



AKADÉMIAI KIADÓ

# Occurrence and significance of smectite in the Pliensbachian (Lower Jurassic) at Lókút (Bakony Mts., Hungary)


Central European  
Geology

64 (2021) 1, 8–17

DOI:

10.1556/24.2021.00002

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Received: April 16, 2020 • Accepted: December 14, 2020

Published online: March 20, 2021

ORIGINAL RESEARCH  
PAPER



## ABSTRACT

Although the Mesozoic rocks of the Transdanubian Range have been the subject of a multitude of different studies, mineralogical research is largely underrepresented. The clay mineralogy of Lower Jurassic (especially the Pliensbachian and Toarcian) strata was broadly investigated earlier; however, systematic high-resolution clay mineralogical studies remain scarce. Here we present a mineralogical study focusing on the Upper Pliensbachian strata of the Lókút-Hosszúárok section, located near the Eplény Manganese Ore Field. We identified dioctahedral smectite, randomly interstratified illite/smectite, illite as 10 Å phyllosilicate, quartz and cristobalite. Based on our new results we propose that the smectite was formed by aging of Mg or Fe hydroxide-silica precipitates. The smectite and cristobalite were presumably formed from the siliceous tests of radiolarians, whose abundance was controlled by a local upwelling system. The occurrence of Pliensbachian smectite in the Lókút outcrop shows similarities with the Úrkút smectites known from both Pliensbachian and Toarcian strata, which implies that similar processes controlled the sedimentation during the Pliensbachian as well as during the black (gray) shale-hosted ore accumulation in the Eplény and Úrkút basins.

## KEYWORDS

Toarcian oceanic anoxic event, Transdanubian Range, Lókút, smectite

## INTRODUCTION

Jurassic sedimentary strata of the Transdanubian Range are dominated by carbonates and have been the broader focus of scientific research. However, from a mineralogical aspect, beside the carbonate minerals, only a few studies have been devoted to accessory minerals; investigation of clay minerals has been scarce. An exception is the comprehensive study of [Viczián \(1995\)](#) who carried out a large-scale clay mineral investigation in the entire Transdanubian Range but only at a low stratigraphic resolution. In the last decade, [Polgári et al. \(2013\)](#) and [Leskó et al. \(2019\)](#) reported new results from topical investigations in a small area but at high spatial resolution. Clay mineralogy is remarkably suitable for reconstruction of depositional environments, because the formation of the different clay minerals is well-defined, and such information will complement and enhance our previous knowledge from other studies. We conducted a clay mineralogy study at the Lókút-Hosszúárok outcrop (“long trench” near the village of Lókút), a stratigraphic key section in the Bakony Mts. ([Konda, 1970](#)), which is located close to the Eplény manganese ore deposit. We investigated the occurrence of clay minerals within the Pliensbachian strata, with a special focus on the occurrence of smectite.

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Previously, smectite was only known to occur in the Lower Jurassic of the Transdanubian Range in the in the Úrkút manganese ore field and within the footwall of this deposit (Viczián 1995; Polgári et al. 2013; Leskó et al. 2019). The other economically significant manganese ore deposit in the Transdanubian Range occurs near Eplény, and previous comparisons of the Úrkút and Eplény deposits assumed similar genetic models. The hypothesis that their formation process was the same implies that the depositional environment and the sedimentary strata should be the same or similar (Konda, 1970; Polgári et al., 2000; Szabó, 2006; Bíró et al., 2012; Horváth et al., 2014; Bíró & Pál-Molnár, 2015; Papp et al., 2015; Papp & Zajzon, 2016; Papp & Zajzon, 2019). To test this, we investigated the occurrence of smectite in the Pliensbachian limestone at the Lókút-Hosszúárok outcrop, because of its proximity to the Eplény manganese deposit.

