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Characteristics, origin and geoarchaeological significance of carbonate concretions from the early Bronze Age locality „Orovački vinogradi“, (Bjelovar, Croatia)

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ABSTRACT

“White horizon” archaeological units consisting of carbonate concretions occur in the archaeological settlement of “Orovački vinogradi”, located near Bjelovar (NW Croatia). During archaeological excavation these horizons were considered to be the inhabitation-levels within the historical layering profile. Further sedimentological and mineralogical research and comparison of the profile with an anthropogenically unaffected site nearby (“Ciglenska strana”) required re-evaluation of this interpretation. These horizons of carbonate concretions are interpreted as the result of diagenetic processes developed within the Upper Pontian sequence of sands, clayey sands and sandstones. A sedimentary-diagenetic model is suggested for their formation. These results had consequences for the archaeological evaluation of the settlement and influenced further investigations at the locality.

Keywords: carbonate concretions, diagenesis, geoarchaeology, Bronze Age, Upper Pontian Vučedol-culture, Vinkovci-culture, Croatia

1. INTRODUCTION

When trying to problem solve in field investigations, it is often necessary, and useful for both disciplines, to relate geology to archaeology. Results of such multidisciplinary research are mostly published jointly by the disciplines, and are known as geoarchaeological results. In this article an example of the mutual application of archaeological and geological methodology will be shown. Research activities were conducted on an early Bronze Age locality of the Vinkovci-culture (2100–1900 BC), named “Orovački vinogradi”, located near Bjelovar (North-Western Croatia). The site was first investigated according to archaeological methodology, followed by later geological investigation in order to solve some problems of interpretation. A similar approach is often used but the opposite approach is also known (archaeological methods after geological investigation), GOLDBERG & MACPHAIL (2006).

Archaeological research at the locality resulted in numerous multidisciplinary data, relevant for understanding everyday life at the beginning of the Bronze Age in North-

Western Croatia (JAKOVLJEVIĆ, 2005, 2007). The settlement of the Vinkovci-culture period (2100–1900 BC), which was the one of the successors of the great Vučedol-culture, is so far the only settlement from this period systematically investigated in North-Western Croatia. Research indicates aspects of the everyday life of the settlement inhabitants. Valuable material discoveries including ceramics, bone and stone artifacts have been made. The results also testify to the ethnological and spiritual origin of the inhabitants as part of an extensive civilization that spread from the Aegean to the Baltic Sea (JAKOVLJEVIĆ, 2005).

"White horizon" archaeological units observed in the investigated locality occur as an interesting and puzzling phenomenon. Their characteristics and origin are described and their geoarchaeological significance is discussed. Carbonate concretions within the profile are interpreted and a model for their formation is proposed. Both the results and the model influenced the archaeological interpretation and evaluation of the site and future excavation activities were directed according to the geological interpretation.

2. ARCHAEOLOGICAL AND GEOLOGICAL SETTING

The "Orovački vinogradi" archaeological settlement is located 15 km west of Bjelovar in North-Western Croatia, on a small hill within the forest (Fig. 1). It is located within Upper Pontian clastic sediments, consisting mostly of sands and sandstones (KOROLIJA & CRNKO, 1985; KOROLIJA et al., 1986).

The settlement is situated on a plateau with three steep sides covered with dense vegetation (beech and hornbeam forest), while the southern side is flat and covered by meadows (Fig. 2). Such a location of the settlement aided preservation of the cultural layers, immediately below the humic horizon (mostly occupation deposits – dug out objects and waste pits) (JAKOVLJEVIĆ, 2007).

In an area of approximately 300 m^2, about 40 different archaeological stratigraphic units were investigated and registered, (including traces of inhabitation objects, layers with bones and ceramic finds etc.). Archaeological data on the "horizontal stratigraphy" (the spatial arrangement of the archaeological objects and finds) obtained during the excavation provided the ground-plan of the settlement (Fig. 2). Collected data on the "vertical stratigraphy" (the vertical arrangement of finds in the archaeological units revealed during excavation), suggest the basic chronology of the settlement. After the archaeological excavation, two questions still remained: (i) how to precisely define the basic chronology of the settlement; and (ii) of what type were archaeological objects in the settlement - traditional buried dug out objects vs. combined dug out objects and surface objects. In order to answer these questions, geological research of the "white horizon" archaeological units proved very useful.

Some archaeological stratigraphic units (i.e. "hard rock" archaeological units – Fig. 3a) are found to be archaeologically negative. Rounded and triangle-shaped palisade traces (Figs. 3b and 3c), which probably defined constructed shelters, have been discovered in different vertical archaeologi-

