess average:  $64 \pm 2$ ), indicating stronger weathering of fossil soils. The YLS samples have slightly lower CIA values, ranging from 60 to 68 (Figure 5). The PLF samples plot subparallel to the A–CN line, suggesting ideal weathering of a slightly more felsic source than the UCC (Rudnick & Gao 2003). Furthermore, a general trend of decreasing chemical weathering intensity from the TRCF to the PLF is unequivocal, as demonstrated by CIA (Figure 4).

#### 4.4. Palaeoprecipitation and palaeotemperature

Reconstructed palaeoclimate results indicate that during the development of red clays and palaeosols, the climate for most of the time was considerably wetter than the modern climate (Table). The modern climate of the Carpathian Basin (Hungary) is dominated by *Cfb* climate with hot summers and mild winters (Fábián & Matyasovszky 2010). In the Köppen climate classification, *Cfb* means temperate marine west coast climate (Kottek *et al.* 2006). Current MAP within the Carpathian Basin is 500–750 mm/year, and the MAT is 10–11 °C, with a seasonal range from 20 °C in July to –1 °C in January (Justyák 1998). The MAP values obtained from palaeosols (Eq. [1]) had a range of 890–1370 ± 182 mm/year and a mean of 1200 mm/year

(Figure 5a) and 884–1774 mm/year (using Eq. [3]). The MAT values from palaeosols (Eq. [2]) ranged from 9 to 14 °C (Figure 5b), with a mean value of 11.5 °C (SD = 1.5 °C).

The MAP values from TRCF, Beremend Mb., vary from 1200 and 1400 mm/year using Eq. (1) and fall between 1400 and 1800 mm/year using Eq. (3). The estimated MAP values from TRCF (Tengelic Mb.) range from 1100 to 1200 mm/year (Eq. [1]) and 1200 to 1400 mm/year (Eq. [3]). The MAP values from KRCF vary from 900 and 1000 mm/year (Eq. [1]) and are 1000–1200 mm/year (Eq. [3]). The calculated precipitation values of PLF (lower palaeosols) range from 800 to 900 (Eq. [1]) and 900 to 1000 mm/year (Eq. [3]).

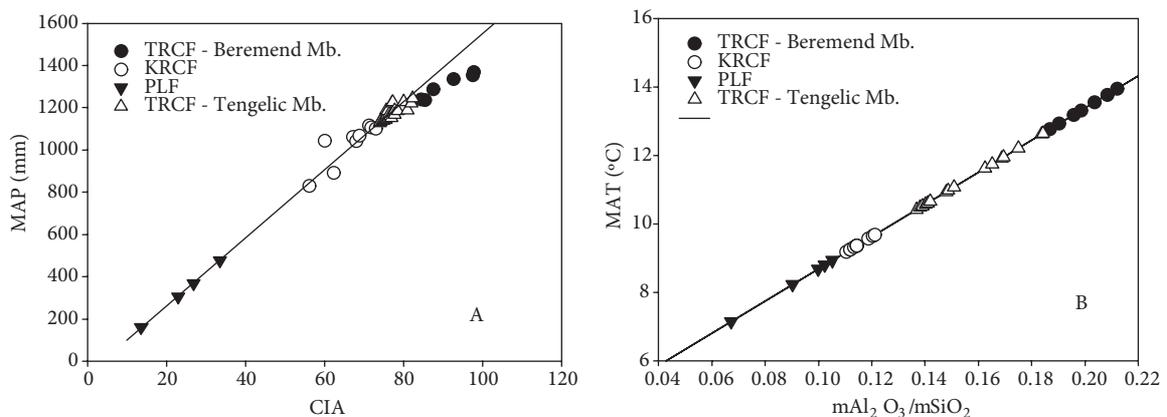
The MAT values of TRCF, Beremend Mb., are 13–15 °C, and for the Tengelic Mb. they fall between 10 and 13 °C. The calculated temperature values of KRCF range from 8 to 10 °C and 7 to 10 °C for the PLF, lower palaeosol.

## 5. Discussion and conclusions

### 5.1. Palaeoclimatological interpretation

Reddish palaeosols are common in Pleistocene loess–palaeosol sequences throughout Central and SE Europe and East and Central Asia, as well as throughout the geological record. Thus, colour alone may not be diagnostic of palaeosol climate (Sheldon & Tabor 2009).