### Geologic setting

The study area is located in the Bakony Mts., (Transdanubian Range, TDR), Hungary (Fig. 1). The TDR belongs to the Alps-Carpathians-Pannonian Basin region, to the ALCAPA Mega-unit (Csontos et al., 1992), with an original paleogeographic position reconstructed as between the Upper Austroalpine and the South Alpine lithosphere fragments (Kázmér & Kovács, 1985; Haas et al., 1995, Gawlick et al., 1999; Csontos & Vörös, 2004). The Mesozoic sedimentation in the entire TDR was influenced by the opening of the western part of the Tethys Ocean (Fig. 2). The Mesozoic sedimentary cycle as preserved in the TDR began with a latest Paleozoic transgression. By the end of the Triassic an extensive, coherent, few km-thick Dachstein-type platform developed (Haas, 1988; Haas & Budai, 1995; Haas et al., 1995; Balog et al., 1997; Haas & Budai, 1999; Haas, 2004; Péró & Haas, 2004). The carbonate platform started to disintegrate near the Triassic-Jurassic boundary because of the continuing rifting and spreading of the Neotethys Ocean. In the Early Jurassic the extensive Triassic carbonate platform was fragmented; by the late Early and Middle Jurassic a dissected submarine topography of “seamounts” and

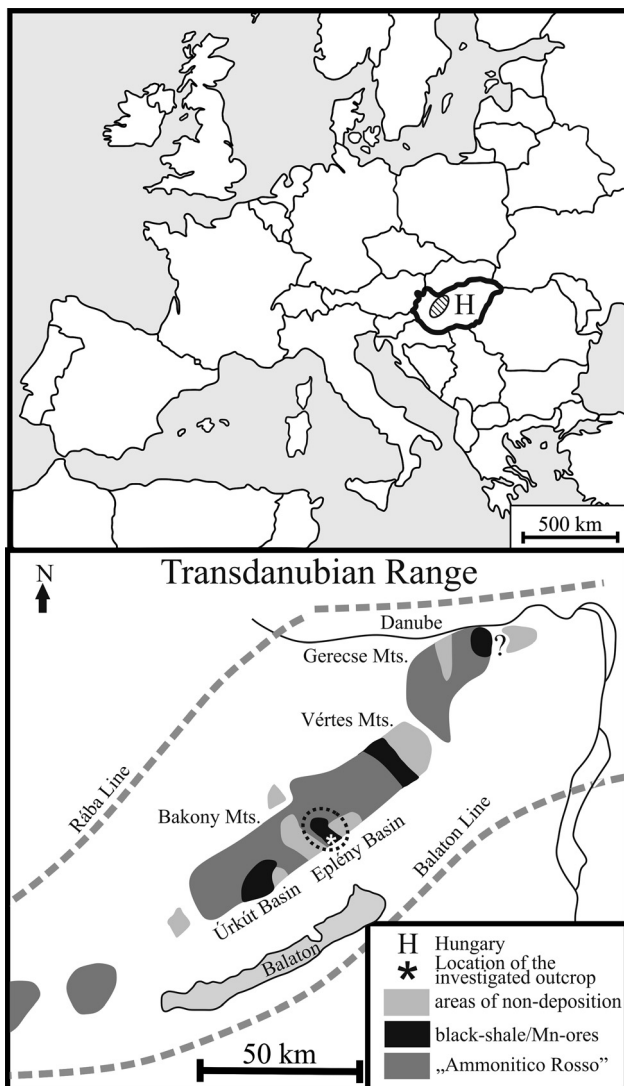


Fig. 1. Location of the studied area in the Eplény Basin, Bakony Mts., within the Transdanubian Range, Hungary (after Vörös & Galács 1998)