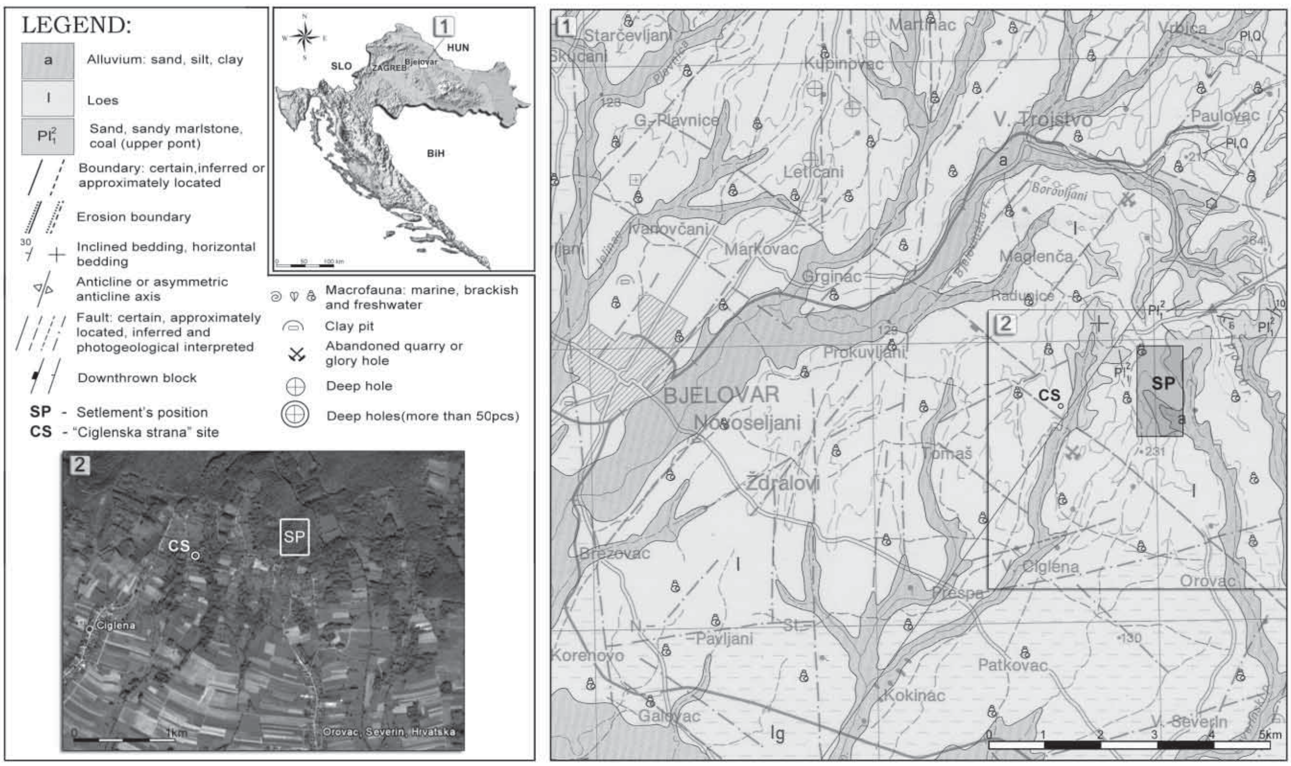


Figure 1: Location map, based on the Basic Geological Map of Croatia, 1:100 000, sheet Bjelovar (KOROLIJA & CRNKO, 1985; KOROLIJA et al., 1986).

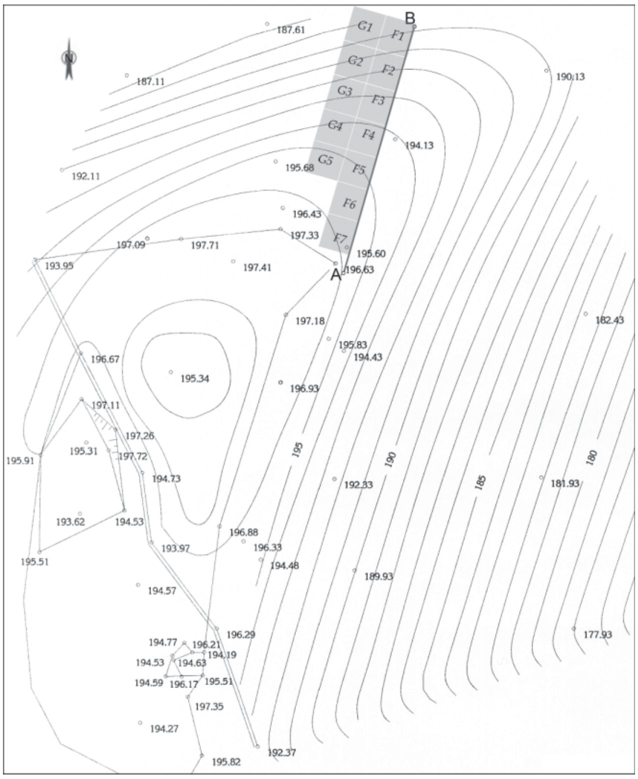


Figure 2: Topographic map of the archaeological settlement located on the hill slope. Archaeological probes are marked G1-G5 and F1-F7 and distributed along the profile of the settlement (A–B). Scale 1:1 000.

cal stratigraphic units. The origin of "white horizon" archaeological units – Figs. 3d and 3e) remained puzzling.

They consist of semi-consolidated fine-grained, white marly material (Fig. 4) and were considered during excavation as the inhabitation-levels within the historical layering profile. The puzzling nature and origin of these archaeological units temporarily suspended further excavation at the locality. Therefore, answers to questions concerning their nature (anthropogenic vs. natural-geological), origin (syn-sedimentary, diagenetic, epigenetic etc.) and their archaeological significance for the future treatment of the locality (conservation of the settlement vs. continuing excavation) depended on geological research of the profile.

At the near-by site of "Ciglenska strana", (approximately 800 m away – CS on Fig. 1), at the same altitude and on the slope of the nearby hill, there is a vertical outcrop with an almost identical layering sequence in the lower part of the outcrop (containing Upper Pontian deposits). The top of the outcrop contains younger loess deposits and is indurated and covered with vegetation. It is claimed that this outcrop was "anthropogenically unaffected" and formed solely by natural (geological) processes. Observations made on this outcrop served for comparison and as a tool to understand the anthropogenically affected site "Orovački vinogradi".

3. METHODOLOGY

Sedimentological field-work methodology (after STOW, 2005) was undertaken at the locality in order to determine the lithology and to describe any sedimentary textures and structures. Particular representative layers were sampled for petrographic and mineralogical analysis and to precisely characterize the mineralogical and petrological composition of the "hard rock" and "white horizon" archaeological units. All results are summarized in the geological column (Figs. 5 and 6a–d). The same sedimentological field-work methodology (after STOW, 2005) and sampling procedure were also conducted at the "Ciglenska strana" site (Fig. 7), in order to compare results with the "Orovački vinogradi" site.

Thin-sections of characteristic samples from the well-lithified sandstones were prepared for petrographic and XRD analysis. Thin-sections of sandstone samples (samples SJ-17 and CS-2) were stained with "Alizarin Red S" and K-ferricyanide and analyzed in plane-polarized light. Samples were described and interpreted in terms of the rock classification after FOLK (1980), PETTIJOHN et al. (1987), TUCKER (2001) and TIŠLJAR (2004).

Whole-rock samples of the "white horizon" archaeological units (samples SJ-24, SJ-27 and CS-1) and sandstones (samples SJ-20 and CS-2) were analyzed by X-ray powder diffraction (XRD). XRD-patterns were taken using a Philips diffractometer (Cu$K\alpha$ radiation, U = 40 kV, I = 20 mA) with graphite monochromator and proportional counter. Samples were treated with 5% acetic acid and the diffraction patterns of air dried and variously treated (with glycerine; with ethylene glycol; 2 hours heated on 600ºC; dissolved for 24 hours in warm 18% HCl) insoluble residues are also recorded.

