Pedogenic calcrete records in southern Transdanubia, Hungary: a brief review with paleoenvironmental and paleogeographic implications

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In the Hungarian part of the Tisia block, four occurrences of rhizolith-bearing pedogenic calcrete have been published, three of them are located in southern Transdanubia. Nodular calcrete with beta fabrics was documented from the lower Permian (Cisuralian) continental Korpád Sandstone Formation where the subaerial exposure profile was developed on strongly altered volcanic shard-rich siliciclastic substrate. Additionally, two locations with *Microcodium*-bearing calcrete developed on lower Jurassic carbonate substrate were published in the last few years. The scope of this study is to briefly summarize pedogenic calcrete records known from the Permian to the Cenozoic of southern Transdanubia (Tisia block, Hungary), and to highlight their regional paleoenvironmental and paleogeographic importance.

Keywords: *calcrete, beta fabrics, subaerial exposure, Permian, Eocene, Hungary*

Introduction

Calcrete is a near-surface, terrestrial accumulation of predominantly calcium carbonate (Wright and Tucker 1991; Alonso-Zarza et al. 1998, Alonso-Zarza 2003). Calcretes which were formed only when evaporation exceeded precipitation, i.e. when the climate was at least seasonally arid, are generally considered as reliable palaeoenvironmental and palaeoclimatic indicators (Wright and Tucker 1991; Alonso-Zarza 2003; Alonso-Zarza and Wright 2010; Brasier 2011). According to the descriptive morphological terminology of Wright (1990), at the simplest level two end-member microfabric types occur in calcretes. Alpha calcretes (e.g. groundwater calcrete) consist of micritic to microsparitic groundmasses, typically with such features as crystallaria, floating skeleton grains, large euhedral crystals, crystal size mottling and displacive growth features. On the other hand, beta calcretes (e.g. pedogenic calcrete) exhibit microfabrics dominated by biogenic features such as rhizoliths (organo-sedimentary structures produced by roots; Klappa 1980), microbial tubes, alveolar textures, and *Microcodium*.

The genus *Microcodium* was originally created by Glück (1912) for aggregates of unusual shaped calcite crystals in Miocene marine deposits, and it was placed in the Codiaceae of the Chlorophyta. Two kinds of *Microcodium* were defined by Esteban (1972): '*Microcodium* a' refers to typical *Microcodium*, consisting of prismatic calcite crystals; whereas '*Microcodium* b' was defined by its smaller grain size and subquadrangular sections of its prisms (atypical *Microcodium*). Based on a large number of reports, the original interpretation of *Microcodium* as siphonaceous alga or

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any other phototrophic microorganism has been rejected; however, the process of *Microcodium* formation still remains unsolved (Klappa 1978; Alonso-Zarza et al. 1998; Košir 2004; Kabanov et al. 2008). *Microcodium* was interpreted as a calcification product of a mycorrhizae-cortical root cell association by Klappa (1978). Alonso-Zarza et al. (1998) suggested that ’*Microcodium* b’ formed through calcification of root cells. Similarly, Košir (2004) stated that morphology and structure of the typical *Microcodium* aggregates indicate that they formed through biologically controlled precipitation of calcium carbonate within the root cortical cells. On the other hand, Kabanov et al. (2008) proposed a non-rhizogenic biologically induced origin for typical *Microcodium* (’type a’) formation. According to these authors, it may be produced by actinobacterial or fungal substrate mycelia probably in association with other bacteria capable of consuming metabolites of the mycelial organism. Therefore, the typical *Microcodium* is a biologically induced mineralization driven by a saprotrophic microorganism or a microbial association decomposing dead terrestrial organic matter (rootlets and earlier generations of fungi, humified organic matter) (Kabanov et al. 2008).

Kabanov et al. (2008) provided an extensive review of the available literature for Paleozoic and Cretaceous–Cenozoic *Microcodium* occurrences. Unfortunately, however, there was no mention about Hungarian *Microcodium* records in that article. Among the paleosol sections in Hungary, calcretes from the Triassic cyclic peritidal–lagoonal platform carbonate succession and from the Pleistocene lacustrine setting of the Gerecse Hills, as well as Quaternary paleosols at the pediment of the Mátra Hills have received special attention (Bakacsi 1993; Bakacsi et al. 1994; Mindszenty and Deák 1999; Bajnóczi et al. 2006). *Microcodium*-bearing calcretes, however, have not been investigated in detail. On the other hand, pedogenic calcrete reports in the Hungarian part of the Tisia block published in English are rare (Lelkes 1994; Varga et al. 2002a, 2002b; Varga et al. 2012); and some papers intend to reach only a Hungarian readership (Varga 2000, 2002, 2009, 2011; Varga et al. 2002c). The scope of this study is to summarize pedogenic calcrete records known from the Permian to the Cenozoic of southern Transdanubia, Hungary, and to highlight their regional paleoenvironmental and paleogeographic importance.

Moreover, as *Microcodium* can be seen as a natural product of actinobacterial or fungal activity, corresponding to the nano- to microscale interaction between decomposing organic matter and a microbial association (Kabanov et al. 2008), this topic can trigger multidisciplinary attention of researchers interested in study of interactions of artificial nanostructures and biological systems.