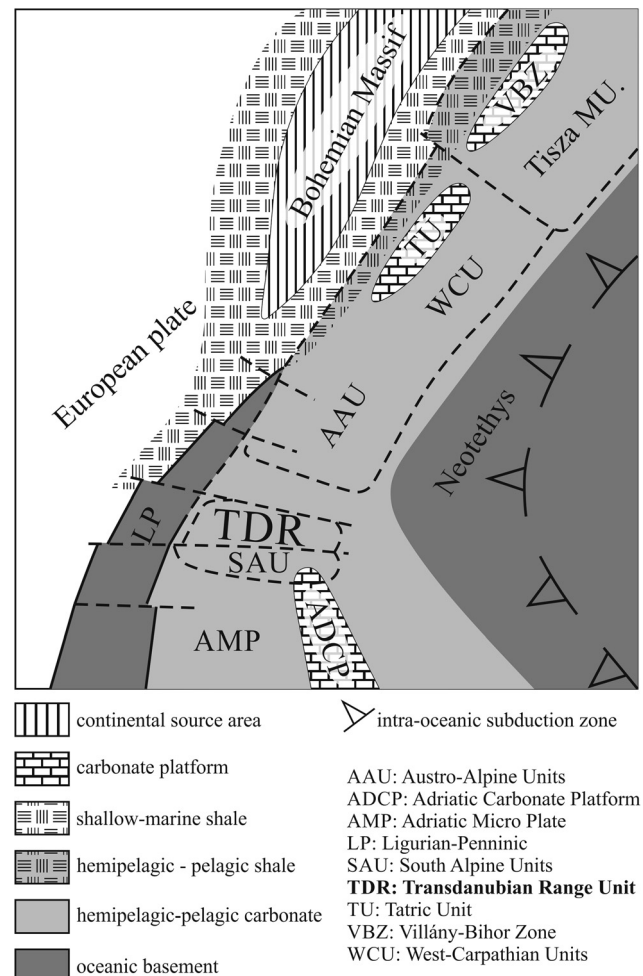


Fig. 2. Paleogeographic position of the Transdanubian Range in the latest Early to early Middle Jurassic (after Haas 2012)

intervening deep basins developed; whereas the Late Jurassic was characterized by a relative shallowing of the pelagic basin (Galác & Vörös, 1972; Vörös & Galác, 1998).

During the Jurassic, some parts of the fragmented platform remained in an elevated position (“seamounts”), where condensed red nodular limestone was deposited and hardgrounds were developed. Other blocks subsided and occupied deeper parts of the basin. Sediment was redistributed from the seamounts to the deep basins by bottom currents, resulting in the lack of sedimentation, condensed sections and hardgrounds on the submarine topographic highs. At the bottom of the fault-controlled slopes Hierlatz Limestone, a peculiar brachiopodal–crinoidal limestone facies was developed. Farther from the slope, deep marine sediments were deposited in the basin. In summary, whereas the Triassic is characterized by extensive, widespread and thick shallow marine sedimentary units, in the Jurassic only a relatively thin package of strata was formed (Konda, 1970; Galác & Vörös, 1972; Galác et al., 1985; Galác, 1988; Haas, 1988; Vörös & Galác, 1998; Haas, 2012).

The Lókút-Hosszúárok outcrop is an artificial trench, excavated in the late 1960s on the Lókúti-domb (Lókút Hill), 1.5 km east-southeast of the village of Lókút (Konda, 1970), which exposes one of the most complete and thickest Lower Jurassic successions of the Bakony Mts. in a local basinal setting. A detailed ammonite biostratigraphy of the fossiliferous strata documented the presence of all three zones of the Lower Pliensbachian in condensed, only a few meter-thick red nodular limestone (*ammonitico rosso*) facies of the Tűzkövesárok Formation (Géczy, 1976), intercalated in the Isztimér Formation (Konda, 1987). The upper part of this latter formation is only 4.5 m thick, and it consists of grey, cherty, sparsely crinoidal limestone and yielded an ammonite fauna that is assigned to the Spinatum Zone of the Upper Pliensbachian (Géczy, 1972). In addition, it also contains a moderately diverse brachiopod assemblage (Vörös, 2009). The base of this unit is an unconformity and the lower part of the Upper Pliensbachian, i.e., the Margaritatus Zone, is missing (Géczy, 1972). This unit is also unconformably overlain by the Toarcian Úrkút Manganese Ore Formation above a manganese-bearing hardground. Locally, the Úrkút Formation is ~4 m thick, represented here by dark grey and brown pelitic rock and radiolarian-bearing marlstone. Figure 3 illustrates the detailed stratigraphy of the Upper Pliensbachian layers as measured during our sampling.