4. RESULTS

During the archaeological excavation we examined a 7 m thick, undisturbed, vertical succession, consisting of sand and sandstone beds and horizons with white concretions. Individual brownish-yellow sand beds are 0.24–1.40 m thick (Fig. 5). Thicker sand beds occur in the lower part of the geological column. The main structural characteristic of the sand beds is plane parallel lamination. In the upper part, well-lithified sandstones up to 5 cm thick (so-called "hard rock" archaeological units) occur. Sandstones alternate with thick-bedded unlithified sand. Boundaries between the sands and sandstones are sharp and wavy. Within some of the sand beds, (usually in the upper part of the column), an upward increase in clay content was observed. Such an increased clay content accompanied the "white horizon" archaeological units.

Sand beds consist of fine- to medium-grained sand (0.1–0.5 mm grain size) (Figs. 6a–d). The main mineral component is quartz with subordinate feldspars and micas. The amount of clay minerals and Fe-oxides varies. In some of the beds these components are concentrated in discrete laminae on the top of the normally graded beds.

Sandstones from the "Orovački vinogradi" site ("hard rock" archaeological units) are predominantly composed (> 60 %), of angular, well-sorted quartz grains. Feldspars and lithic fragments, (cherts and magmatic rocks), (Figs. 8a and 8b) are subordinate components. Between these three main types of grains there is a significant amount of inter-

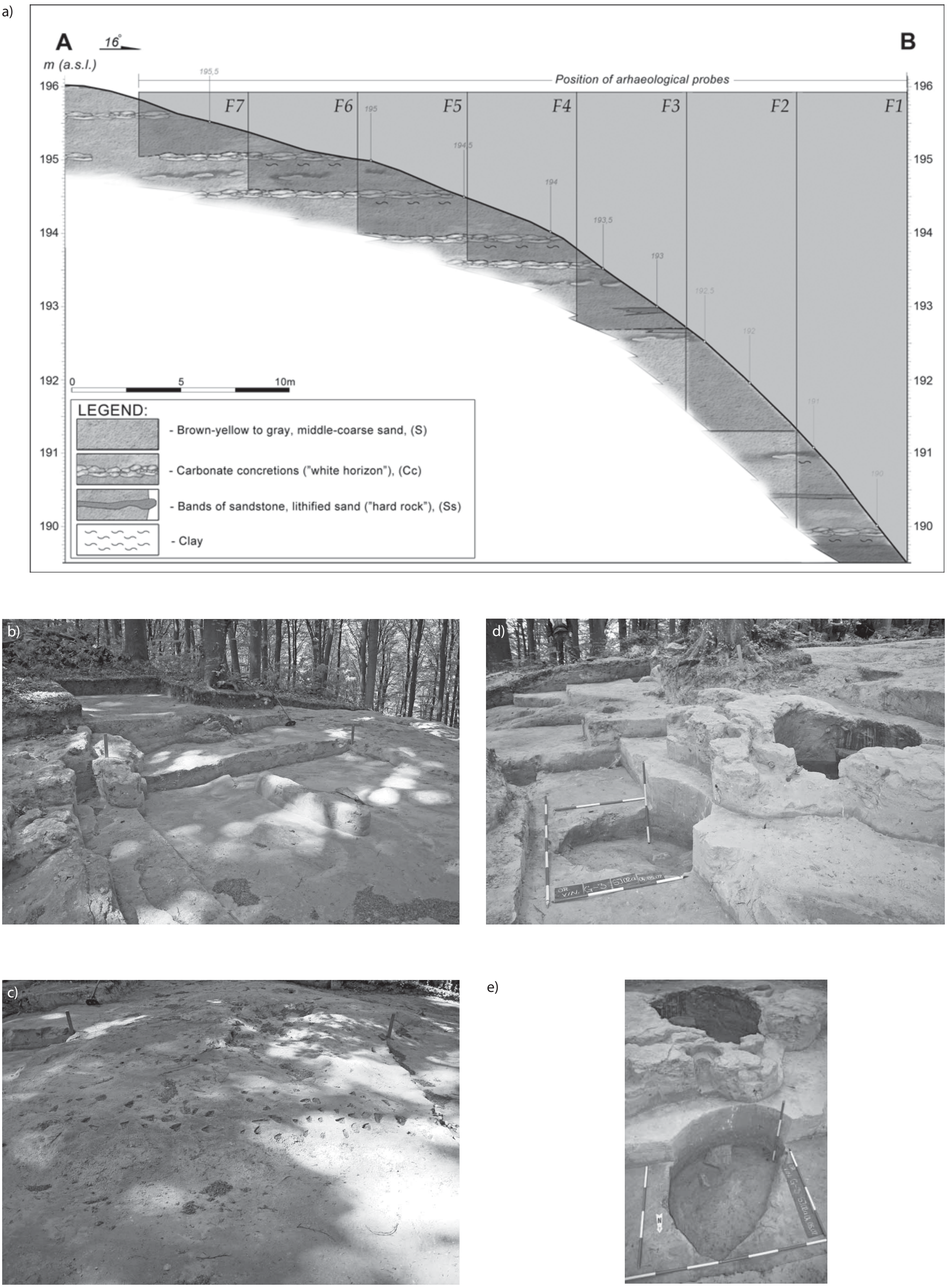


Figure 3: a) Geological profile A–B along the settlement (see Fig. 2); b) Archaeological probe F4 with rounded and triangle-shaped palisade traces; c) Archaeological probe F4 with rounded and triangle-shaped palisade traces; d) Archaeological probe F4 containing "white horizon" archaeological units; e) Archaeological probe F4 containing "white horizon" archaeological units.