Pedogenic calcrete records in southern Transdanubia, Hungary

In the central Carpathian–Pannonian area, the Mid-Hungarian line, a key element in the tectonics of the Intra-Carpathian area, subdivides the pre-Tertiary basement in two parts (Fig. 1A): Alcapa (Alpine–West Carpathian–Pannonian) block on the north and Tisia (Tisza–Dacia unit or Tisza Mega-unit) on the south (Csontos and Nagymarosy 1998; Csontos et al. 1992, 2002). In the Hungarian part of the Tisia block, four occurrences of pedogenic calcrete with *in situ Microcodium* or poorly preserved *Microcodium*-like structures have been published (Fig. 1B), three of them are located in southern Transdanubia.

Paleozoic (Permian) occurrence

In southern Transdanubia (Fig. 1C), the Permian siliciclastic and volcaniclastic rocks were deposited in continental strike-slip and rift-related basins, belonging to the internal part of the Variscan orogenic domain (Barabás and Barabásné Stuhl 1998; Vozárová et al. 2009). Among these units, the fine-grained siliciclastic deposits of the Cisuralian Korpád Sandstone Formation are distinctive in terms of presence of nodular calcretes with rhizocretions (Varga 2009; Varga et al. 2012).

The alluvial Korpád Formation occurs in the subsurface in southern Transdanubia and ranges up to 700 m in thickness, consisting of polymictic conglomerate, breccia, sandstone, and mudstone (Fazekas 1987; Barabás and Barabásné Stuhl 1998). This formation contains a sparse Early Permian macroflora (e.g., *Pecopteris*, *Voltzites*) and a lowermost Permian microflora composed of the *Potonieisporites* and *Vittatina* assemblage (Barabás and Barabásné Stuhl 1998). Calcretes and calcareous paleosols were not recognized previously in this unit; however, a large amount of individual dolomite concretions and concretionary aggregates together with animal burrows were described by Jámbor (1964) from the red siltstone samples (drill core 9015, near the village of Dinnyeberki, Mecsek Mts.; Fig. 1C). Varga (2009) and Varga et al. (2012) reported that these carbonate concretions are, at least partially, of rhizogenic origin, representing nodular horizons of calcrete profiles.

The studied subaerial exposure profile developed on strongly altered volcanic shard-rich siliciclastic substrate where calcium could be derived from the hydrolysis of volcanic glass and plagioclase feldspar. Calcrete microfabric is characterized by the presence of micritic mottles, root traces (e.g., rhizocretions and smaller root casts; Fig. 2), and associated biogenic structures such as faecal peloids (rounded micrite pellets resulting from invertebrate defaecation in the soil) and relic structures of *in situ Microcodium*-like aggregates (in the sense of Klappa 1980 and Kabanov et al. 2008). Unfortunately, *Microcodium* appears as partly to totally recrystallized calcite grains, so primary morphology could not be identified. The rhizocretions are complex tubular structures up to 1 cm in diameter, with a wall structure of irregular micritic laminae which form roughly concentric layers around the central hollow filled by drusy calcite spar cement (Fig. 2B). In the pedogenic micritic laminae of rhizocretions, carbonate is replaced by tiny authigenic quartz showing clear evidence for localized silicification (Fig. 3).

Eocene Microcodium corroding Jurassic (Pliensbachian) substrate

Eocene calcrete-sourced pebbles were found in a lower-middle Miocene conglomerate sequence (Szászvár Formation) in the western part of the Mecsek Mts. (Fig. 1C). Based on evidence from spore and pollen remains (Varga 2000; Varga et al. 2002a, 2002b, 2002c), local geology (Wéber 1982, 1985) and an ancient analogue (Gierlowski-Kordesch et al. 1991), the reworked calcrete seems to be related to the continental sequence of the middle Eocene–early Oligocene(?) Szentlőrinc Formation. Rocks of this unit are not exposed in Hungary; they were penetrated in significant thickness by boreholes in southern Transdanubia (Wéber 1982, 1985; Varga et al. 2004).

The depositional environment of the Szentlőrinc Formation is interpreted as an alluvial system containing a carbonate–clastic alluvial plain (floodplain environment), locally with brown coal seams (Wéber 1982, 1985; Varga et al. 2002a, 2004). The provenance area surrounding the basin comprised Paleozoic (e.g., metamorphic and acidic volcanic fragments, fine-grained sandstones) and Mesozoic (e.g., Triassic limestone and dolomite, Jurassic limestone and marl clasts, Cretaceous limestone clasts derived from the Mecsek-Villány zone, and altered basic volcanics) rocks supplying the clastics (Varga 2002; Varga et al. 2002a, 2004). The basement, partially, constituted a supposed karst system correlated with the Jurassic Szársomlyó Limestone of the Villány area delivering bedload and dissolved carbonates into the sedimentary basin (Varga et al. 2004).