The older type (Beremend Mb of the TRCF) is red kaolinitic clay containing typically disordered kaolinite, mixed-layer smectite/kaolinite, smectite, and rare gibbsite (Viczián 2007; Kovács *et al.* 2011). According to the previous model (Dezső *et al.* 2007; Viczián 2007), gibbsite in low amounts was most probably formed during the Csarnótan period, together with kaolinite in the weathering crust on the surface. In exceptional cases the preservation of high-gibbsitic clays in an older generation of fissures cannot be completely excluded. Transformation in the ground water may have produced smectites but



**Figure 5.** Relationship between A) mean annual precipitation (MAP) and CIA–K and B) mean annual temperature (MAT) and the molecular weathering ratio of  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  in the red clays, palaeosols, and loess deposits.

**Table.** Climate data set of Carpathian Basin obtained from previous studies and the new results. 1 – Eronen & Rook 2004; 2 – van Dam 2006; 3 – Montuire *et al.* 2006; 4 – Haywood *et al.* 2000; 5 – Haywood & Valdes 2004; 6 – Chandler *et al.* 2008; 7 – Mosbrugger *et al.* 2005; 8 – Justyák 1998.

Age (Ma)	MN zone	Munsell colour (dry)	MAT	MAP	Ref.	MAT (this study)	MAP <sup>a</sup> (this study)	MAP <sup>b</sup> (this study)	Studied deposits
4.2–3.2	MN15	5YR 4/4	10–15	1150	1, 2, 3	13–15	1200–1400	1400–1800	TRCF, BM
3.15	–	–	n.d.	951	4, 5, 6	13–15	1200–1400	1400–1800	TRCF, BM
3.2–2.5	MN16	5YR 4/6	n.d.	700	1	10–13	1100–1200	1200–1400	TRCF, TM
2.5–1.8	MN17	7.5YR 7/4 5YR 5/6	10.6–12.4	990	1, 7	8–10	900–1000	1000–1200	KRCF
1.8–0.9		2.5YR 4/8	n.d.	n.d.	–	7–10	800–1000	900–1000	PLF, lower palaeosol
recent	–	–	10–11	500–750	8	–	–	–	–

Note: MAT values in °C; MAP values in mm; Ref. – references; n.d. – no data; MAP<sup>a</sup> – Eq. (1); MAP<sup>b</sup> – Eq. (3).

did not go far enough to produce kaolinite or gibbsite. The vertebrate fauna of the Csarnótan biostratigraphic stage (Kretzoi 1969) indicates warm and humid climatic conditions, which were compared by Kaiser (1999) to the recent climate along the Atlantic coasts of Portugal, and by Koloszar *et al.* (2000) to the recent climate of SE Asia. According to Marsi *et al.* (2001) the source material of the red clay fill in the Beremend quarry was mainly an autochthonous weathering crust on the top of the isolated elevation of the limestone block at Beremend. Viczián (2007) considered that it was formed in the local subaerial weathering crust in a warm, humid, subtropical, or monsoon climate. Our palaeoclimatic results show that this red clay was developed under a humid subtropical climate (Köppen climate classification *Cfa*), which is a climate zone characterised by hot, humid summers and generally mild to cool winters (Kottek *et al.* 2006). This type of climate is found in northern Vietnam, the south-eastern quarter of mainland China, the northern half of Taiwan, and narrow coastal areas of South Korea.

It is important to note, however, that kaolinitic palaeoprofiles may have not all formed in tropical to subtropical climates, and some may even not have formed under wet conditions (e.g., Chamley 1989; Thiry 2000). Therefore, other effects (e.g., tectonic rejuvenation and the role of surface uplift) on the mineralogy and the CIA values must be taken into consideration as well (e.g., Kuhlemann *et al.* 2008; Mikes *et al.* 2011; Varga *et al.* 2011; Újvári *et al.* in press), although the climatic control seems to be obvious in explaining our dataset.

On one hand, Bronger (2007) has stated that the efficiency of weathering under tropical climates has often been overestimated. In South India, above a threshold of about 2000 mm (6 humid months), deep weathering is a recent process leading to the formation of kaolinites; above

2500 mm (10 humid months) on the windward side of the Western Ghats, it leads also to the formation of gibbsite. Pedogenic formation of kaolinites also in the seasonal tropics needs a longer time, probably some 100 ka. In the Atlantic coastal region of Morocco, in a time span of several 100 ka, the direction of weathering goes towards strong pedogenic kaolinite formation, showing poor crystallinity of the fireclay type.