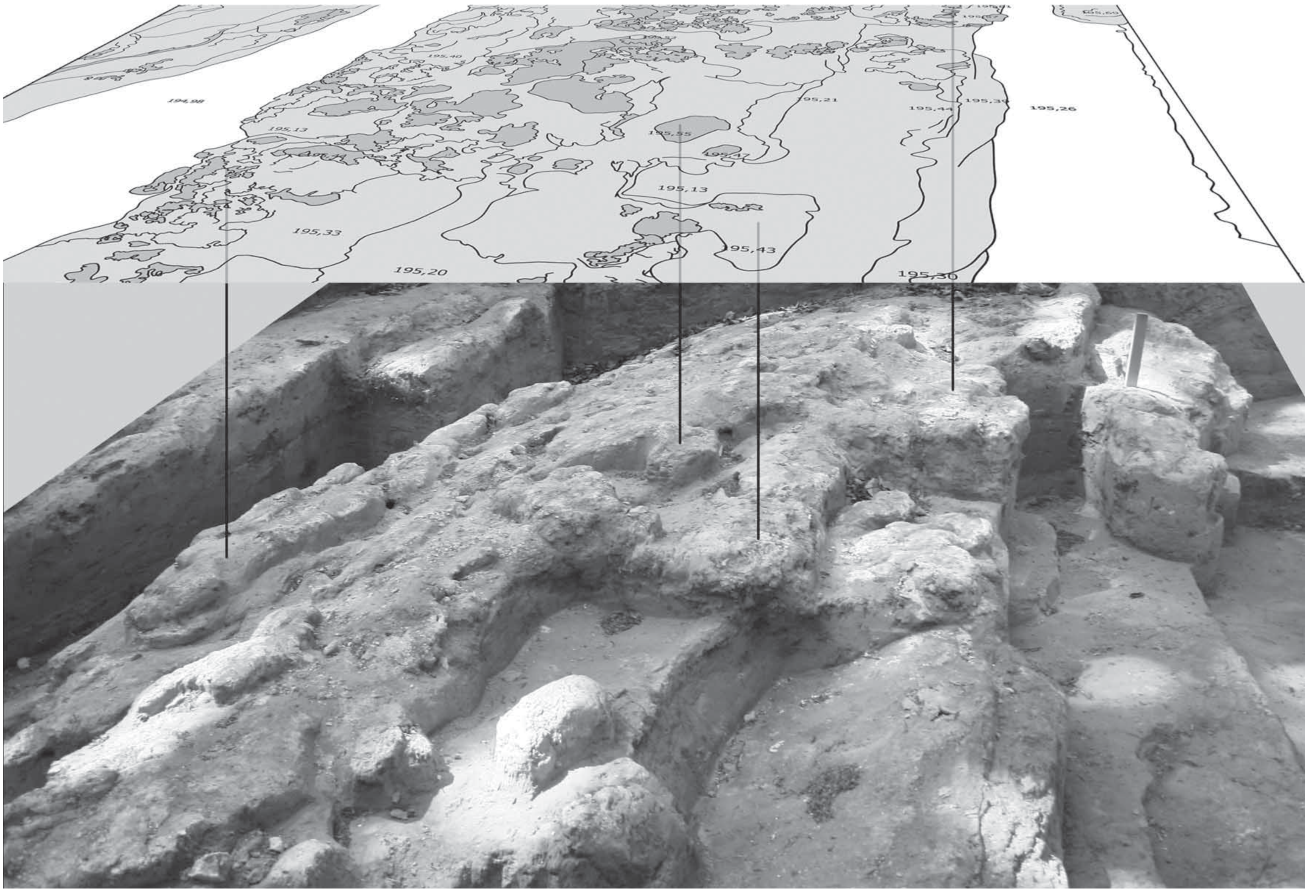


Figure 4: Photography and ground-plan of the archaeological probe F-6. "White horizon" archaeological units are marked with lines on the photography (bellow) and with dark grey areas on the ground-plan of the probe (above).

locked flakes of micas (mostly detrital muscovite), without obvious orientation of flakes in parallel laminae. All grains are well-cemented with sparry calcite. There is minimum primary or secondary porosity (almost none). Sandstones have been determined as micaceous sublitharenites. Semi-quantitative mineral analysis (XRD) of the sandstone samples (samples SJ-20 and CS-2 in Table 1) showed similar mineral composition, considering recalculation of the calcite and dolomite as the cementing minerals. The composition (defined by petrographic and semi-quantitative XRD analysis) of sandstone samples collected at the "Ciglenska strana" locality are almost identical to those at the "Orovački vinogradi", and therefore their origin can be compared (compare Figs. 8a and 8b, and samples SJ-20 and CS-2 in Table 1).

Archaeological stratigraphic units, the so-called "white horizon" archaeological units that randomly appear in the vertical section "Orovački vinogradi" and Ciglenska strana consist of spherical, (up to 5 cm in diameter), and partly elongated, (up to 15 cm long and from 5 to 10 cm wide) white concretions, which are mostly coalesced into semi-consolidated layers. Coalescence of these concretions can be seen on the upper bedding planes of these horizons that were exposed during archaeological excavation, and preserved as possibly significant vertical archaeological stratigraphic units. Internally, the texture of the concretions showed no obvious nucleus for concretionary growth (e.g. fossil or some lithic fragment). They contain silty particles and clay, showing thin ovoid lamination that continues from the sand beds (Figs. 9 and 9b). Finer quartz grains, together with some clay material are incorporated into the concretions. The frequency of concretion layers in the geological column coincides with the increase in clay content in the sand beds below, which indicates a significantly close relationship between the two features. Semi-quantitative mineral analysis (XRD) for sev-

Table 1: Results of the semi-quantitative mineralogical analysis (XRD) for the representative concretions samples (samples CS-1, SJ-24 and SJ-27) and sandstone samples (samples SJ-20 and CS-2) from the both sites ("Orovački vinogradi" site – SJ-samples; "Ciglenska strana" site – CS-samples). Cal: calcite; Dol: dolomite; Qtz: quartz; Pl: plagioclasses; Kfs: K-feldspars; M: micas; Kln: kaolinite; Chl: chlorite; Chl-Vrm: chlorite-vermiculite; S: smectite; AC: amorphous component; +: mineral present in the sample; -: mineral not present in the sample; ?: mineral questionably present in the sample.

Sample	Cal	Dol	Qtz	Pl	Kfs	M	Kln	Chl	Chl-Vrm	S	AC
CS-1	72	-	5	3	+	8	+	?	6	+	+
SJ-24	72	5	4	3	?	6	+	-	4	+	+
SJ-27	82	5	2	+	+	3	?	?	+	?	+
SJ-20	40	12	14	15	+	12	+	+	3	-	+
CS-2	43	-	22	17	4	6	-	+	+	-	+