The beige colored reworked calcrete consisting of sand and gravel-sized clasts coated by laminated micrite could develop on carbonate rock substrate associated with an alluvial fan system containing fragments of Mecsek-type lower Jurassic rocks (Varga 2000; Varga et al. 2002c). Calcrete microfabrics reveal coated grains, peloids, rhizoliths, alveolar textures, and well preserved *in situ Microcodium* aggregates (Fig. 4). The root structures are mostly represented by root casts and rhizocretions but root petrifaction also occurs. Some root casts are associated with alveolar texture and *Microcodium*. In most cases the isodiametric to elongated *Microcodium* grains with characteristic opaque inclusions and uniform extinction distinctly cluster in aggregates and radiate from central cavities filled with limpid calcite spar cement. The aggregate shape is generally „corn-cob" or cylinder with rosette appearance in transverse sections, corresponding to the typical *Microcodium*. In the studied calcrete, peloids and coated grains are also very frequent (Fig. 5). Ooids and pisoids are micritic, rather indistinctly laminated, and have cores of carbonate rock fragments, rarely bioclasts, calcite spar or detrital quartz grains. Additionally, a few coated grains with cores of root-cast fragments, *Microcodium* grains or intraclasts have been recognized in the calcrete samples, which indicate that the horizon formed through multiple phases of brecciation resulted from penetrative growth of roots and subsequent cementation by micrite (Varga et al. 2002b, 2002c).

Origin of the reworked calcrete clasts is unknown, however, pisolithic and brecciated horizons are common at the top of calcrete profiles where intense brecciation favors the formation of calcrete-sourced clasts (Alonso-Zarza 2003). Alternatively, Wórum (1999) suggested a late Oligocene compressional stress field in the area studied, so the occurrence of calcrete-sourced pebbles in the lower Miocene conglomerate layers may reflect the tectonically controlled physical erosion of the Szentlőrinc rocks.

Pedogenic calcrete developed on lower Jurassic (Sinemurian) carbonate substrate

In a narrow belt between the villages of Ófalu and Zsibrik, southern Transdanubia (Fig. 1B), occurrences of Jurassic rocks are known in an elongated tectonic zone at the SE part of the Mecsek Mts. (details see in Császár et al. 2007). Relevant part of this Jurassic sequence belongs to the so-called Zobákpuszta Sandstone Formation (formerly a part of Vasas Marl Formation; Raucsik 2012) which is composed of alternating gray sandstone and sandy, calcareous siltstone, claystone and marl beds with gray sphaerosiderite concretions and *Gryphaea* coquinas. The formation has a general fining upward character and the average carbonate content of the sediments increases upsection suggesting transgressive depositional dynamics. Based on its sedimentary structures and fossil assemblage (e.g., bivalves, foraminifers, ostracods, and echinoderm fragments), the formation must have been deposited in littoral to shallow sublittoral zone of a marine basin with normal salinity during the Sinemurian (Császár et al. 2007).

In the study area, characteristic features of some ancient calcrete formation are restricted to few small outcrops developed over mixed carbonate–siliciclastic sandstones and conglomerates (L4, L12, and L16 outcrops; Császár et al. 2007), showing week to moderate degree of actual surface weathering. The altered substrate is characterized by corroded siliciclastic grains (quartz, K-feldspar, and rare muscovite and zircon) with circumgranular calcite rims (Fig. 6) and, sometimes, root structures represented by micrite- and/or microspar-filled curved channels, alveolar-septal structures, and probably *in situ* corrosive *Microcodium*. Their aggregates enclose internal areas filled with micrite cement (Fig. 6D). In this area, moderately preserved *Microcodium-like* structures of elongate calcite prisms (measuring 50–80 μm in length and 10–20 μm in width) has been reported only from outcrop L16 (Varga 2011).

Jurassic rocks here are locally covered by shallow marine Miocene, Pannonian or Quaternary deposits with unconformities between them (Császár et al. 2007); however, the calcrete formation event has not been dated.

Paleoenvironmental and paleogeographic significance of calcrete records

Rhizoliths and related features are products of pedodiagenesis; therefore they are indicators of palaeosols and hence of subaerial vadose environments in ancient successions (Klappa, 1980). During pedogenesis, root respiration produces significant levels of CO_2 (and hence of carbonic acid), additionally, microbial decomposition also releases CO_2 that controls the dissolution and precipitation of pedogenic carbonate (Wright and Tucker 1991; Alonso-Zarza 2003; Brasier 2011). All workers studying *Microcodium* in relation to the environment of its formation agree on its biogenic subsurface non-marine nature (e.g., Klappa 1978; Kabanov et al. 2008). Two peaks in *Microcodium* abundance occur at the Moscovian–early Permian and latest Cretaceous–Paleogene intervals which are distinct by low pCO_2 in the atmosphere (Berner 2006; Royer 2006; Kabanov et al. 2008). Most of the accumulations of *Microcodium* occur within continental depositional settings that are affected by pedogenesis and/or calcrete formation within palustrine, fluvial, and, rarely, karstic settings. Furthermore, *in situ* occurrence of *Microcodium* aggregates in shallow marine facies always indicates subaerial exposure and pedogenic modification of the sediment (Košir 2004). On the other hand, calcretes as indicators of unconformities could be used in subsurface stratigraphy, providing one more element of basin-architecture analysis in buried deposits (Alonso-Zarza and Wright 2010).