## ANALYTICAL METHODS

The analytical work for this project was carried out in the laboratories of the Institute of Mineralogy and Geology at the University of Miskolc. For clay mineralogical studies, a total of 18 carbonate samples collected from the section were dissolved in 5 m/m% acetic acid (CH<sub>3</sub>COOH) and then washed with distilled water until they reached the pH of 5–5.5. The samples were dissolved in one piece, without any grinding in order to possess intact insoluble material for further investigations including SEM analysis. The <2 μm

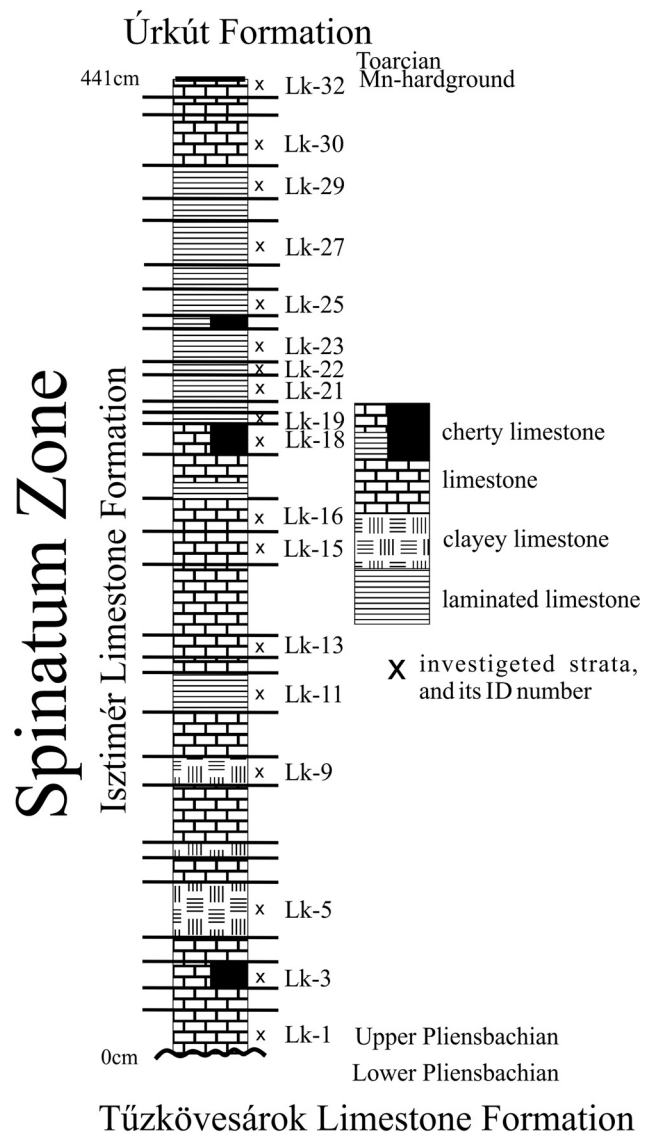


Fig. 3. Schematic section of the sampled Upper Pliensbachian layers of the Lókút-Hosszúárok outcrop

grain size fraction was separated by the gravitational method in a distilled water column. The settling time was determined using Stokes’ law (Gibbs, 1965). From the settled material, bulk X-ray powder diffraction measurements were made to identify the (060) reflections of the clay minerals for octahedral type assignment. Subsequently, the “pipette-on-glass-slide” method (Brindley, 1961) was used to prepare oriented specimens.

X-ray powder diffraction (XRD) measurements were carried out using a Bruker D8 Advance diffractometer, applying Cu-K $\alpha$  radiation, 40 mA tube current and 40 kV accelerating voltage, parallel beam geometry obtained with a Göbel mirror in reflection geometry, Vantec-1 position-sensitive detector with 1° detector window opening and 0.007° 2 $\theta$ /14 s goniometer speed. Instrumental parameters of the oriented clay mineral measurements were the same, the only difference being a longer, 29 sec-counting time. From the residual material the <2 μm grain size fraction was