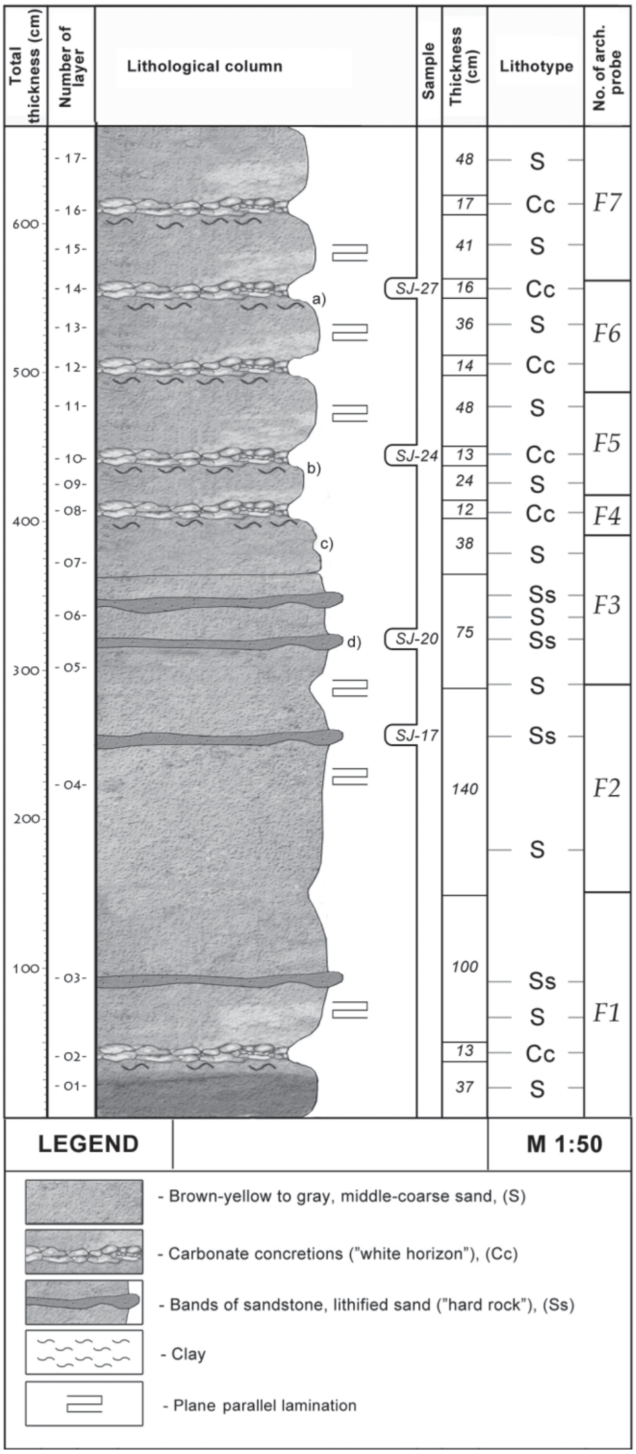


Figure 5: Geological column in the site „Orovački vinogradi".

eral samples of concretions from both sites ("Orovački vinogradi" and "Ciglenska strana") showed very similar mineralogical compositions (samples CS-1, SJ-24 and SJ-27 in Table 1). The concretions are predominantly calcareous, containing between 72% and 82% calcite, up to 5% dolomite, < 8 % feldspars (both plagioclases and K-feldspars), < 8% micas, clay minerals (predominantly chlorite-vermiculite and smectite, chlorite and kaolinite) and also some amorphous material.

5. INTERPRETATION AND DISCUSSION

Results of the sedimentological and mineralogical research revealed that the investigated profile consists of sands, clayey sands and sandstones and that it is formed by deposition of sedimentary material. The major enigma during the archaeological investigations was the presence of unusual "white horizon" archaeological units, cropping out frequently along the vertical profile. During the geological field work, it was discovered that they have been formed by the development and coalescence of concretions in particular horizons. These processes are discussed and a possible model for their formation is inferred below.

Concretions are usually formed by various combinations of processes, under different sedimentary and geochemical conditions. Concretion morphologies strongly depend on the physico-chemical conditions operating during their formation (SEILACHER, 1991). Their mineralogical compositions are often considered to be an important factor for the development of morphologically different concretion types (SELLES-MARTINEZ, 1996). Mineral composition varying from mostly calcite, siderite and silica, together with concretions morphologies, can be indicative of their origin (syngenetic, diagenetic or epigenetic). Examples of epigenetic concretions formed by meteoric groundwater are calcareous concretions in loess, or sideritic concretions in desert sandstones. The former are also known as "loess dolls", and sometimes are even wrongly named as "Neolithic mother goddesses". Concretions from the investigated profile do not match that type of epigenetic concretions because of their different morphology, fabric and composition, together with the characteristics of the host sediments, which are not aeolian sediment (loess). Diagenetic concretions (pre- or post-compaction), usually contain calcite, dolomite or silica, depending on the material available from the surrounding sediment. Concretions can be formed around some nucleus or, more commonly, without any nucleus, along particular horizons, reflecting a level at which super-saturation of pore-waters was achieved (TUCKER, 2001). Super-saturation of the pore-waters is often achieved when the clay content dispersed in the host sediment increased, usually forming a kind of vertically impermeable screen. Concretions may also be preferentially developed in horizons with increased organic carbon, compared to the surrounding sediments. In both of the investigated sites, an increased amount of clay was noticed immediately below the "white horizon" archaeological units, suggesting their close relationship. Therefore localized precipitation from adequate pore-waters under suitable geochemical conditions controlled the mineralogy and growth rates of concretions. In the case of carbonate concretions from the "Orovački vinogradi" and "Ciglenska strana" sites, the available material from the surrounding sediment, (predominantly quartz sands with micas, occasionally cemented with sparry calcite cement, thus forming micaceous sublitharenites) was most-likely calcite precipitated from pore-waters. Finer quartz grains, together with some clay material were incorporated in particular horizons during growth of the concretions.