With respect to the lower Permian (Cisuralian) nodular calcrete belonging to the Korpád Formation, micromorphological features mentioned above together with the mineralogy suggest a relatively dry climate with low amount of rainfall (100–500 mm/year) during pedogenesis (Varga et al. 2012). This result supports that the southern Transdanubian part of the Tisia block was located in the arid paleoclimatic belt during the early Permian. Interestingly, however, another calcrete record has never been documented in the Cisuralian continental sequences of the Tisia (see Vozárová et al. 2009, 2010, and references therein). Permian deposits from the Apuseni Mountains (Romania) developed also in continental facies with molassic characteristics where red beds are dominant (Seghedi et al. 2001) but basal black-colored and bituminous shaly deposits suggest somewhat more humid climate. On the other hand, considered in a

Circum-Pannonian context, the depositional basin of the Korpád Formation is well correlated with the Permian basins of the Western Carpathians and the Eastern Alps, Alcapa. The Cisuralian sediments of the Zemplinicum (the Cejkov and Černochov Formations) were deposited in an alluvial fan setting alternating with floodplain or ephemeral lake deposits with calcrete horizons, all showing the typical features of semiarid/arid climatic conditions (Vozárová et al. 2009, 2010). Additionally, calcrete horizons together with lenses of dolomite and gypsum occur locally in the Hronicum Cisuralian sequence (the Malužiná Formation) comprising a thick succession of alternating conglomerates, sandstones and shales deposited in braided alluvial and fluvial-lacustrine environments under a semiarid/arid climate (Vozárová et al. 2009, 2010; Vdačný et al. 2013). Furthermore, in the Drauzug and in the Gurktal Nappe (the Laas Formation and the Werchzirm Formation, respectively), proximal to distal alluvial red-beds, grading into fine-grained sandflat-playa complexes locally with calcrete crust, characterize the Cisuralian (Vozárová et al. 2009, 2010).

In contrast to the early Permian subaerial exposure profile, only sporadic information exists for correlation of younger Transdanubian pedogenic calcretes developed on lower Jurassic substrates. The aforementioned correlation of the Eocene microflora-bearing calcrete-sourced gravels found in the lower Miocene Szászvár conglomerate sequence with the Szentlőrinc Formation is somewhat speculative, because of insufficient petrographic and biostratigraphical control. Additionally, we are unaware of any *Microcodium* findings in Paleogene successions from the Hungarian part of the Tisia block. It is important to note, however, that *Microcodium* is most extensively reported from the latest Cretaceous–Eocene of the Mediterranean regions (Kabanov et al. 2008, and references therein). Corresponding to the Circum-Pannonian area, excellently documented subaerial exposure surfaces, including calcretes with typical *Microcodium*, occur within a succession of upper Paleocene and lower Eocene shallow-marine limestones (the Trstelj Formation and Alveolina-Nummulites Limestone) in southwestern Slovenia (Košir 2004), so our result fits well to the younger peak of *Microcodium* abundance in a global context.

Finally, regarding the pedogenic calcrete developed on lower Jurassic (Sinemurian) carbonate substrate, the calcrete formation event has not been dated. Apart from the aforementioned Eocene unconformity, there are several Neogene and Quaternary subaerial exposure events with calcrete formation in the study area. Calcretes of shallow-marine carbonate systems have been described from the Sarmatian sediments from various parts of Hungary (Fig. 1B). According to Lelkes (1994), sedimentation on a restricted carbonate platform was interrupted by subaerial periods and pedogenesis during the Sarmatian. The diagnostic features for pedogenic calcrete facies identified from 27 core sections are rhizoliths (root moulds, root casts, root tubules, rhizocretions, and root petrifactions), alveolar textures, rare typical *Microcodium* (core Soltvadkert–4 only), calcified insect eggs, peloids, and various types of pedogenic voids (Lelkes 1994). Based on the neighboring location sites (Fig. 1B) the beta calcrete developed on lower Jurassic carbonate substrate could probably form during the Sarmatian subaerial exposure event. Nevertheless, other cycles separated by unconformities reflecting subaerial vadose environments have also been mentioned in the Neogene to Quaternary basin fill (e.g., Bajnóczi et al. 2006) so the time of the calcrete formation developed on the Sinemurian substrate and, therefore, its paleoenvironmental relationship cannot be estimated in a satisfactory manner.

Concluding remarks

This paper summarizes Hungarian examples of calcified plant roots and *Microcodium* from subaerial exposure profiles in southern Transdanubia, Tisia block, which are characterized by root-influenced fabric but differ in form and stage of development. Three occurrences of rhizolits together with *in situ Microcodium* or poorly to moderately preserved *Microcodium*-like structures related to calcrete profiles have been documented in the area studied. These are the followings: nodular calcrete belonging to the lower Permian (Cisuralian) Korpád Sandstone Formation (Western Mecsek Mts.), Eocene calcrete-sourced clasts developed on Jurassic substrate from the Miocene Szászvár conglomerate sequence (Western Mecsek Mts.), and calcrete of uncertain age developed on carbonate-siliciclastic substrate of the Sinemurian Zobákpuszta Sandstone Formation in a narrow belt between the villages of Ófalu and Zsibrik (Eastern Mecsek Mts.).

