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Palynofacies analysis, sedimentology and hydrocarbon potential of the Menilite Beds (Oligocene) in the Slovakian and Romanian Outer Carpathians

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The sedimentary organic matter (SOM) assemblages and sedimentology of the Menilite Beds from the Dukla, Grybów and Vrancea units in the Slovakian and Romanian Outer Carpathians are described. Qualitative and quantitative analyses of the SOM help ascertain depositional conditions, while the thermal maturity of the organic matter studied is estimated utilizing the Spore Colour Index and UV light excitation techniques. The sedimentary organic particles were grouped into ten SOM categories: marine palynomorphs (dinoflagellate cysts), sporomorphs (saccate and non-saccate, pollen and spores), freshwater algae (*Botryococcus* sp., and other freshwater microplankton), phytoclasts (cuticles, translucent wood, opaque wood), resin and amorphous organic matter (AOM). All samples are dominated by AOM. The presence of *Botryococcus* sp., *Pediastrum* sp., *Pterospermella* sp. and *Campenia* sp., in some samples points to deposition under hyposaline conditions. It is interpreted that the freshwater influx induced water column stratification in the basin, leading to the development of dysoxic to anoxic bottom-water conditions that enhanced the preservation of AOM. Kerogen analysis in UV light and evaluation using the Spore Colour Index demonstrated different thermal maturation patterns from the Slovakian (post-mature) and Romanian (immature) sections. Integrated palynofacies analysis (notably, the presence of freshwater algae) and sedimentological observations (e.g., hummocky cross-stratification) lead to the conclusion that the deposition of the Menilite Beds in the Vrancea Unit (Romania) was relatively proximal to the shoreline, above storm wave base, whereas the Slovakian units (Dukla and Grybów) were deposited in a more distal setting.

Key words: Menilite Beds, palynofacies, UV fluorescence, thermal maturity, Carpathians.

INTRODUCTION

Organic-rich sediments deposited in the Paratethys Ocean during the Oligocene have long been considered to be potential hydrocarbon source rocks (Zuber, 1918; Szajnocha, 1920; De Cizancourt, 1931; Gucik, 1980; Ziegler and Roure, 1999). The Menilite Beds are an example of these deposits, in which total organic carbon (TOC) can exceed 20% (Köster et al., 1998; Kosakowski et al., 2009, 2018). Consequently, this stratigraphic unit has been investigated primarily for its geochemical properties and potential for hydrocarbon generation (Koltun, 1992; Köster et al., 1998; Kotarba et al., 2007; Belayouni et al., 2009; Kosakowski et al., 2009, 2018; Sachsenhofer et al., 2015; Wendorff et al., 2017) over many years, a particularly relevant topic given the numerous controversies regarding the depositional setting of the Menilite Beds (Kotlarczyk et al., 2006; Jankowski, 2015; Dziadzio et al., 2016). However, existing studies rarely use data from sedimentary organic matter (SOM) analyses in their environmental interpretations, relying instead on palynomorph assemblages (Olaru, 1970; Ţabără, 2010, 2017; Ţabără et al., 2015).

Biostratigraphic studies based on calcareous nanoplankton (Belayouni et al., 2009; Garecka, 2012), foraminifers (Olszewska, 1982, 1985), dinoflagellate cysts (Ţabără, 2010, 2017; Ţabără et al., 2015), and ichthyofauna (Kotlarczyk et al., 2006) suggest that the Menilite Beds of the Outer Carpathians were largely deposited in the Rupelian (Early Oligocene). However, in the marginal parts of the basin, deposition continued until the Late Oligocene. In some places, deposition extended into the late Egerian–earliest Aquitanian (Miocene, NN1 Calcareous Nannoplankton Zone; Andreyeva-Grigorovich and Gruzman, 1994; Andreyeva-Grigorovich et al., 1997; Garecka, 2012; e.g., the Skole Unit). The development and deposition of Menilite-type facies (organic-rich, fine-grained Oligocene strata) across the entire Outer Carpathians (Czech Republic, Poland, Slovakia, Ukraine, and Romania) was initially driven by progressive extension in the Early Oligocene. Concurrent tectonic motion in the Paratethys region caused the isolation of the basin from the open ocean. Therefore, the restriction of saline oceanic circulation and considerable freshwater influx into the basin (Baldi, 1980) spurred water column stratification (Bojanowski et al., 2018) and the periodic development of dysoxic conditions at the sediment-water interface (Kotlarczyk and Uchman, 2012). However, Miclăuş et al. (2009) suggested that anoxic conditions might have also been favoured by increased biological productivity caused by the isolation of the Parathetys, global climate changes, or relative sea-level fluctuations. Oxygen-deficient conditions, combined with relatively

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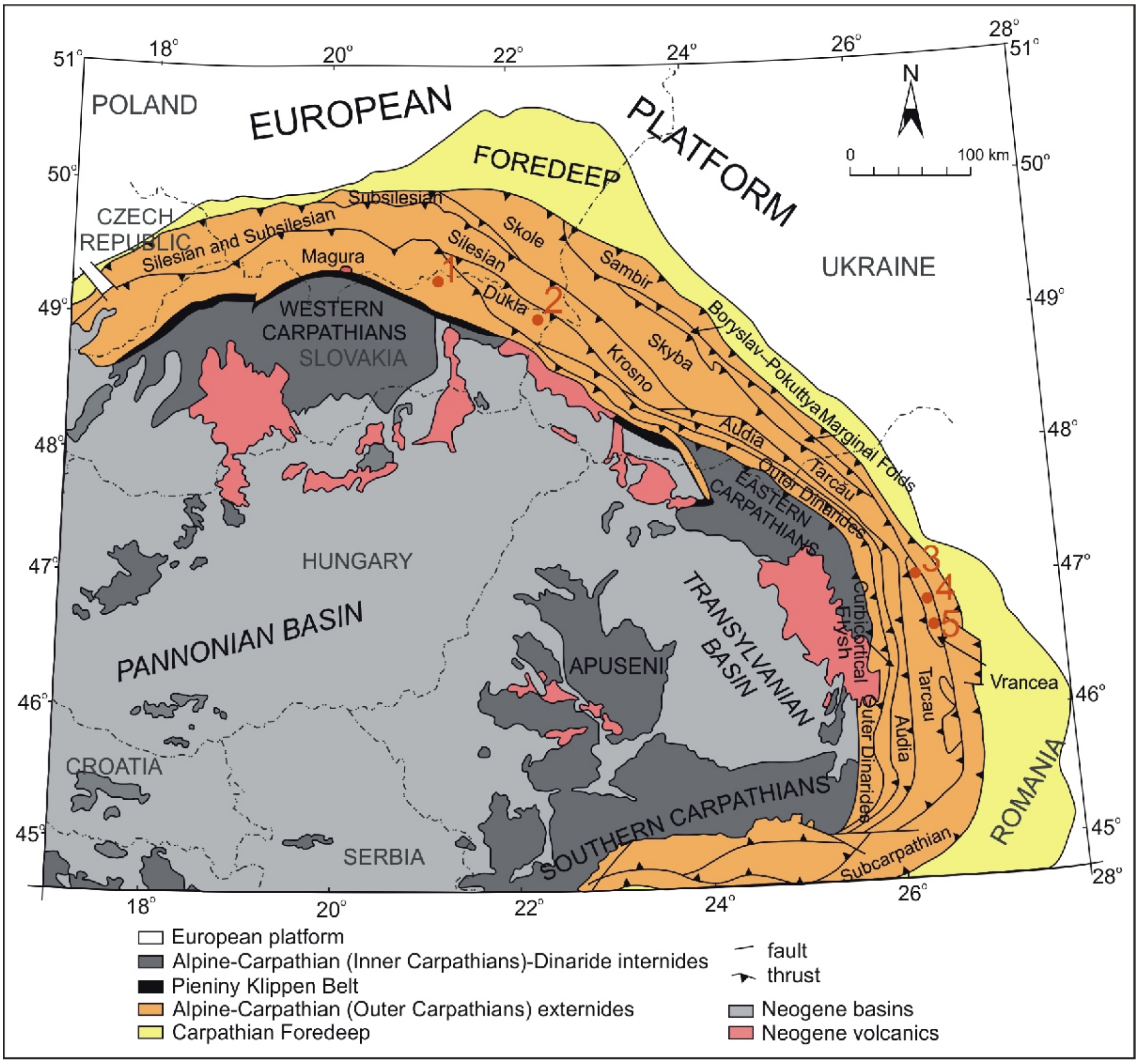

Fig. 1. Schematic map of the Alpine-Carpathian-Dinaride domain, with location of the sections studied indicated (after Kováč et al., 2007)

Slovakia: 1 – Smilno, 2 – Dara Prislop; Romania: 3 – Agapia, 4 – Piatra Neamt, 5 – Nechit

stable sedimentation, drove the deposition of organic-rich, fine-grained sediments throughout the entire basin(s), leading to a high degree of facies unification (Jankowski, 2004; Kotlarczyk and Uchman, 2012). In the geological record, the presence of anoxic events is often reflected in the enhanced preservation of SOM. A qualitative and quantitative analysis of SOM may allow for the determination of sedimentary conditions and fluctuations within the anoxic event interval across isochronous horizons and within vertical successions of macroscopically identical rocks (Pilskaln, 1991). Palynofacies analysis has the potential to reveal hitherto hidden aspects of the depositional setting and changes within the Menilite Basin.