On the other hand, depending on the global climate, weathering conditions in the Alpine realm changed from tropical (Eocene) to subtropical (Early and Middle Miocene) to temperate wet (Late Miocene–Pliocene) conditions, reflecting regional cooling and continuing uplift (Kuhlemann *et al.* 2008). The Late Cretaceous–Early Tertiary kaolinitic event with bauxite formation on the carbonate platforms of the Alpine belt is well identified in the sedimentary record, especially in western Europe (Thiry 2000; Kuhlemann *et al.* 2008). Additionally, the Hercynian basement was coated with thick kaolinitic palaeosols formed throughout the Cretaceous (Thiry 2000 and references therein). During the Early Neogene, corresponding to the first Alpine tectonic movements together with drying of the climate, the kaolinitic palaeosols were eroded, causing the onset of the most important detrital discharge of the whole Neogene in western Europe (Thiry 2000). In the Eastern Alps, reexhumation probably started in the Late Miocene and accelerated from the Pliocene (~2.7 Ma) onwards (Kuhlemann *et al.* 2002, 2008; Kuhlemann 2007; Willett 2010). According to Westaway (2002), long-term river terrace sequences indicate a global increase in uplift rates in the Late Pliocene, followed by a calm period and then a renewed increase around the Early–Middle Pleistocene boundary. Additionally, the amount of uplift in the Carpathians was greatest also in the Late Pliocene and Early Quaternary (Zuchiewicz 1998, 2009).

Red clays from Cenozoic palaeosols of the Eastern Alps record periods of stagnating uplift and decrease of relief (Kuhlemann *et al.* 2008). Exposure of kaolinitic palaeosols to temperate weathering conditions due to accelerating uplift may, therefore, have been fairly short in order to modify the mineralogy and chemical composition of previously formed clay minerals (e.g., Bronger 2007; Kuhlemann *et al.* 2008). Therefore, formation of the older type (Beremend Mb.) of the TRCF under humid subtropical climate during the Pliocene in the Alpine realm seems to be improbable. Consequently, kaolinite together with gibbsite in this type of the studied red clays may be inherited from pre-Pliocene lateritic soils, potentially formed under subtropical climatic conditions during the Eocene–Middle Miocene.

The younger member of the TRCF contains red (or “reddish”) clay beds. It contains relatively fresh material (illite, chlorite); the weathering products are predominantly smectite and goethite formed in a warm and dry climate in environmental conditions of savannah and steppe or forest steppe (Viczián 2007). The enrichment of resistant minerals such as quartz in the samples of the Tengellic Member indicates long-lasting semiarid weathering (Marsi 2000). Our data suggest that this type of red clay was developed under a warm-summer Mediterranean climate (*Csb*). This subtype of the Mediterranean climate experiences warm (but not hot) and dry summers, while winters are rainy and can be mild to chilly (Kottek *et al.* 2006). *Csb* climates are found in north-western Iberia, coastal California, and parts of the Pacific Northwest (Kottek *et al.* 2006).

The basal red clay layers of the Paks Loess Formation and KRCF contain similar material to the underlying red clays belonging to the younger member of the TRCF (Viczián 2007; Kovács *et al.* 2011).

They contain relatively abundant quartz and other detrital minerals. In both formations typical clay minerals

are well crystallised detrital illite and illite/smectite mixed layer minerals. The relative abundance of smectite and the smectite/illite ratio is significantly lower in loess samples relative to palaeosol and red clay samples, suggesting clear fluctuations in weathering intensity during the evolution of the Paks sequences (Újvári *et al.* in press). Additionally, relative abundances of smectitic material are higher in palaeosols, whereas the illite (illite ± muscovite) content is higher in loess. The apparent inverse behaviour of illitic material and smectite in the depth profile can indicate the transformation of illite into smectitic material in periods of soil formation. Less frequent clay minerals are smectite + kaolinite or vermiculite + chlorite, depending probably on slight climatic fluctuations during this period. The slightly but significantly lesser degree of weathering (more illite and chlorite, less smectite) indicates cooling of the climate. It is expressed in minor but clearly defined differences in the quantity of minerals (Viczián 2007). Based on the results, the climatic conditions were similar to those previously discussed. It was also *Csb*, but cooler with less precipitation.

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