Interestingly, in a Circum-Pannonian context, Permian calcrete-bearing sediments have not been reported from the Apuseni Mountains (Tisia), but the depositional basin of the Korpád Formation can be correlated to the Permian basins of the Western Carpathians (e.g., Zemplinicum and Hronicum) and the Eastern Alps (e.g., Drauzug and Gurktal Nappe), Alcapa. In contrast to the Cisuralian subaerial exposure profile, only sporadic information exists for correlation of younger Transdanubian calcretes developed on Jurassic substrates. According to our opinion, if calcretes as indicators of unconformities receive more attention in the area of the Tisia block they may be used more efficiently in surface/subsurface stratigraphy as well as for regional paleoenvironmental and paleogeographic correlations.

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Figure captions

Fig. 1. A) Major tectonic units of the Carpathian–Pannonian area after Csontos et al. (1992, 2002). B) Location map of the pedogenic calcretes from the Hungarian part of the Tisia block. 1 = Permian calcrete (Varga et al. 2012); 2 = Eocene calcrete-sourced clasts (Varga et al. 2002b); 3 = Calcrete developed on Sinemurian substrate (Varga 2011); 4 = Calcrete developed on Miocene (Sarmatian) substrate (Lelkes 1994). C) Structural framework and generalized geological map of the Western Mecsek Mts. (Konrád and Sebe 2010). 1 = Neogene; 2 = Jurassic and Cretaceous; 3 = Triassic; 4 = Upper Permian – Lower Triassic; 5 = Palaeozoic in general; 6 = observed fault; 7 = supposed fault; 8 = observed reverse fault; 9 = supposed reverse fault; 10 = strike-slip fault; 11 = syncline; 12 = anticline.

Fig. 2. Photomicrographs of petrographic thin sections of the Permian calcrete (A–C: sample 15.22.1; D: sample 15.22.2). A–B) Rhizocretion with complex tubular structure. Smaller second order rhizocretions and root casts are obvious around the well developed central rhizocretion, suggesting vertical root systems with taproot and laterals (A: Plane-polarized light; B: Crossed nicols). Smaller root casts are indicated by arrows. C) Strongly recrystallized *Microcodium*-like calcite aggregates with relics of dark finely

dispersed inclusions in close vicinity to rhizoliths. Plane-polarized light. D) In surroundings of the second order rhizocretions small pellets are present. Plane-polarized light.

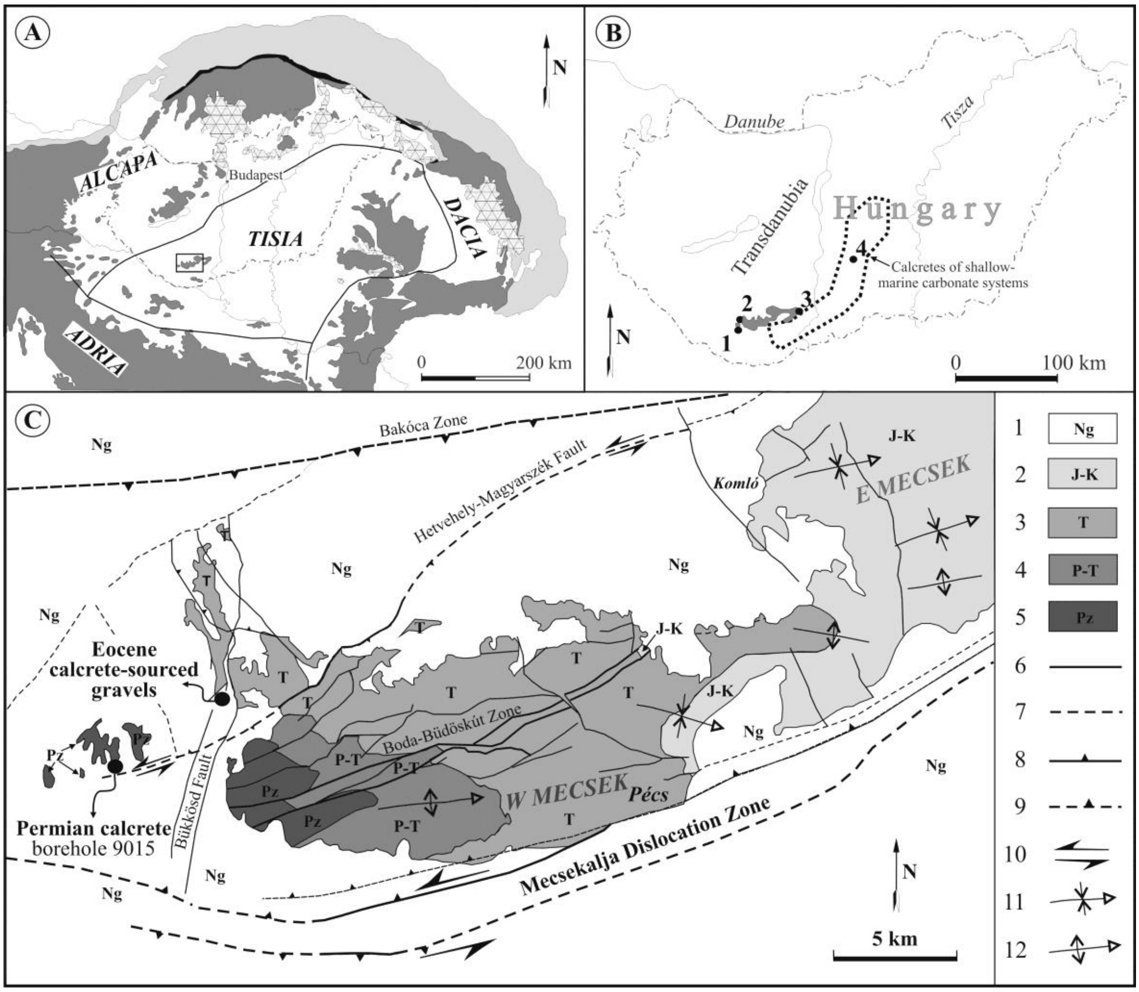
Fig. 3. Photomicrographs of petrographic thin sections of the Permian calcrete (sample 15.22.1). A–D) In the pedogenic micritic laminae of rhizocretions (Rh), carbonate is replaced by tiny authigenic quartz having euhedral crystal terminations (arrows). Plane-polarized light.