Palynofacies analysis is based on the optical microscopy of organic matter released from the mineral phases of the rock (Combaz, 1964). This method, based on the diversity of observed organic matter, allows for the determination of the petroleum potential (by estimation of kerogen type) of the sedimentary rocks and the reconstruction of their depositional environments (Tyson, 1995; Batten, 1996). Organic sedimentary particles visible under a transmitted light microscope may show a large diversity of colours, morphologies, opacities, and recognisable structures (Tyson, 1995), which is why both quantitative and qualitative palynofacies analysis are useful in reconstructing depositional settings.

This study focuses on Menilite Beds outcrops currently separated by great distances (~600 km) and located within different tectonic units (Fig. 1). Palynofacies and sedimentological analysis should allow for the verification of (1) sedimentary conditions within the Oligocene Carpathian basin(s) in potentially distant depositional zones and (2) organic matter distributions as reflected in the present tectonic pattern.

The aims of the present study include: (1) a palynofacies characterisation of the Menilite Beds within the Vrancea Unit (Romania) and the Smilno and Dara Prislop units (Slovakia); (2) a determination of their respective sedimentary settings; and (3) an assessment of the hydrocarbon potential of the Menilite Beds.

GEOLOGICAL SETTING

The study area is located within the Outer Carpathians, alternatively referred to as the Flysch Carpathians (e.g., Kováč et al., 2007; Jankowski et al., 2012). The Carpathians are the eastern prolongation of the Alpine arc and were formed after the closure of the Tethys Ocean during the Cretaceous to the Miocene, as a consequence of the collision of a series of microplates with the European Platform (Froitzheim et al., 2008). According to the majority of conceptual models, this process may have terminated with the subduction of the European Platform margin under a group of microplates, coincident with the complete closure of the Tethys Ocean. This, in turn, resulted in the deformation and imbrication of strata accumulated within the so-called Outer Carpathian Basin (Jankowski, 2015; Olszewska and Szydło, 2017), the Carpathian Foreland Basin (e.g., Grasu et al., 1999; Puglisi et al., 2006; Miclăuş et al., 2009), or the Outer Carpathian Ocean (Golonka et al., 2006) and their partial overthrusting onto the platform (Maheľ and Buday, 1968; Książkiewicz, 1972; Maheľ, 1974). Traditionally, in its western extent the Carpathian Orogen is subdivided into the Inner Carpathians and the Outer Carpathians (see Maheľ and Buday, 1968; Książkiewicz, 1972).

In Romania, the Outer Carpathians are referred to as the Moldavide Belt (Săndulescu, 1975, 1980). In some reports (Andrusov, 1968; Plašienka et al., 1997), the term “Central Western Carpathians” is used interchangeably with “Inner Western Carpathians”. The boundary between the Inner and Outer Carpathians is traditionally interpreted to occur along the Pieniny Klippen Belt, a narrow structure presently interpreted in two ways: (1) as a Early/Middle Miocene suture between the European foreland and the Alcapa Block of the Inner Carpathians (Birkenmajer, 1986; Csontos and Nagymarosy, 1998), or (2) as a tectonic melange zone (Jankowski, 2015) developed along a strike-slip fault (Birkenmajer, 1977; Jurewicz, 2005) and characterized by the presence of both older (Triassic, Jurassic, Cretaceous) and younger (Paleogene) strata. In the marginal Inner and the southern part of the Outer Carpathians, Oligocene sediments were deposited in basins during the extensional phase (Central Carpathian Paleogene Basin – Janočko and Jacko, 1998; Soták, 1998; East Carpathian Paleogene Basin – Jarmołowicz-Szulc and Jankowski, 2011). In the Romanian portion of the Carpathians, Oligocene sediments were deposited in the hinterland of the Outer Carpathians or in the Moldavides. Traditionally, the following units are distinguished in the Slovakian and Polish segments of the Outer Carpathians: from south to north, the Magura Unit, the Dukla Unit, the Silesian Unit, a narrow belt of the Węglówka Unit, and the Skole Unit. Some researchers (see Kotlarczyk, 1988; Jankowski et al., 2004) also distinguish the Boryslav–Pokuttya Unit in the vicinity of Przemyśl.

The Romanian Carpathians are also subdivided into two regions: an inner one, composed of crystalline basement nappes and Mesozoic sedimentary rocks, and an outer one, primarily composed of Cretaceous and Paleogene flysch deposits (Săndulescu, 1975, 1980; Balla, 1986; Badescu, 1997). The innermost tectonic units (i.e., the Macla and Audia nappes) and the outermost units (the Tarcău, Vrancea and Subcarpathian nappes) constitute the Moldavide Nappe Complex (Săndulescu, 1975; Badescu, 1998; Fig. 1).

According to many studies, deposition within the Carpathians took place in a series of basins or sub-basins that correspond to the present-day units of the Outer Carpathians (Książkiewicz, 1972; Golonka and Krobicki, 2004; Ślączka et al., 2006, 2012; Oszczypko-Clowes et al., 2015). The Skole, Sub-Silesian, Silesian, Dukla, and Magura sub-basins and their eastern prolongation formed part of the rifted European margin (Oszczypko and Oszczypko-Clowes, 2003; Golonka et al., 2006). Adjacent basins were locally separated by ridges: for instance, the Silesian ridge separated the Silesian and Magura basins (e.g., Golonka et al., 2006). According to Birkemajer (1986) and Golonka et al. (2006), it is envisioned that the Silesian and Magura basins were underlain by oceanic basement. Traditionally, these basins have been interpreted as primarily filled with deep-water, mostly tubiditic deposits (e.g., Kotlarczyk and Uchman, 2012). However, some authors have recently suggested that deposition in these areas took place in a single basin, subject to multi-stage rebuilding as a result of changes in the tectonic regime (Jankowski, 2004). In this case, the formation of the Carpathian basins was driven mainly by decomposition of the East European Platform margin. Contractional stages pinpoint the formation of a foreland basin (Jankowski and Wysocka, 2019). Jankowski (2007, 2015) and Jankowski and Wysocka (2019) have additionally distinguished an extensional stage, during which sedimentation took place in half-graben structures. The process of basin closure and sedimentation cessation was diachronous, progressing from west to east; in Romania, this process extended into the Pliocene (e.g., Royden and Baldi, 1988; Linzer et al., 1998; Golonka et al., 2006).