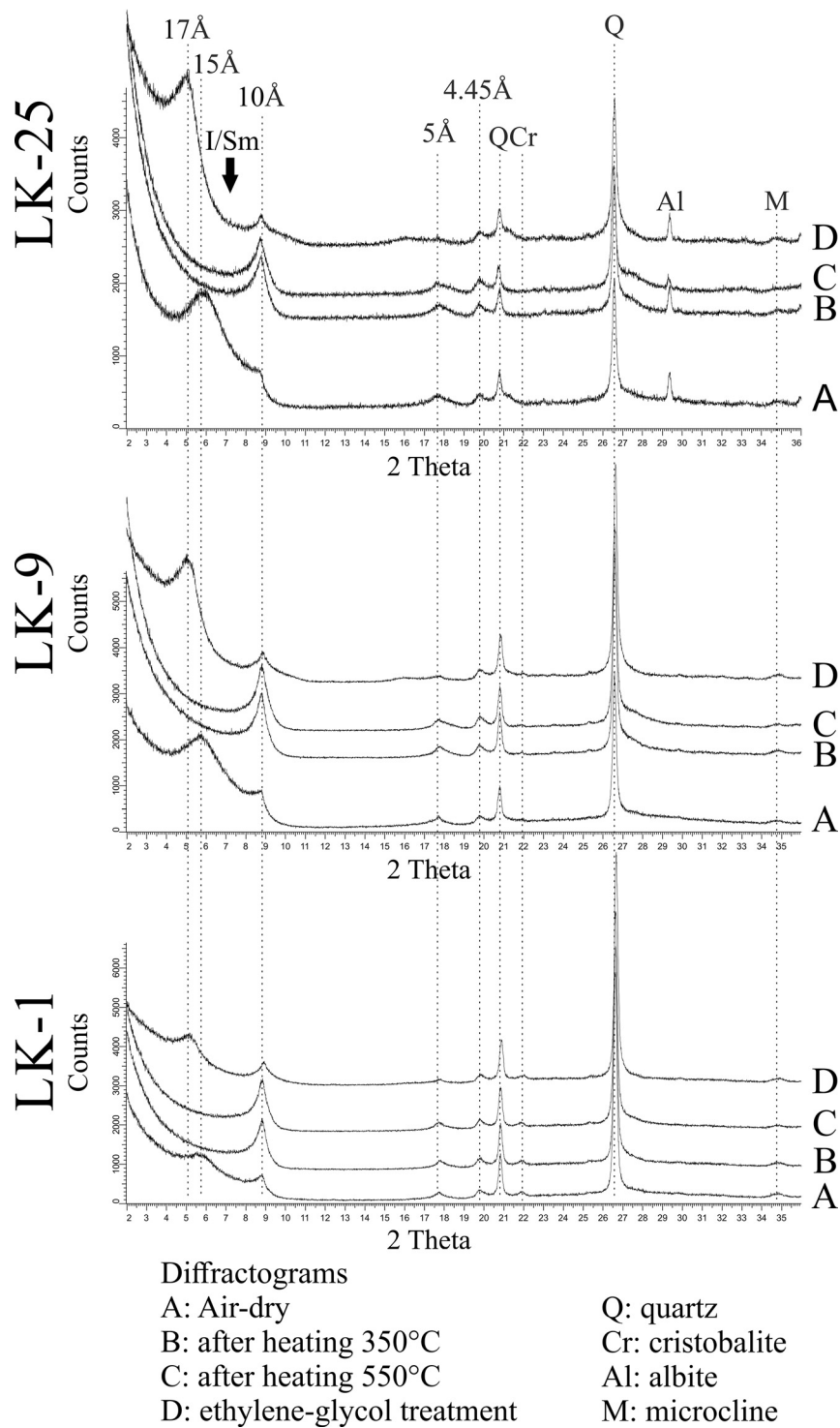


Fig. 5. Representative XRD patterns of the lower, middle and upper part of the Upper Pliensbachian section. Note the smectite peaks shifting from 15 Å to 17–17.5 Å after ethylene-glycol treatment and to 10 Å after heating

## DISCUSSION

According to previous studies (Kaeding et al., 1983; Viczián, 1995; Polgári et al., 2013), the smectite content of Lower Jurassic strata in the Transdanubian Range was unique, restricted to the Úrkút Manganese Ore deposit, in the Toarcian black or grey shale and ore bodies. However, Leskó

(2014) reported smectite for the first time below the Úrkút manganese ore deposit, from the Pliensbachian footwall. Leskó et al. (2019) proved that smectite is ubiquitous close to the Úrkút Manganese deposit within the Pliensbachian footwall.