Fig. 4. Eocene calcrete developed on lower Jurassic carbonate substrate. A) Polished slab of the Eocene calcrete-sourced gravel derived from the brecciated horizon of a partially or totally eroded calcrete profile. Highly irregular to subrounded clasts coated with micritic laminae composed of predominant Pliensbachian limestone fragments. Root-induced brecciation is evidenced by rhizolith (arrow). Coin for scale: 23 mm. B) *In situ Microcodium* aggregates within root channel. Plane-polarized light. C) Typical *Microcodium* („rosette") structure in peloidal calcrete. Plane-polarized light. D) *Microcodium* aggregate with characteristic opacity of individual grains due to fine inclusions. Plane-polarized light.

Fig. 5. Eocene calcrete developed on lower Jurassic carbonate substrate (Plane-polarized light). A–B) Spar cement filled root casts with *Microcodium* aggregates. C–D) Calcrete peloids (sand-silt sized micritic grains) and indistinctly laminated, micritic vadose pisoids. Some pisoids have cores of carbonate rock fragments (e.g. bioturbated packstone–wackestone with sponge spiculae and echinoderm fragments; arrow).

Fig. 6. Photomicrographs of pedogenic calcrete developed on lower Jurassic (Sinemurian) carbonate substrate (Plane-polarized light). A–C) Floating siliciclastic (Q: quartz and K: K-feldspar) grains coated by displacive asymmetric to circumgranular cement rim showing a typical feature of calcretes. The remaining void spaces are filled with micrite or microspar. Root-structures are indicated by arrows. D) Weekly recrystallized *Microcodium*-like calcite aggregates composed of a single layer of individual, elongate prismatic crystals of calcite.

Fig. 1.


A
N
ALCAPA
Budapest
TISIA
DACIA
ADRIA
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200 km
B
Danube
Tisza
Transdanubia
Hungary
Calcretes of shallow-marine carbonate systems
1
2
3
4
N
0
100 km
C
Ng
Bakóca Zone
Hetvehely-Magyarszék Fault
Komló
E MECSEK
J-K
T
Eocene calcrete-sourced gravels
Pz
Permian calcrete borehole 9015
Bükkösd Fault
Boda-Büdöskút Zone
P-T
W MECSEK
Pécs
Mecsekalja Dislocation Zone
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4 P-T
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Fig. 2.