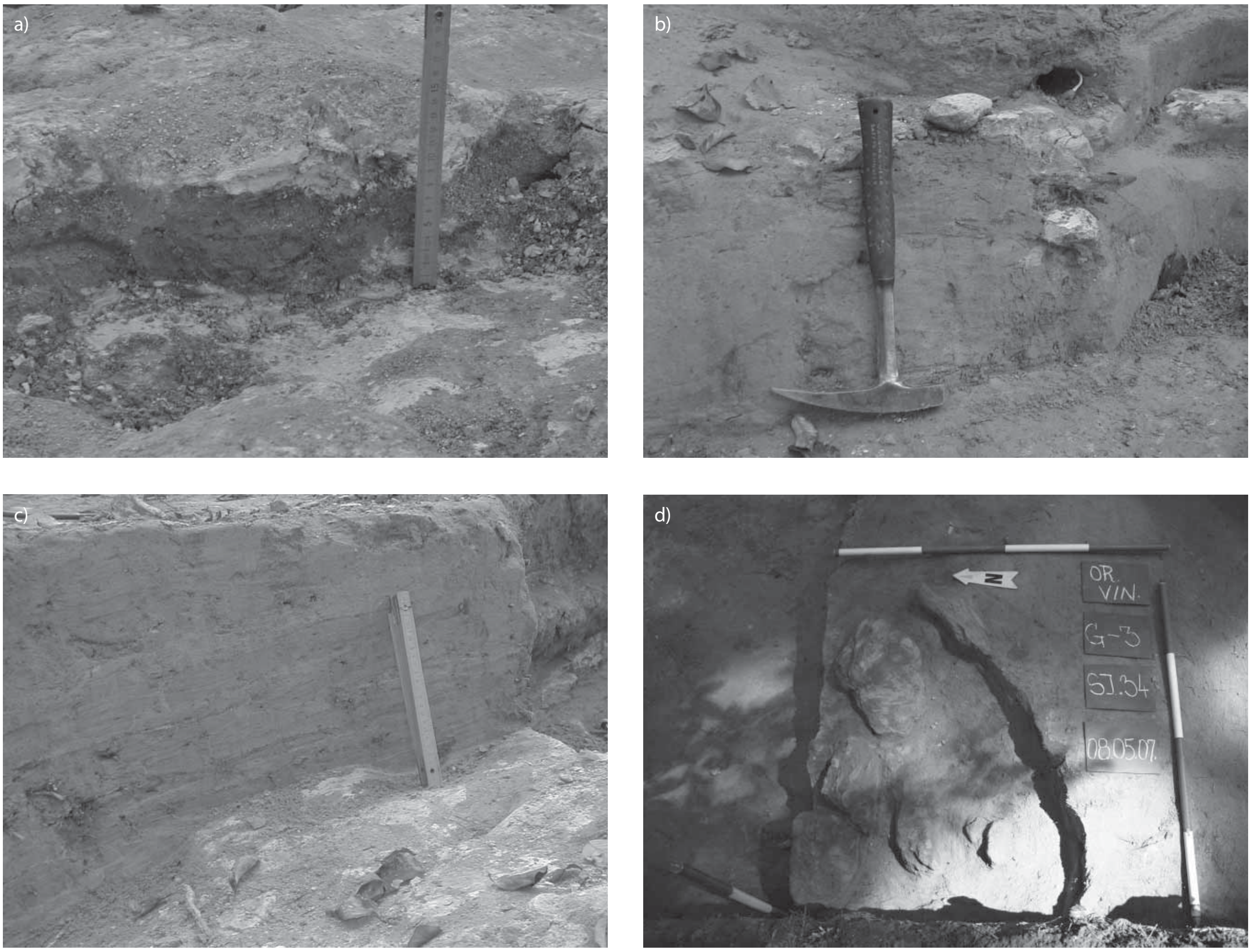


Figure 6: a) "White horizon" unit. Ruler (20 cm) for scale (see in Fig. 5); b) Individual carbonate concretions within "white horizon" unit. Hammer (33 cm) for scale (see in Fig. 5); c) Plane parallel lamination in the sand bed. Ruler (20 cm) for scale (see in Fig. 5); d) "Hard rock" unit (sandstone). Scale bar segments are 10 cm (see in Fig. 5).

In terms of concretion morphology, all concretions usually occurred as two radically different morphospaces, either as dendrites or as Liesegang rings (SEILACHER, 2001). Both morphotypes result from the distribution and percolation of pore fluids that contain dissolved ions within the sediment. However, their appearances are not the subject of this article. Instead, we would like to pay attention to the most commonly appearing concretion morphologies – ovoid and

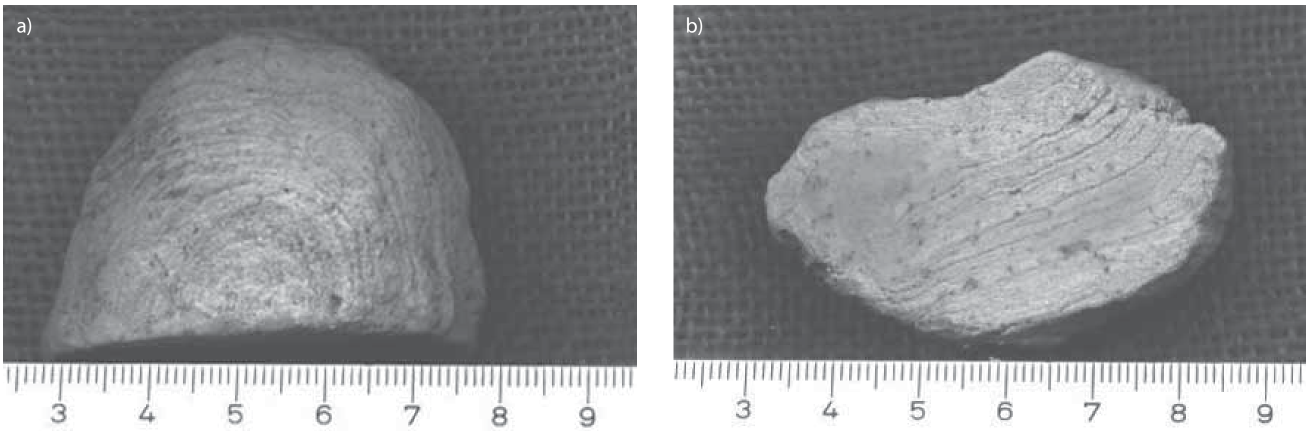
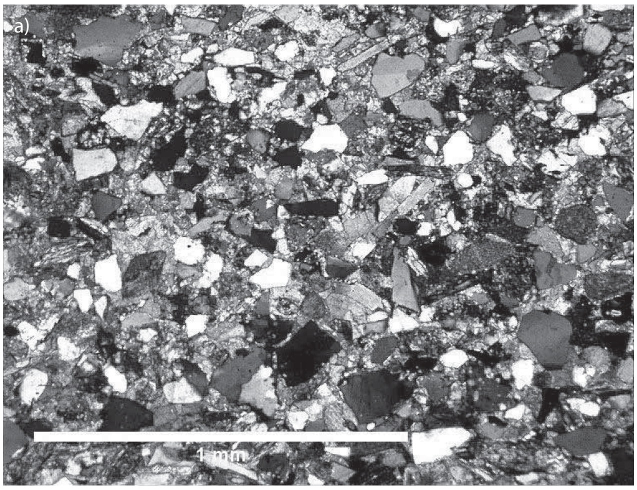


Figure 7: a) Internal structure of the concretion – continuing lamination from the sand bed. View from the top of the concretion. Scale bar is in centimetres; b) Internal structure of the concretion – continuing lamination from the sand bed. Cross-cut of the concretion. Scale bar is in centimetres.