MENILITE BEDS

The Menilite Beds (the name is derived from a variety of opal, known as menilite; Glocker, 1843) occur in almost all tectonic elements of the Western and Romanian Carpathians (Murgeanu et al., 1970; Lexa et al., 2000; Jankowski et al., 2004, 2007, 2012; Wagner, 2008). The unit is also referred to as the Menilite Formation, Amphisyla, the Disodilic Beds (Cordier, 1808, derived from disodil, a variety of menilite), and the Meletta, Czeczwin, and Smilno beds (Glocker, 1843; Jankowski et al., 2004, 2007, 2012). In the western part of the Romanian Carpathians, the Menilite Beds are divided into two units: (1) the Menilite Beds (lower), which mainly consists of cherts and shales, is separated by the Bituminous Marl from (2) the shale unit (upper) with cherts and sandstones, here referred to as the Disodilic Beds (upper; e.g., Jankowski et al., 2012; Ţabără, 2017; Fig. 2). In the Slovakian Carpathians, the Menilite Beds are divided into three subunits: (1) the Cherts, (2) the Lower Menilite Beds, and (3) the Upper Menilite Beds (Stránik and Hanzlikowá, 1963). The Menilite Beds are variable in thickness, which is largely controlled by the development of sandstone units (e.g., Gucik and Wójcik, 1982; Gucik, 1987; Kotlarczyk and Leśniak, 1990; Kotlarczyk et al., 2006). Jarmołowicz-Szulc and Jankowski (2011) interpreted the occurrence of different lithologies (i.e., conglomerates, sandstones, diatomites, marls, carbonates, and different types of shale) as associated with deposition in morphologically diverse basin(s).

The deposition of the Menilite Beds initiated in the early Oligocene in an isolated, marginal part of the Paratethys. The basin was characterized by fluctuating salinity and restricted circulation (Baldi, 1980; Popov et al., 2010). The Menilite Beds are dominated by organic-rich, fine-grained strata deposited under anoxic conditions (e.g., Pauča, 1936; Vetö, 1987; Sachsenhofer et al., 2015). The basal boundary of the Menilite Beds is considered to be isochronous (Olszewska, 1985). In contrast, their upper boundary is diachronous (Garecka, 2008, 2012). The Menilite Beds are progressively overlain from the south by the Krosno Beds. In the marginal portions of the Outer Carpathian Basin(s), sedimentary breccias, such as the Slon Beds, the Gura Soimului Beds, and the Vorotyshcha Beds, overlie the Menilite Beds (Fig. 2; Grasu et al., 1988; Jankowski et al., 2012).

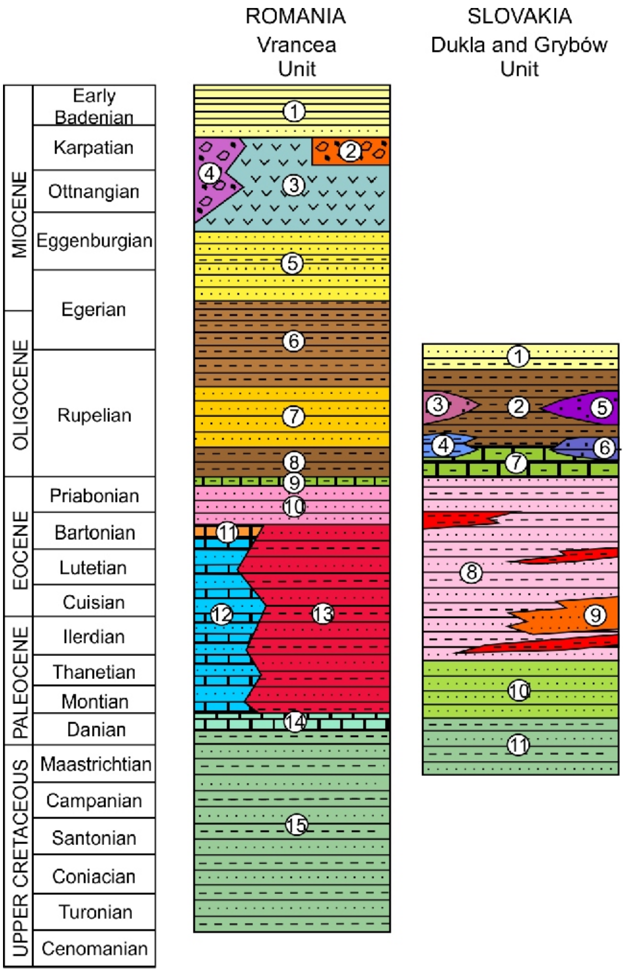

Fig. 2. Correlation of regional lithostratigraphic units (based on Jankowski et al., 2012) with the exposures studied indicated

Units from which samples were collected are underlined; Romania, Vrancea Unit: 1 – Doftana Beds, 2 – Brebu Conglomerate, 3 – Salt Beds, 4 – Slon Beds, 5 – Gypsum Beds (Gura Soimului Beds, Gura Misina Beds), 6 – Upper Dysodilic Shale, 7 – Kliva Sandstone, 8 – Lower Dysodilic Shale, Bituminous Marl, Menilite Beds, 9 – Globigerina Marl, Lucăceşti Sandstone, 10 – Bisericani Beds, 11 – Doamna Beds, 12 – Jgheabu Marl, 13 – Greşu Beds, 14 – Piatra Uscata Beds, Caşin Beds, 15 – Lepşa Beds; Slovakia, Dukla and Grybów units: 1 – Cergowa Beds, 2 – Menilite Beds, 3 – Kliva Sandstone, 4 – Cergowa Sandstone, 5 – Papin Beds, 6 – Mszanka Beds, 7 – Globigerina Marl, 8 – Submenilite Beds, 9 – Bukovec Sandstone, 10 – Cisna Sandstone, 11 – Łupków Beds

Thin layers of coccolithic limestone, presumably related to phytoplankton blooms, are found in the Menilite Beds (Uhlig, 1882; Nowak, 1965; Haczewski, 1989; Ciurej and Haczewski, 2016). These layers – the Tylawa, Wujskie, and Jasło limestones – are considered to be isochronous (Jucha and Kotlarczyk, 1961; Koszarski and Żytko, 1961; Jucha, 1969; Kotlarczyk et al., 2006). In the lower part of the Menilite Beds succession, regional tuff horizons (Sikora et al., 1959; Koszarski and Wieser, 1960) and diatomites (the Futoma Diatomite) are recognized (Krzyżanowski, 1962; Kotlarczyk, 1988; Kotlarczyk and Leśniak, 1990).

Numerous lithostratigraphic equivalents of the Menilite Beds have been distinguished, which are referred to as the "Menilite facies" due to their common features (Oligocene organic-rich, primarily fine-grained strata; Jarmołowicz-Szulc and Jankowski, 2011). The Menilite Beds are present within the successions of the Central Carpathian Paleogene Basin (Poland, Slovakia, and the Czech Republic), the Buda-type Paleogene (Croatia, Slovenia and Hungary), the Transylvanian Basin (Romania), and the Black Sea region (Ukraine and Russia; Puglisi et al., 2006; Jarmołowicz-Szulc and Jankowski, 2011; Filipek et al., 2017; Mayer et al., 2018). The Menilite Beds are stratigraphically equivalent to a portion of the fine-grained source rocks from the eastern Paratethys called the Maikop Group, which was deposited in similar, brackish-water conditions during the "Solenovian Event" (Voronina and Popov, 1984; Popov et al., 1993; Popov and Studencka, 2015). In particular, marlstones with different regional nomenclature (the Dynów Marl, the Ostracoda Beds and the Romanian Bituminous Marl) are considered to be a Paratethys-wide marker horizon (Mayer et al., 2018). Uniform, shallow-water faunas of small bivalves, with the notable presence of Solenovian-type endemic genera, locally together with various faunal groups (e.g., freshwater calcareous nannofossils, marine fish), have been identified in these marlstones in Poland, Ukraine, the Czech Republic, Kazakhstan, Georgia and Romania (Kazakhashvili, 1984; Voronina and Popov, 1984; Rusu, 1999; Popov and Studencka, 2015; Studencka et al., 2016).

HYDROCARBON POTENTIAL OF THE MENILITE BEDS

The Outer Carpathians are one of the oldest petroleum-producing regions in the world: the first oil exploration in this region was documented in 1853 (Karnkowski, 1999). Petroleum accumulations have been identified in almost all sandy lithostratigraphic members (the Istebna, Hieroglyphic and Krosno Beds; Fig. 2) and in the Paleozoic–Mesozoic basement of the Outer Carpathians (Kotarba and Koltun, 2006; Więcław et al., 2011).