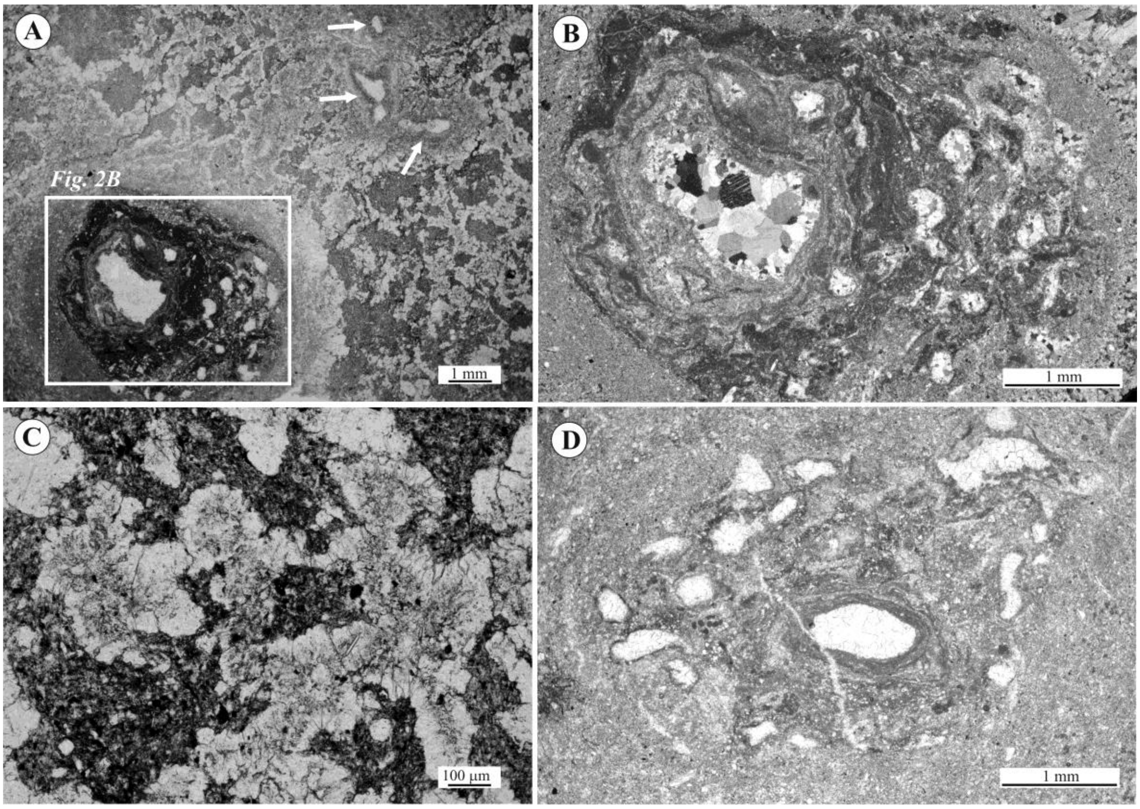

Fig. 3

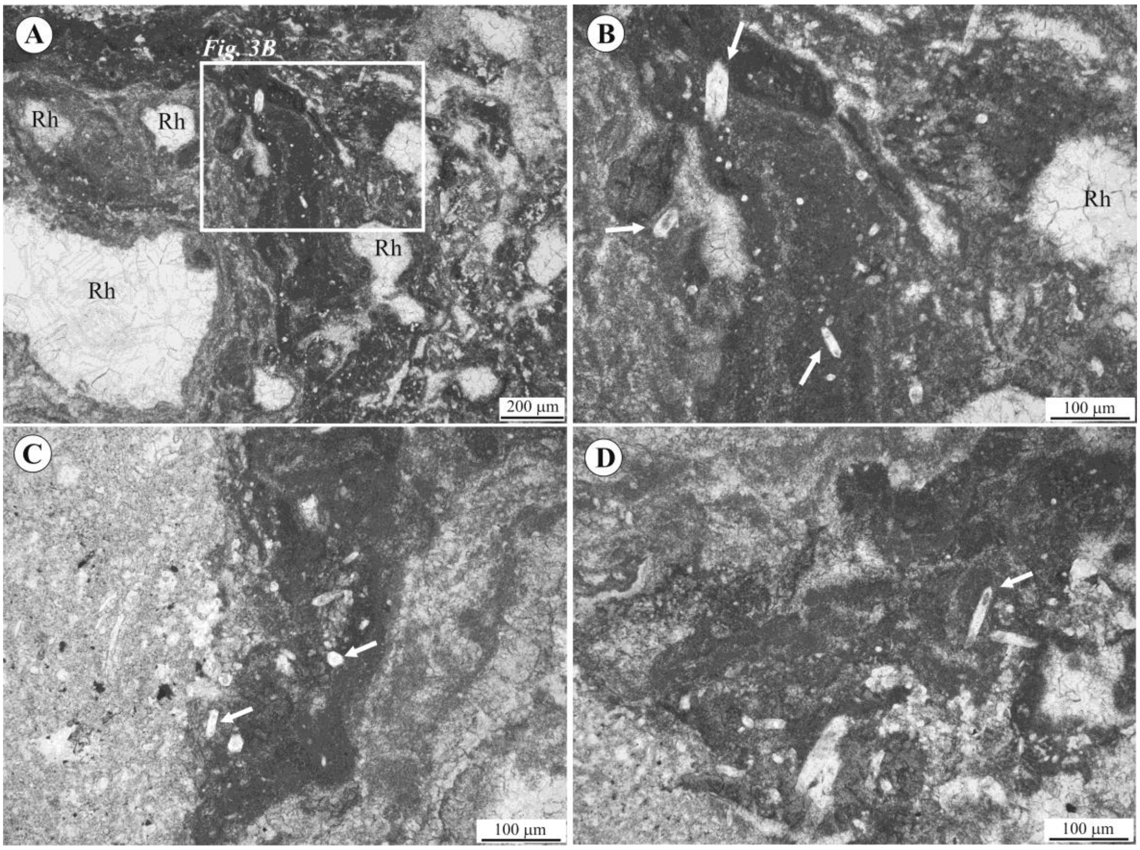

Fig. 4.