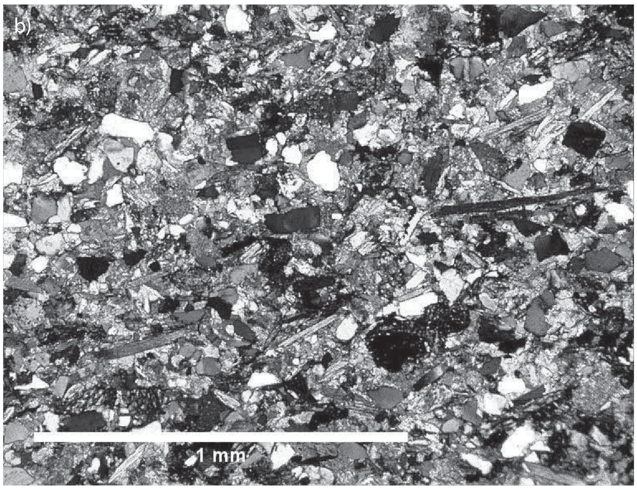


Figure 8: Microphotographs of the representative sandstone samples from "Orovački vinogradi" (sample SJ-17 – Fig. 8a) and "Ciglenska strana" sites (sample CS-2 – Fig. 8b)). Both samples are determined as micaceous sublitharenites with angular, well-sorted, (0.1 – 0.5 mm of grain size) of quartz grains that predominates, feldspars and micas and with some lithic fragments (cherts and magmatic rocks). Grains were cemented with spary calcite cement. a) Sample SJ-17 (Sample stained with Alizarin red S and K-ferricyanide); b) Sample CS-2. (Sample stained with Alizarin red S and K-ferricyanide).

globular (SCOTCHMAN, 1991; COLEMAN & RAISWELL, 1995), because similar morphologies appear in the investigated archaeological sites. Concretionary growth is the result of radial concentric precipitation of material and carbonate is usually supplied by diffusion from the surrounding sediment. If supersaturated ground-water flowed past the growing calcite concretion, dissolved material could be added not only by diffusion, but by mass flow. However, very low groundwater flow-rates have a relatively low influence on the growth rate of concretions (BERNER, 1968, 1971). Elongate concretions may be sometimes preferentially oriented, reflecting the direction of high pore-water movement (McBRIDE et al., 1994). The observed concretions are sometimes elongate, but they do not show any evidence to indicate the possible direction of pore-water movement.

An increased clay component in some horizons helped precipitation and concretions preferentially developed along these horizons suggesting circulation of water through the bed, and then horizontally along an impermeable clayey screen. Concretions are therefore interpreted as diagenetic – they developed after deposition of the sand beds due to the circulation of pore water precipitating calcite around some invisible cells. Geometries of the concretions are the result of the host lithologies, their fabric and their characteristics. The most important characteristics are the permeability and anisotropy of the host sediment. An important step in concretion geometry analysis and interpretation of their formation was to explain the interference of concretionary growth and compaction. Spherical growth reflects isotropic sediments (i.e. sand or mud), ellipsoidal growth reflects the vertical reduction of permeability caused by compaction, and so-called *bread-loaf* geometries (with a flattened lower side) reflects a steep compaction gradient (SEILACHER, 2001). Lensoid geometries of the concretions, in which the original lamination pinches out towards the edges within the calcified part, implies the pre-compaction phase of concretionary growth. Similarly, the absence of pinching out of the original lamination and the presence of a hard nucleus with the general symmetry indicate a post-compaction phase of concretionary growth (SEILACHER, 2001; STOW, 2005).

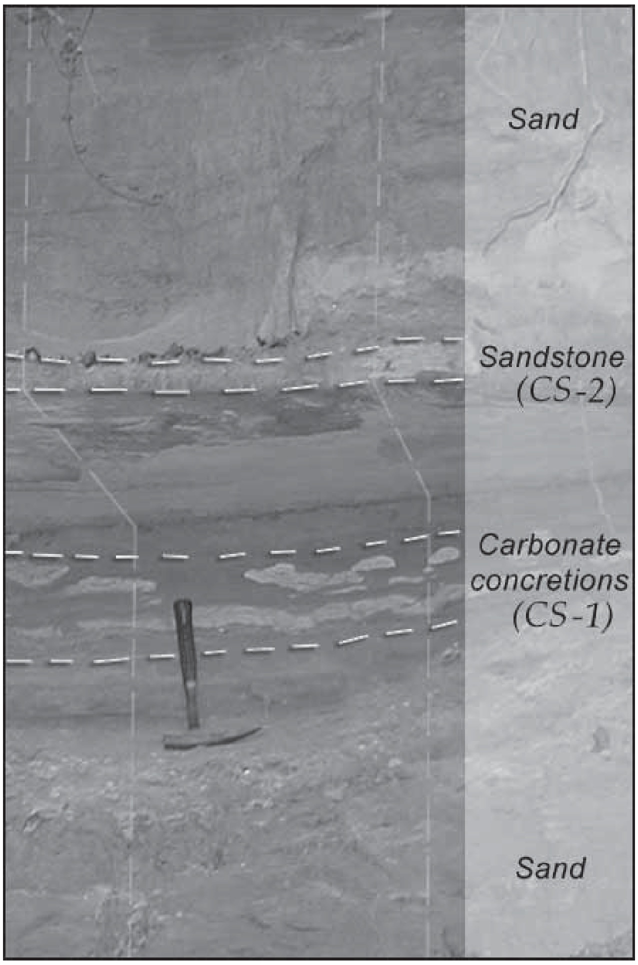


Figure 9: Vertical profile in the anthropogenically unaffected site „Ciglenska strana". Hammer (33 cm) for scale.