The bituminous Menilite Beds are considered to be the one of the source units in the Carpathian region (see Zuber, 1918; Ionescu, 1994; Dicea, 1995; Kotarba and Koltun, 2006; Stefanescu et al., 2006; Belayouni et al., 2009; Kosakowski, 2013). The high hydrocarbon potential of the Menilite Beds is interpreted and deduced based on its high TOC values and kerogen type. In some localities, TOC exceeds 20% (Köster et al., 1998; Kosakowski et al., 2009, 2018; Sachsenhofer et al., 2015; Jirman et al., 2018; Rauball et al., 2019). The kerogen content of the Menilite Beds consists mainly of oil-prone, low-sulphur Type-II kerogen, with the occasional occurrence of Type-III kerogen (Curtis et al., 2004; Lewan et al., 2006; Kosakowski et al., 2009; Rauball et al., 2019). In the Menilite Beds, variability in kerogen type reflects a combination of the input of different sources of organic matter, together with variable environmental conditions and bathymetry (Kotarba and Koltun, 2006). Within the Outer Carpathians, the eastern part of the Silesian Unit and the Ukrainian region are characterized by the greatest hydrocarbon potential, primarily due to the dominance of oil-prone Type-II kerogen (Kotarba and Koltun, 2006). The Menilite Beds reached a spectrum of distinct thermal maturation levels, primarily driven by variations in organic matter type, burial history, and geothermal gradient (e.g., Stefanescu et al., 2006).

Geochemical analyses of the Menilite Beds in several outcrops in the Polish, Ukrainian and Romania Carpathians indicate that the stratigraphic unit is thermally immature to marginally mature, reaching into the early stages of the oil window (Curtis et al., 2004; Anastasiu, 2016; Wendorff et al., 2017; Kosakowski et al., 2018; Rauball et al., 2019). In the Polish and Ukrainian Carpathians, thermal maturity of the Menilite Beds increases from the outer to the inner tectonic units, from imma-

Table 1

GPS coordinates for the investigated exposures (source: ArcGIS Basemap)

Locality	Sample	GPS coordinates	
Dara Prislop	DP 10, DP 9B, DP 9A, DP 8B, DP 8A, DP 7, DP 6, DP 5C, DP 5B, DP 5A, DP 4B, DP 4A, DP 3B, DP 3A, DP 2, DP 1A	49°2'50.644"N	22°17'25.598"E
Smilno	S 2	49°21'22.026"N	21°25'45.778"E
	S 12	49°21'22.026"N	21°25'45.778"E
	S 16	49°21'22.026"N	21°25'45.778"E
	S 24	49°21'22.421"N	21°25'45.500"E
	S 30	49°21'22.421"N	21°25'45.500"E
Nechit	N 1008b, N 1008a	46°45'55.166"N	26°26'19.824"E
	1003a	46°46'15.899"N	26°25'37.715"E
	1001	46°46'14.949"N	26°25'31.904"E
	1000	46°46'15.107"N	26°25'27.847"E
	993b	46°46'21.166"N	26°25'12.933"E
	991	46°46'20.298"N	26°25'8.461"E
	977	46°46'16.468"N	26°24'11.765"E
	971	46°46'14.246"N	26°23'58.661"E
	962	46°45'53.777"N	26°23'4.056"E
	961	46°45'53.042"N	26°23'1.766"E
	960	46°45'52.801"N	26°23'1.277"E
	952	46°45'53.064"N	26°22'50.754"E
	949	46°45'51.595"N	26°22'46.762"E
Piatra Neamt	PN KG	46°56'27.840"N	26°22'10.320"E
	PN SPK	46°56'13.200"N	26°22'10.320"E
	PN PM	46°56'5.220"N	26°22'4.320"E
	PN PK	46°56'17.940"N	26°22'7.860"E
Agapia	A 1040b, A 1040a	47°9'43.715"N	26°13'50.696"E
	A 1041	47°9'44.525"N	26°13'50.308"E
	A 1042j, 1042c	47°9'46.019"N	26°13'54.415"E

ture in the Skole and Boryslav–Pokuttya units to overmature in the southern parts of the Silesian and Dukla units (Kotarba et al., 2007; Kosakowski et al., 2018). A similar relationship between thermal maturity and location within the orogen was also noticed in Romania (Vrancea and Tarcău units) by Wendorff et al. (2017). However, at some localities in Ukraine (the Boryslav–Pokuttya Unit and locally the Skiba Unit), where the unit was buried to between 3 and 6 km depth, the Menilite Beds are in the oil or gas window (Koltun, 1992; Kosakowski et al., 2018). The organic geochemical compounds of oils, and in particular the presence of oleanane, at these localities suggest that they were generated from the Menilite Beds (Kotarba and Koltun, 2006; Więcław et al., 2012) from an early stage to the peak of the oil window.

The presence of sandstone and conglomerate lithosomes in the Menilite Beds (e.g., the Kliva, Magdalena and Cergowa Sandstones, and the Maly Vyžen Beds; Jankowski et al., 2012) is important, as these coarse-grained siliciclastic strata serve as potential hydrocarbon migration routes or reservoir rocks. Coarse-grained deposits of Oligocene age have good reservoir potential in both shallow and deep structures of the Outer Carpathians (Dziadzio et al., 2006). In some areas, the Menilite Beds may be source rocks as well as reservoir rocks.

MATERIAL

In the present study, samples were collected from three geological units: the Dukla and Grybów units (Slovakia), and the Vrancea Unit (Romania; Fig. 1 and Table 1). In total, 42 samples from the Menilite Beds were collected from the following exposures: Smilno (Slovakia; 5 samples), Dara Prislop (Slovakia; 16 samples), Piatra Neamt (Romania; 4 samples), the Nechit River (Romania; 14 samples), and Agapia (Romania; 3 samples from the Menilite Beds and 2 samples from the Slon Beds (equivalent of the Vorotyshcha Beds; Fig. 1). It can be assumed, based on lithostratigraphic studies (Kotlarczyk et al., 2006; Jankowski et al., 2012), that the Menilite Beds of Romania (Nechit, Bituminous Marl) and Slovakia (Dara Prislop, Cergowa Beds) were deposited at approximately the same time. To evaluate differences in organic matter distribution between the various parts of the sedimentary basin, the samples were collected at locations originally distant from each other during the deposition of the Menilite Beds. Furthermore, these localities are located in different tectonic units at present, allowing for an assessment of post-depositional variability in their development (e.g., thermal maturity differentiation).

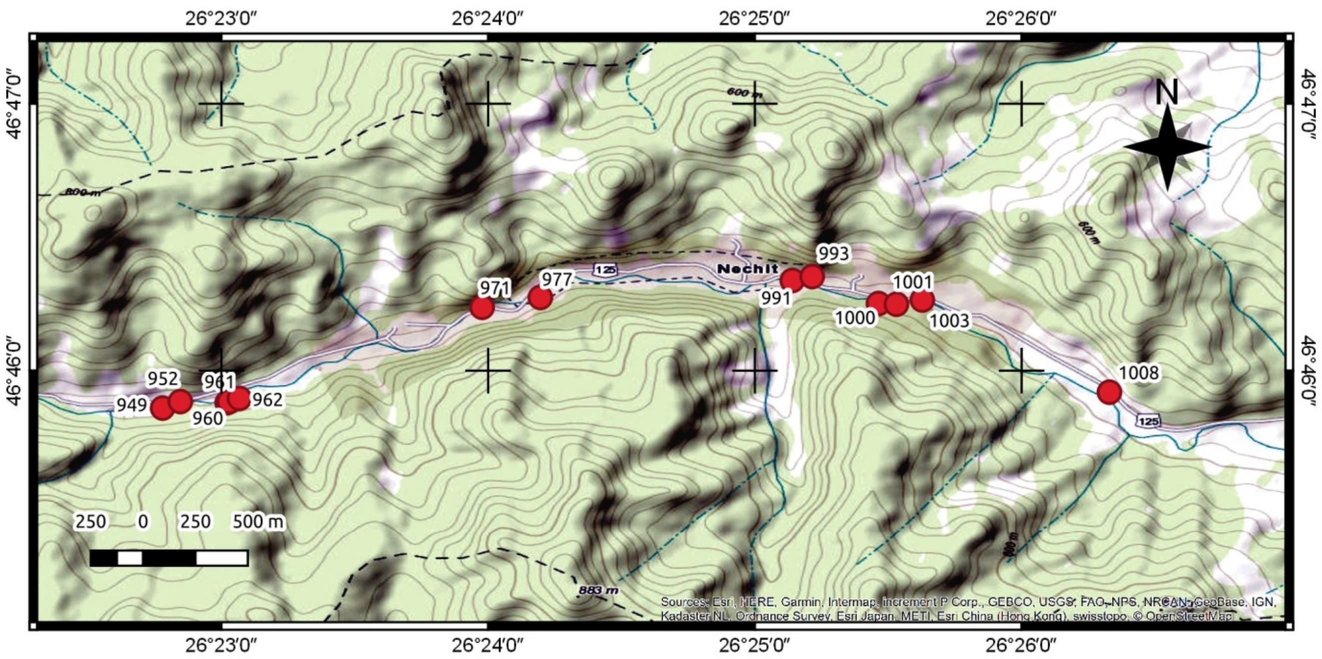

Fig. 3. Topographic map of the Nechit River area (Vrancea Unit, Romania), with sampling sites marked