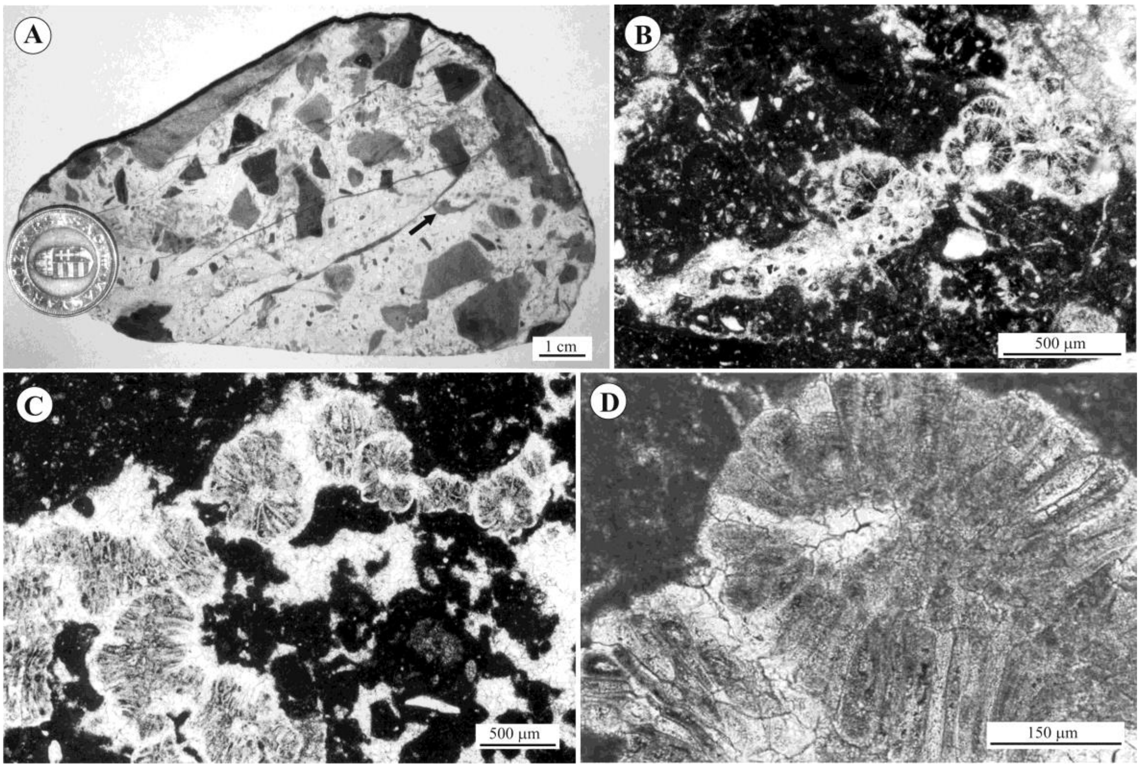

Fig. 5

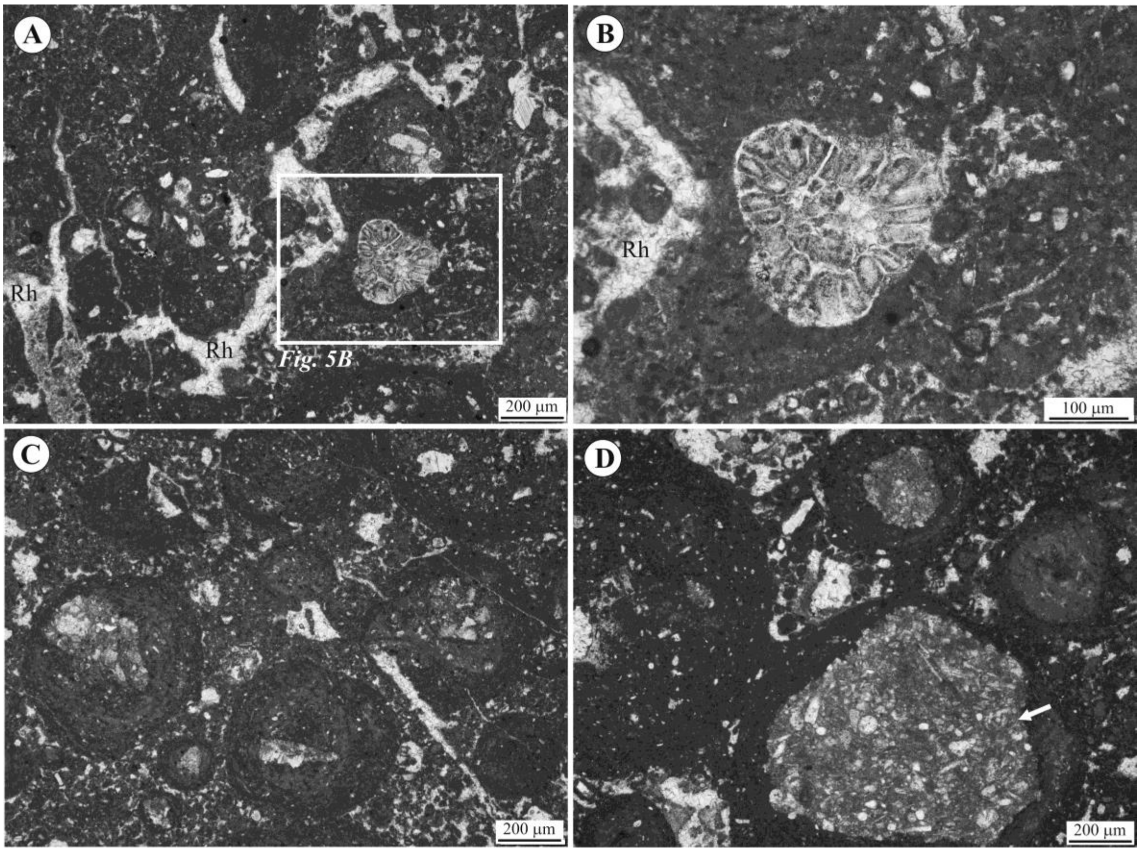

Fig. 6