Timing of concretion development is determined from analysis of their geometry and their geometric relationships with the surrounding beds. Continuation of depositional laminae from the sand beds to the concretions was observed, as well as slight pinching of the lamination in the surrounding sands towards the edges of concretions. Concretions show ovoid geometry, which again suggests development during the diagenetic phase, (after sedimentation of the overlying sand bed). Concretions grow from pore-waters super-saturated with carbonate, along anisotropic horizons enriched in clay. The main processes were diffusion followed by laminar pore-water flow along partly developed concretions. Carbonate concretions in all horizons, at both sites, are therefore interpreted as having developed in the pre-compaction stage of diagenesis. However, the geometries of some elongated concretions observed in the profiles also suggest a post-compaction influence seen in the morphology of the concretions. Considering the relatively small volume of overlying sediments (only a few metres or even a few decimetres), together with their semi-lithified nature, a post-compaction influence can be considered as the minor control for development of the observed concretions. Consequently, the pre-compaction factors (precipitation caused by super-saturation of pore-waters along the particular horizons enriched in clay component), can be considered as the major control for development of the concretions.

All the above-mentioned data can be summarized in the 4-stage diagenetic model of carbonate concretion development during the pre-compaction phase of diagenesis (Fig. 10):

1. Deposition of a sedimentary succession consisting mostly of sand beds with enrichment in clay particles along particular horizons.
2. Development of concretions by diffusion from carbonate-rich pore waters. The source of carbonates was the surrounding sediment, (sand with carbonate-rich pore-waters). Concretions grow by mineral precipitation around anisotropic centres within horizons enriched in clay.

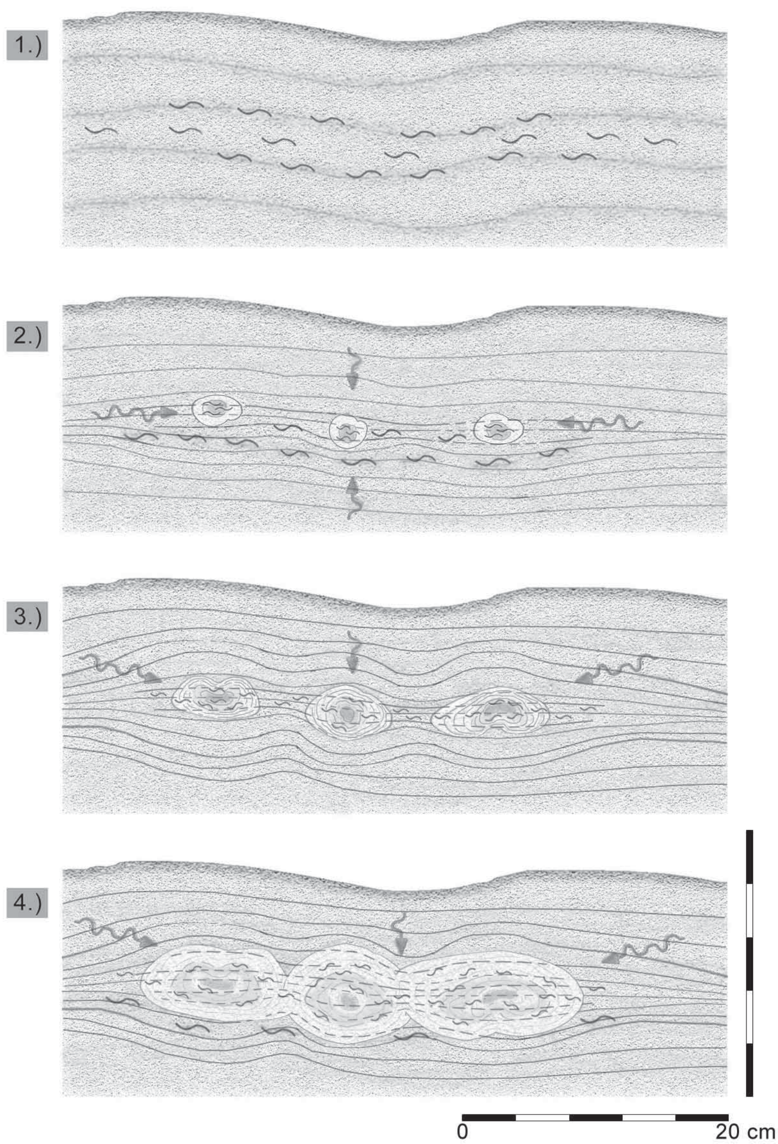


Figure 10: Model for diagenetic development of carbonate concretions in the pre-compactional phase of diagenesis. Stage 1) Development of sedimentary column and enrichment in clay particles along the particular horizons. Stage 2) Beginning of the concretionary growth by diffusion. Stage 3) Further development of ovoidal geometry of the concretions by laminar pore-water flow along partly developed concretions. Stage 4) Coalescence of the concretions in a single semi-consolidated layer and the formation of the "white horizon" archaeological units.

3. Further growth of the concretions by pore-water circulation along partly developed concretion horizons. Crystallization of cortices covering preformed parts of concretions and preservation of original lamination from sand beds. Partial incorporation of silty material from the surrounding sediment, (quartz, feldspars and micas).
4. Coalescence of concretions into a single semi-consolidated layer and formation of the "white horizon" archaeological units. Minor post-compaction influence on the concretion geometries.

6. CONCLUSIONS

According to the results of the field-work presented here, and comparison of the analysis from the two investigated localities with Pontian sediments, – archaeological profile at "Orovački vinogradi" site and geological profile at "Ciglenska strana" site – several conclusions about the characteristics, origin and geoarchaeological significance of the observed carbonate concretions can be made:

a) The investigated profile on the archaeological site "Orovački vinogradi" comprising beds of sand, clayey sand, sandstone and "white horizons" archaeological units, developed by natural (geological) processes of sedimentation and diagenesis.
b) The "white horizons" archaeological units consist of carbonate concretions, developed by precipitation of carbonate from super-saturated pore-waters, along particular anisotropic sand beds enriched in clay.
c) Morphologies of the carbonate concretions and their geometric relationships with the surrounding sediments indicate their development in the pre-compaction phase of diagenesis.
d) The main processes involved in development of the concretions were diffusion and subsequent pore-water movement along previously developed parts of the concretions.
e) Coalescence of the concretions in semi-consolidated layers, and the formation of the "white horizon" archaeological units within the investigated archaeological profile, gives a false impression of anthropogenic affected horizons. The interpretation presented here and the geoarchaeological significance of these concretions indicates continuing excavation on the site, without the specific need for conservation of these horizons.
f) The given interpretation and model for the origin of the observed carbonate concretions suggesst the need for a final re-evaluation of basic chronology of the settlement, provided by archaeological data on the "vertical stratigraphy".
h) The definition of types of archaeological objects in the "Orovački vinogradi" settlement will be less complicated, considering the "white horizon" archaeological units are naturally developed and coalesced carbonate concretions.

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