The number and distribution of samples from each section was dependent on the accessibility and type of exposure. In tectonically undisturbed sections of considerable thickness (Dara Prislop, Smilno, Piatra Neamt, and Agapia), samples were collected within the stratigraphic order of the section. At the Nechit River site, owing to regional folding and the partly inaccessible nature of the Menilite Beds, the samples were collected from exposures located along the stream (Fig. 3). Samples were collected from various lithologies (mainly shales) at each of these localities (Figs. 4 and 5).

METHODS

All samples collected were processed following modified palynological techniques used at the laboratory of the Faculty of Geology, University of Warsaw. 40.23–43.00 grams of crushed rocks were treated with 37% hydrochloric acid to remove carbonates. Different times of HCl were used according to the carbonate content of individual samples. At least one hour of HCl treatment was enough for shales and mudstones. In case of samples of marl and limestone, small amounts of HCl are successively added until acceleration of the reaction ceases. After that samples were washed with H_2O, which is removed during decanting. Then 70% hydrofluoric acid was used to remove silica and silicates. Samples with HF were left for one week. The organic residuum was sieved through a 15 μm mesh sieve. In order to concentrate palynomorphs, a heavy liquid separation was used ($ZnCl_2$; density 2.0 g/cm^3) and the residuum was sieved once again through a 15 μm mesh sieve. Slides were made for each sample, using UV-cured glue as the mounting medium. Microphotographs were taken in transmitted and UV light using a *Nikon Eclipse E-600* microscope equipped with a digital camera.

In the present study, at least 300 SOM particles were counted, and statistical analyses were conducted using various organic components of the SOM (Tyson, 1995). The abundant structureless AOM was studied under UV light to determine its primary components and the degree of subsequent reworking (e.g., Van Gijzel, 1961). The SOM recognized in this study is grouped into ten categories (*sensu* Combaz, 1964), shown in Table 2. The maturity of the Menilite Beds was also evaluated using the Spore Colour Index Chart and this was used for the first time in the Slovakian area (Fischer et al., 1980).

RESULTS

LITHOLOGY AND SEDIMENTOLOGICAL FEATURES OF THE SECTIONS STUDIED

SMILNO

The section studied in Smilno is located in the Grybów Unit (Slovakia). In this exposed section the Menilite Beds are dominated by fine-grained deposits (laminated siltstones, shales), which are in places interbedded with sandstone layers characterized by sedimentary structures including horizontal lamination, lenticular bedding, flaser bedding, ripple cross-lamination, sole marks (Fig. 4E) and deformation structures. The shales are mainly black and locally contain traces of oil. Samples were collected from the shales.

DARA PRISLOP

In the Dara Prislop section (Slovakia), the Cergowa Beds sandstones were observed. They are represented by several repetitive sequences of clastic rocks (mostly characterized by a fining-upwards sequence; Fig. 4B). Most packages start with micaceous, fine-grained sandstones, which transition into grey sandy mudstones, ultimately capped by black siltstones/shales. Sole marks (tool marks and flute moulds), indicative of different palaeotransport directions (from E to W, and from N to S), were identified in the section (Fig. 4C). Convolution and load structures were also present in the sandstone layers (Fig. 4D). Samples for palynofacies analysis were collected from different lithologies, with the intention of recognising the diversity of SOM in various rock types.

Fig. 4. Menilite Beds and Slon Beds in the Slovakian and Romanian sections

A – general overview of the exposure at Dara Prislop; **B** – packages represented by repeated clastic rock sequences (fine-grained sandstones through sandy mudstones passing into siltstones), Dara Prislop, triangles indicate fining-up successions; **C** – sole marks (Dara Prislop), arrows point in the direction of transport; **D** – load structures (Dara Prislop); **E** – sole marks (Smilno); **F** – general overview of the Bituminous Marl exposure at Piatra Neamt; **G** – general overview of the Menilite Shale exposure at Piatra Neamt; **H** – strongly deformed clasts of the Menilite Shale incorporated into a yellow matrix (Slon Beds, Agapia); photographs A–D, F–H by Anna Wysocka, photograph E by Marcin Barski

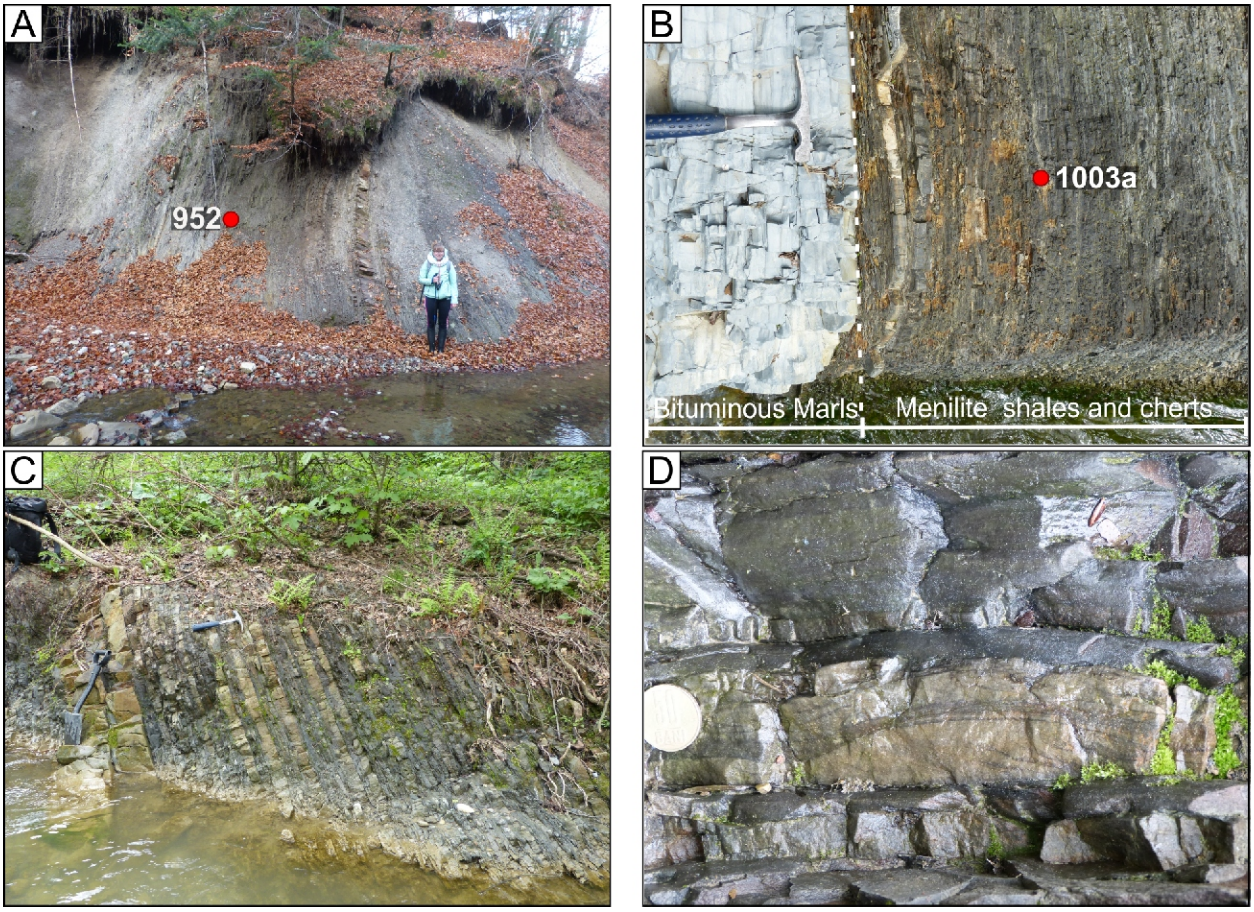

Fig. 5. Selected photographs of the Nechit River section

A – general overview of an exposure in the Nechit River section dominated by black shales and thinly-bedded cherts (red dot marks sample 952); **B** – boundary between the Menilite shales and cherts and the Bituminous Marl (red dot marks the location of sample 1003a); **C** – shale-sandstone bed packet with hummocky cross-stratification structures; **D** – sandstones with hummocky cross-stratification structures, sample 993b