Smectite in such facies could form in several different ways; its genesis is most commonly related to various

Table 1. Chemical composition of the dissolution residue from representative samples determined by SEM-EDS. Concentrations are expressed in m/m%

	Na <sub>2</sub> O	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	P <sub>2</sub> O <sub>3</sub>	SO <sub>2</sub>	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	FeO
LK-11	0.66	1.60	7.48	82.67	1.65	0.38	1.44	0.67	0.15	3.30
	0.99	1.78	7.49	82.32	1.55	0.58	1.41	0.66	0.15	3.06
	0.42	1.32	6.84	83.66	1.64	0.40	1.29	0.70	0.16	3.56
LK-23	0.81	2.17	9.57	78.30	1.06	0.46	1.73	0.65	1.37	3.87
	0.50	1.98	9.80	79.86	0.84	0.39	1.69	0.70	0.25	3.98
	1.60	2.78	9.30	78.60	1.19	0.65	1.53	0.49	0.32	3.53
LK-27	0.47	1.88	9.79	79.59	0.66	0.13	1.85	0.56	0.14	4.93
	0.66	2.33	10.36	74.98	1.51	0.48	1.71	0.94	1.60	5.43
	0.48	1.87	9.69	80.18	0.68	0.49	1.50	0.65	0.13	4.33