AGAPIA

In the Agapia section (Romania), samples were collected from 2 exposures, which are 20 m apart. The exposures are separated by a stream, so the contact between them is not visible. In the first exposure, the Bituminous Marl was exposed in the lower part, overlain by bituminous siliceous shales with some sandstone intercalations in the upper part. At the top of the section, the proportion of thin-bedded sandstones increases. Load structures are occasionally visible in the lower parts of the sandstone beds. The Slon Beds (Oligocene–Miocene) were present in the second exposure, and are composed of sandstone-claystone breccias, primarily cemented by dark grey clays and mudstones. Menilite shale clasts have been identified within the Slon Beds ("Vorotyshcha Beds" facies). The strongly deformed clasts of Menilite shales are incorporated into a yellow matrix. Samples were collected from both the matrix and clasts to perform a comparative analysis.

PIATRA NEAMT

In the Piatra Neamt area (Romania), samples were collected from 4 exposed sections. The Bituminous Marl was present in the first (PN KG) and the second exposure (PN SPK). Marls characterized by horizontal laminations, with fish skeletons and scales, are interbedded with brown carbonate-rich shales. The third exposure (PN PM) represents mainly monotonous brown siliceous shales typical of the Menilite Beds. The last exposure (PN PK) is dominated by grey shales and thin-bedded sandstones.

NECHIT

Fieldwork in Nechit was focused on sedimentological analysis and sampling along the Nechit stream. Due to intense regional folding and the partly inaccessible nature of the Menilite Beds here it was not possisble to create a single log (Fig. 3). In exposure 949, cherts are exposed in the lower part and overlain by black and brown shales, which are interbedded with thin- and medium-bedded sandstones. In the upper part of the exposure, slumped deposits occur. The Menilite Beds, in exposures 952, 1000, 1001, 960 and 962, are represented only by brown shales. In exposures 961 and 991 siliceous bituminous brown marls (Bituminous Marls) were identified. In exposure 961, the sample was taken from the upper part of a marl package.

Exposure 971 is characterized by a reversed stratigraphic order, as shown by load structures. The lower part is dominated by dark shales, which are intercalated with locally horizontally laminated sandstone beds. In the upper part, thick-bedded massive sandstones and conglomerates with erosional boundaries occur. In one of these, chert and sandstone pebbles were identified.

Table 2

Classification of sedimentary organic matter (SOM; modified after Tyson, 1993, 1995; Roncaglia, 2004; McArthur et al., 2016)

Group	Subgroup	Category	Characteristics/Example	
Structured	Palynomorphs	Organic-walled marine phytoplankton	Dinoflagellate cysts	Resting cysts produced by Dinoflagellata *sensu* Williams et al. (1998)
		Other microplankton	*Botryococcus*	Brackish-water tolerant freshwater colonial green algae *Botryococcus* sp.
			Other microplankton	e.g., Quadriflagellate, spherical algae *Tasmanites* sp., colonial freshwater algae *Pediastrum* sp., prasinophyta (*Pterospermella* sp.) and *Campenia* sp. (assigned to algae, but with unclear taxonomic affinity, due to smooth walls and lack of morphological features, i.e. appendages, pores or laminated walls Tyson, 1995)
		Sporomorphs	Saccates	Saccate pollen (pollen grains with buoyancy sacs)
			Other non-saccate pollen and spores	Megaspores and miospores, non-saccate pollen; variable size, thickness and ornamentation
	Phytoclasts	Macrophyte plant debris	Cuticle	Leaf and plant membranous tissue
			Translucent wood	Brown, biostructured wood, cortex
			Opaque wood	Black, biochemically oxidised wood
Unstructured	Amorphous organic matter	Bacterially or phytoplankton derived amorphous organic matter; diagenetic amorphous products of macrophyte tissues	AOM	All particulate organic components which appear structureless at the scale of light microscopy (single particle of AOM was determined as every piece of AOM in which the minimum size is >15 μm)
		Higher plant secretions	Unstructured lithified resin	Intra-/extra-cellular resins; mainly dark orange, conchoidal

Exposures 977 and 1008 are dominated by black and brown shales, which are in places interbedded with mudstone debris-flow deposits with synsedimentary folds. Exposure 993 is dominated by sandstones with flaser bedding ripple cross-lamination or hummocky cross-stratification and black/brown shales. In Exposure 1000, a sharp contact between the cherts and shales (the lower part of the log) and the Bituminous Marls (upper part) is clearly visible.

PALYNOFACIES ANALYSIS

SMILNO

In Smilno (Fig. 4C), palynofacies in five samples are characterized by a very high abundance of AOM (93.2–99% of SOM) and a relatively low abundance of opaque phytoclasts (1.0–6.1%; Table 3 and Fig. 6C). The AOM is non-fluorescent. The minimum AOM abundance (81.7%) is observed in one sample (S16) with the maximum abundance of opaque wood particles (18.3%). In general, non-fluorescent, granular, and blocky fragments dominate the AOM in this exposure. *Amorphous phytoclasts*, characterized by having a surface 50% intact, were identified in the samples: these indicate an intermediate stage in their conversion to AOM. The prasinophytae alga *Tasmanites* sp., with a dark brown hue, was observed in sample S16.

DARA PRISLOP

From Dara Prislop, palynofacies in 16 samples are analysed. AOM (80.6–99.7%) constitutes a majority of the SOM, with opaque phytoclasts as a minor component (0.3–17.8%) (Table 3 and Fig. 6A–C). The AOM is dominated by granular, dark brown or black fragments in all samples. AOM may reach considerably larger sizes (reaching ~700 μm) with more variable shapes relative to the samples from Smilno. The AOM is non-fluorescent. No lithological dependence was noted in this sample suite. Sample DR 4A differs from the other samples due to the presence of a cuticle content (1.7%) characterized by dark brown colours.

AGAPIA

From Agapia, palynofacies in five samples were analysed. Samples collected from the Menilite Beds (A 1041 from the Bituminous Marls/Dynów Marl, A 1040a and A 1040b from the dark brown bituminous shales), are characterized by high AOM abundances (80.5, 82.4 and 99.4%, respectively). The palynological assemblages from the shale are more diverse than the organic matter in the Dynów Marl: seven SOM components occur in the shale samples, while only two are found in the Dynów Marl samples. Dinocysts in sample A 1040a are limited to *Caligodinium* sp. and the Middle Jurassic dinoflagellate cyst *Nannoceratopsis* sp., suggestive of reworking. UV light analysis revealed variable fluorescence types (brown and dark orange), indicating the presence of two AOM types in different preservational states. The darker one may be reworked. Sample A 1040b is characterized by a more diverse dinoflagellate cyst assemblage, including *Glaphyrocysta* sp., *Rhombodinium* sp., *Chiropteridium* sp., *Caligodnium* sp., *Wetzeliella symmetrica*, and *Deflandrea phosporitica* and AOM is also more homogeneous relative to sample A 1040a. Examination in UV light showed that the palynomorphs have dark yellow and orange fluorescence colours. Additionally, UV fluorescence demonstrated the presence of freshwater algae: namely, *Botryococcus* sp., *Pediastrum* sp. and *Pterospermella* sp. (Fig. 6N), which were not seen in transmitted light.

Sample A 1042cz, collected from clasts within the Slon Beds, is dominated by dark orange AOM with weak or no UV fluorescence. The dinoflagellate cysts in this sample are rare and char-

Table 3

Relative abundances of SOM recognized in this study

Sample	Lithology	*Botryococus* sp.	Cuticle	Other non-saccate pollen and spores	Other microplankton	Saccate pollen	Resin	Translucent wood	Opaque wood	AOM	Dinoflagellate cysts
DP 10	black shale	0.0	0.0	0.3	0.0	0.0	0.0	0.3	4.0	95.4	0.0
DP 9B	grey mudstone	0.0	0.0	0.0	0.0	0.0	0.0	0.3	6.9	92.8	0.0
DP 9A	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.3	12.8	86.9	0.0
DP 8B	black mudstone	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.3	99.7	0.0
DP 8A	grey shale	0.0	0.0	0.0	0.0	0.0	0.0	0.0	11.9	88.1	0.0
DP 7	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.0	2.9	97.1	0.0
DP 6	black shale	0.0	0.0	0.3	0.0	0.0	0.0	0.7	8.0	91.0	0.0
DP 5C	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.0	6.4	93.6	0.0
DP 5B	grey shale	0.0	0.0	0.0	0.0	0.0	0.0	0.3	7.3	92.4	0.0
DP 5A	grey mudstone	0.0	0.0	0.0	0.0	0.0	0.0	0.0	6.9	93.1	0.0
DP 4B	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.6	98.4	0.0
DP 4A	grey shale	0.0	1.7	0.0	0.0	0.0	0.0	0.0	16.8	81.5	0.0
DP 3B	grey shale	0.0	0.0	0.0	0.0	0.0	0.0	1.6	17.8	80.6	0.0
DP 3A	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.3	4.8	94.9	0.0
DP 2	grey mudstone	0.0	0.0	0.0	0.0	0.0	0.0	0.0	17.5	82.5	0.0
DP 1A	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.3	6.6	93.1	0.0
S 2	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.3	98.7	0.0
S 12	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.0	99.0	0.0
S 16	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.0	18.3	81.7	0.0
S 24	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.0	3.1	96.9	0.0
S 30	black shale	0.0	0.0	0.0	0.0	0.0	0.0	0.6	6.1	93.2	0.0
N 1008b	laminated limestone	0.0	0.0	0.0	0.0	0.0	0.0	1.9	0.0	97.8	0.3
N 1008a	laminated limestone	0.0	0.0	0.9	0.0	1.8	0.0	0.3	0.3	96.7	0.0
N 1003a	black shale	0.0	0.0	0.0	0.6	1.3	0.0	0.6	0.3	97.1	0.0
N 1001	black shale	0.0	0.0	0.3	0.0	0.9	0.0	3.1	0.0	90.3	5.3
N 1000	black shale	0.0	0.3	9.6	0.0	10.4	0.3	12.8	10.1	43.5	13.0
N 993b	black shale	0.0	0.0	0.3	0.0	0.0	0.0	1.7	0.0	93.4	4.7
N 991	bituminous marl	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	100.0	0.0
N 977	black shale	0.0	0.0	0.3	0.0	1.0	0.0	0.6	6.1	88.1	3.8
N 971	brown shale	0.0	0.0	1.6	0.0	2.3	1.6	3.2	2.6	75.7	12.9
N 962	brown shale	0.0	0.0	0.3	0.0	0.0	0.0	0.0	0.0	99.4	0.3
N 961	bituminous marl	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	100.0	0.0
N 960	brown shale	0.0	0.0	0.9	0.0	1.9	0.0	0.0	0.3	96.0	0.9
N 952	brown shale	0.0	2.0	9.3	0.0	11.3	0.0	3.0	1.7	71.7	1.0
N 949	grey siltstone	0.0	0.0	1.0	0.0	1.3	0.3	1.6	3.5	92.3	0.0
PN KG	bituminous marl	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.3	98.7	0.0
PN SPK	bituminous marl	0.0	0.0	0.0	0.0	0.0	0.0	4.8	9.6	85.5	0.0
PN PM	shale	0.0	0.0	0.0	0.0	0.0	0.0	2.5	3.1	94.4	0.0
PN PK	shale	1.2	0.0	0.0	0.0	0.0	0.0	0.0	0.9	97.8	0.0
A 1040b	shale	0.3	0.0	6.8	0.0	1.5	0.9	7.1	0.3	80.5	2.7
A 1040a	shale	1.0	0.0	4.2	0.3	8.7	0.6	1.3	1.0	82.4	0.6
A 1041	marl	0.0	0.0	0.3	0.0	0.3	0.0	0.0	0.0	99.4	0.0
A 1042j	breccia matrix	0.0	0.0	0.9	0.0	0.3	0.0	7.8	27.8	29.9	33.4
A 1042c	clast from breccia	0.0	0.0	0.3	0.0	0.0	0.0	0.0	0.7	98.7	0.3