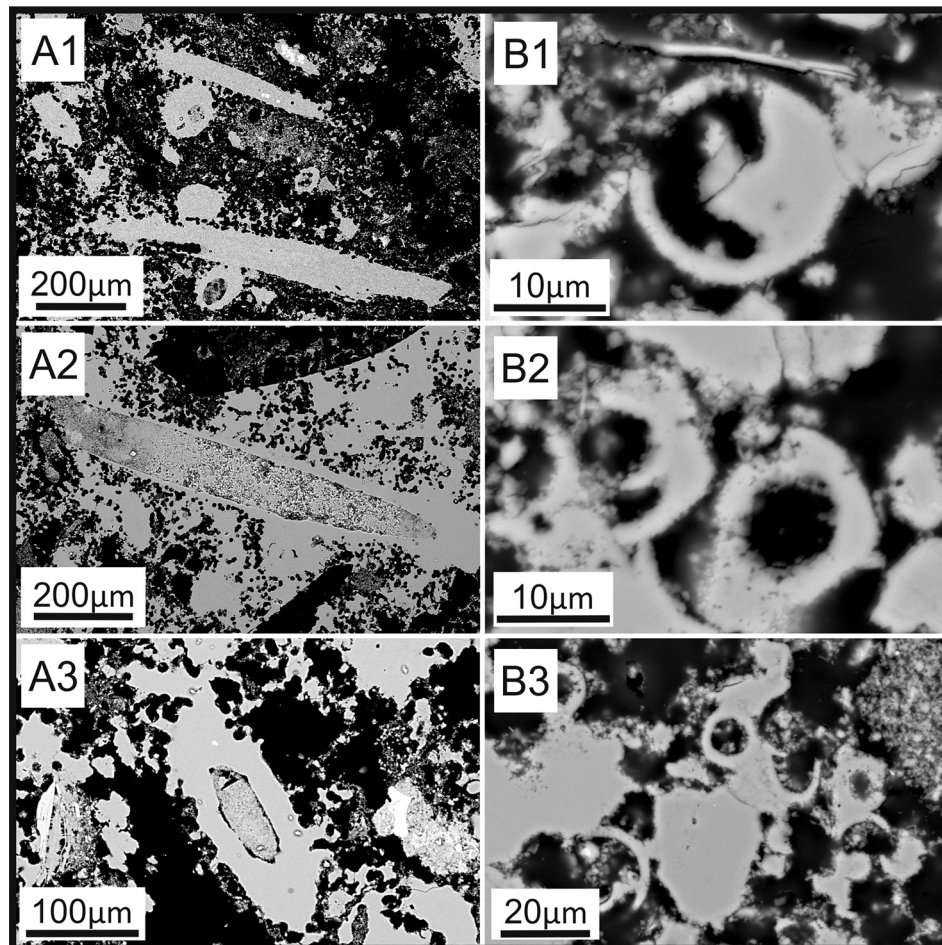


Fig. 6. SEM-BSE images from siliceous skeletons from the investigated section. Images were taken from samples LK-3: A1-B1; LK-15: A2-B2; LK-18: A3-B3

magmatic/volcanic and hydrothermal events or related alteration (Bischoff, 1972; Hein & Scholl, 1978; Seyfried et al., 1978; Cole, 1983; Bustillo & Martínez-Frías, 2003; Lantenois et al., 2008). Smectite could also form at low temperature by aging of hydrated silica colloid in the presence of the required cations, as was proven by laboratory experiments on freshly-prepared hydroxide-silica precipitates by Harder (1972, 1976). A recent research by Schöllhorn et al. (2020) found evidence for Pliensbachian magmatism/volcanism in Switzerland and in England.

Mercury content of the investigated rock units could be related to coeval magmatic/volcanic events (Varekamp & Buseck, 1986; Nriagu & Becker, 2003). Schöllhorn et al. (2020) documented that the increase of Hg concentration correlates with increasing phyllosilicate content. Unfortunately, quantitative details of the abundance and ratio of the different types of phyllosilicates are not provided in their study, where the Hg refers to coeval magmatic/volcanic events. According to their study their smectite and illite ratio change was caused by climate changes. There is no evidence

in the entire Transdanubian Range for Pliensbachian igneous rocks, tuffs, volcanic glasses, elevated mercury content, or outstanding peaks of phyllosilicate content; thus, a genetic model that invokes any magmatic or volcanic event could be excluded (Polgári et al., 2013; Leskó et al., 2019). However, the other model is more plausible for the genesis of smectite reported here and is likely to explain the occurrence of smectite in the studied deposits at Lókút-Hosszúárok. This process is also thought to be responsible for the smectite formation in the Úrkút Basin (Polgári et al., 2013; Leskó et al., 2019).

The randomly interstratified illite/smectite is generally formed through illitization of the smectite during the diagenesis. The proportion (~70–80% smectite and 20–30% illite) shows that the illitization progress was at its beginning; the sediment was buried only at shallow depth and the temperature was low (<100 °C) (Jennings & Thompson, 1986; Velde et al., 1986; Środoń, 1999; Bergaya et al., 2006; Caudros, 2006). This result fits with the models of the local (Bakony Mts.) and regional geologic history as well (Konda, 1970; Galácz et al., 1985).

The SEM-EDS results support the XRD results. The SiO<sub>2</sub> content is related to the high amount of quartz and other silicates. Illite and smectite are the main carriers of Al and Mg. Because of the cation exchange of the acetic acid treatment, Ca can occur within the smectite; K and Na relate to illite. Fe is a constituent of smectite and nanocrystalline Fe-oxide; the latter is responsible for the color of the red or pinkish limestones. P and S are likely tied to the Fe-oxides. However, no unambiguous conclusion can be drawn from the SEM-EDS results because we measured bulk composition, since the grain size of the individual crystals is too small. It was not possible to determine the chemical formula of the smectite, because multiple phases were analyzed simultaneously.

The grain size of the illite suggests the extrabasinal (i.e., detrital) origin, and the mineral composition (phyllosilicates, quartz, feldspars) of the residue assumes a sedimentary sea basin environment (Bergaya et al., 2006), which is in agreement with the previous sedimentary models of the TDR.

The silica content of the silicified limestone layers, cristobalite and partly quartz, which was found in the insoluble residue after leaching, is derived from siliceous fossils: sponges and radiolarians (Fig. 6) (Konda, 1970; von der Borsch et al., 1971; von Rad & Rösch, 1972; Cole & Shaw, 1989; Williams & Crerar, 1985; Dixit et al., 2001). Figure 6 shows remains of siliceous sponges (A1 from LK-3; A2 from LK-13 and A3 from LK-18; Moore, 1967; Matranga, 2015), and presumably radiolarian tests (B1 from LK-3; B2 from LK-13 and B3 from LK-18; Anderson, 1983; Baumgartner et al., 2007). All of the siliceous fossils are strongly recrystallized. The large amounts of radiolarians, which could contribute to bulk silicification of certain layers, suggest the presence of a local upwelling system, as upwelling could cause local radiolarian blooms (Smith, 1968; Hartline, 1981; Molina-Cruz, 1984).

Considerable similarities exist between the Eplény Basin and the Úrkút Basin, including the economically significant accumulation of black shale-hosted manganese ore, which is

restricted to these two occurrences in the Transdanubian Range, and the asymmetric geometry of sedimentation patterns (i.e., similar trends of sedimentation on opposite sides of seamounts and intervening basins; Vörös & Galácz, 1998). In addition, the large amount of biogenic silica, likely derived from radiolarians, is also comparable in the Pliensbachian strata in both basins, in Úrkút in the form of the peculiar lithology referred to as “tripoli” (Polgári et al., 2005; Leskó et al., 2019) and in Lókút represented by cristobalite and silicified carbonate layers. Localized upwelling systems can account for these observations, similar to the paleogeographic-paleoceanographic model developed for older, Middle Triassic silica-rich deposits in the Transdanubian Range (Budai et al., 2017).

Smectite was found in the Upper Pliensbachian strata only in the Eplény and Úrkút Basins, where there is an increased siliceous fossil content (radiolarites and spongiolites) (Polgári et al., 2015; Leskó et al., 2019), which could relate to upwelling events. Furthermore, smectite appears where a large amount of manganese relates to the Jenkyns Event. However, smectite was not identified in other parts of the Transdanubian Range; although other evidence of the Jenkyns Event can be encountered, there is no evidence of elevated siliceous bioaccumulation in the Upper Pliensbachian record, and no significant manganese accumulation in the Toarcian rocks. The Upper Pliensbachian layers do not have any smectite content in the Tölgyhát Quarry, Gerecse Mts., (Leskó & Zajzon, 2015; Leskó et al., 2015), which widely studied location represents the Jurassic sedimentation. These results indicate that the smectite appearance relates to the manganese accumulation. This relation was mentioned in former studies (Kaedin et al., 1983; Viczián, 1995) as well, but the present study and Leskó et al. (2019) show that the smectite appearance is more widespread than previous studies indicated; also, the formation of the smectites was interpreted in a different fashion.

## CONCLUSIONS

The occurrence of smectite in the Lower Jurassic of the Transdanubian Range is not as restricted as previous studies suggested, as it is not confined to the ore bodies of the Úrkút Manganese Ore Formation but also appears in stratigraphically lower units near the thick manganese-rich deposits within the Transdanubian Range. Our new mineralogical results indicate the presence of dioctahedral smectite and increased quartz and cristobalite content from the Pliensbachian sediments of the Lókút-Hosszúárok outcrop, in agreement with a model that emphasizes analogues with the Úrkút Basin on the basis of similar data. Our study strengthens the arguments that the Eplény and Úrkút Basins were analogous areas with similar paleogeographic position and sedimentary environments during the Early Jurassic, and similarly characteristic mineralogical paragenesis, which could have resulted from the same processes and be explained by congruent genetic



models. Our results are in agreement with other local and regional geologic observations which help to better understand the geologic evolution of the Transdanubian Range.

## ACKNOWLEDGMENTS

This study was carried out as part of the EFOP-3.6.1-16-2016-00011 “Younger and Renewing University – Innovative Knowledge City – institutional development of the University of Miskolc aiming at intelligent specialisation” project implemented in the framework of the Széchenyi 2020 program. The realization of this project is supported by the European Union, co-financed by the European Social Fund.”

This is MTA-MTM-ELTE Paleo contribution no. 334.

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