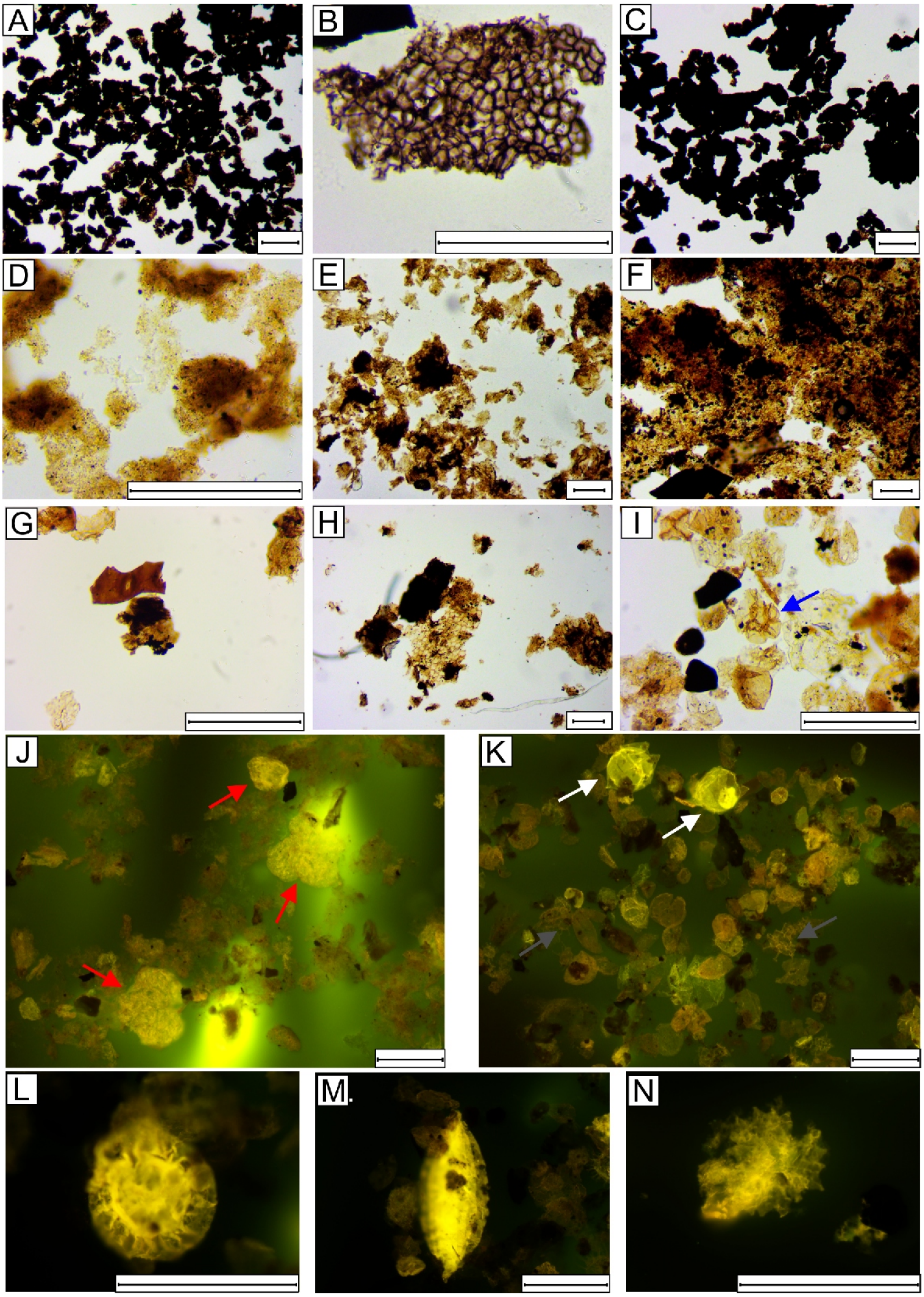

Fig. 6. Palynofacies, phytoclasts and palynomorphs from selected samples

A – palynofacies composed of black AOM and opaque phytoclasts, sample DP 5C; **B** – dark brown cuticle, DP 4A; **C** – organic material characterized by the dominance of black AOM, sample S 2; **D** – palynofacies composed mainly of pale AOM, sample PN KG; **E** – palynological material characterized by dominance of yellow AOM, sample N 962; **F** – organic material characterized by the dominance of yellow AOM with high pyrite concentrations, sample N 952; **G** – translucent phytoclast with sharp edges, sample N 977; **H** – cuticle (in centre), sample N 993b; **I** – foraminiferal linings (blue arrow), sample N 1000; **J** – organic material characterized by a great abundance of the freshwater colonial algae *Botryococcus* sp., sample PN SPK; **K** – palynological material characterized by palynomorphs with different degrees of preservation, white arrows indicate dinoflagellate cysts identified as *Deflandrea phosporitica*, with bright yellow fluorescence; grey arrows indicate dinoflagellate cysts with orange fluorescence, sample N 1000; **L** – *Pterospermella* sp., sample N 1000; **M** – *Campenia* sp., sample N 1000; **N** – *Pediastrum* sp., sample A 1040a; scale bar on all photographs – 100 µm acterized by a very poor state of preservation, often hampering identification. The sample includes *Enneadocysta pectiniformis*, *Charlesdowniea coleothrypta*, *Deflandrea phosporitica*, *Spiniferites* sp. and *Cordosphaeridium* sp. Examination in UV light showed that the palynomorphs have orange fluorescence. By contrast, in sample A 1042j collected from the sediment matrix, dinoflagellate cysts are the most abundant components of the SOM (Table 3). The sample yielded *Enneadocysta pectiniformis*, *Membranophoridium aspinatum*, *Deflandrea phosporitica*, *Homotryblium tenuispinosum*, *Achomosphaera* sp. and *Spiniferites* sp. Under UV light, all cyst specimens exhibited similar fluorescence colours (Fig. 7I, J).

PIATRA NEAMT

From Piatra Neamt, palynofacies in 4 samples were analysed. The SOM from the samples collected at Piatra Neamt is dominated by AOM (85.5–98.7%), with a secondary phytoclast component. Translucent (max. 4.8%) and opaque (max. 9.6%) wood was identified in all four samples. Sample PN PK differs from other samples due to the presence of *Botryococcus* sp. (1.2%), which was observed in transmitted light. In translucent light, the AOM is characterized by yellow and orange colours. Analysis under UV light revealed an additional freshwater algae taxon, *Pediastrum* sp., in the samples. In sample PN SPK, a relatively high abundance of *Botryococcus* sp. was also observed (Fig. 6J).

NECHIT RIVER

From Nechit, palynofacies in 14 samples were analysed. Most samples from the Nechit River section are characterized by a larger degree of variability than in other sections, with AOM as the dominant component (43.5–100%). In transmitted light, most of the AOM shows bright colours, with pyrite present. Samples where AOM was the only type of SOM (N 961 and N 991) were collected from the Dynów Marl. UV light analysis showed that the AOM in these two samples has weak or absent fluorescence and a structureless nature. Therefore, it is not possible to identify primary elements present in the AOM. In some other samples, UV analysis revealed the presence of various components hosted within the AOM: sporomorphs (sample N 952; Fig. 7A) and palynomorphs (sample N 1000). Analysis of organic matter under UV light also allowed for the identification of freshwater algae in the following samples: N 1000 (*Botryococcus* sp., *Pterospermella* sp., *Campenia* sp.; Fig. 6L, M), N 1003a (*Botryococcus* sp.) and N 1008a (*Botryococcus* sp.).

Samples N 971 and N 1000 are characterized by a higher abundance of dinoflagellate cysts (12.9 and 13%, respectively) relative to other samples. The dinoflagellate cysts in these two samples are poorly preserved and are possibly reworked [sample N 971: *Areosphaeridium diktyoplokum* (Fig. 7M) and *Cerodinium wardenense* (Fig. 7D, E); sample N 1000: *Areosphaeridium diktyoplokum* (Fig. 7G)]. Sample N 993b, collected above a bed with typical combined flow structures, including hummocky cross-stratification (Fig. 5C, D), contains cuticle fragments (Fig. 6H). In samples N 949, N 971, N 1000 resin was also identified.

INTERPRETATION

In spite of the large present-day distance (100 km) between the Slovakian Menilite Beds exposures (Dara Prislop and Smilno), their organic matter has similar properties. In all samples, the AOM is dominated by granular, dark brown or black aggregates. Opaque phytoclasts are the second most common component of the SOM (Fig. 5). The black and brown colours of AOM, cuticles, and *Tasmanites* sp. imply a high degree of thermal maturation (i.e., post-mature source rock). The results from Slovakia are probably related to the relatively high geothermal activity in the region, known as the Carpathian Conductivity Anomaly (Majcin et al., 2014). This high heat flux was associated with a deep-seated fault system on the margin of the European Platform (Kučarič et al., 2013; Majcin et al., 2014). This fault system is thought to be responsible for the formation of Cenozoic volcanic rocks and active heat transfer through fluid mobilization, which additionally may increase thermal maturity (Majcin et al., 2014).

The SOM assemblages from the selected Romanian exposures are characterized by high AOM abundances. In contrast to the AOM from the Slovakian sections, their AOM is pale yellow to orange, with the common presence of pyrite. The samples from Romanian sections are also characterized by a larger diversity of organic matter components (in some samples, seven component types are present, of both marine and terrestrial origin) than the samples from Slovakia. Together, this suggests that the Slovakian Menilite Beds were deposited in the centre of the basin, at a greater distance from the organic matter source area than the Vrancea Menilite Beds (Romania). In contrast, the results from Nechit and Piatra Neamt suggest that the Vrancea Menilite Beds were deposited in the marginal part of the basin. An AOM-palynomorph-phytoclast ternary plot suggests that the Menilite Beds were deposited under distal, suboxic-anoxic conditions (Fig. 8). However, the presence of cuticle in these samples is indicative of a relatively proximal depositional setting (Tyson, 1993). This is due to the leaf origin of cuticle, which cannot be transported for long distances because it is extremely prone to mechanical degradation and bacterial decomposition, causing it to rain out of the water column (Spicer, 1991). As an exception, cuticle particles can occur in deep-water environments when they are funnelled down submarine canyons (Shepard, 1964; Cross et al., 1966; Schnyder, 2017). Within the Menilite Beds, the Cergowa Beds are considered to be submarine fan deposits (e.g., Pszonka and Wendorff, 2017). This lithostratigraphic unit was analysed in the Dara Prislop exposure, where typical turbidity current structures were identified, including graded intervals, load structures, convolute bedding, tool marks, and flute marks. In sample DP 4A, from grey sandy mudstones in the lower part of the package characterized by normal grading, cuticle fragments were observed. The preservation of these easily degraded plant remains may have been due to rapid deposition and burial in turbiditic sediments.

In the Vrancea Unit, cuticle fragments were also identified. Moreover, typical submarine fan structures were not observed in this region. Therefore, the deposition of the Menilite Beds in the Nechit River area is unlikely to have occurred in a deep-water setting, because cuticle fragments, which are prone to bacterial decomposition, causing them to rain out of the water column, are present in samples.This is supported by the lithology of the Nechit River exposure: namely, the presence of hummocky cross-stratification structures generally interpreted as indicative of storm wave base conditions (Duke et al., 1991; Dumas and Arnott, 2006; Fig. 5D). Moreover, storm conditions can introduce and preserve plant fragments through rapid transport and provide good burial conditions (Spicer, 1980). Such conditions are probably documented in sample N 993b (Fig. 5D), in which cuticle fragments were common. Tyson (1993) suggests that similar organic matter assemblages, with domination of AOM and low percentages of palynomorphs, may

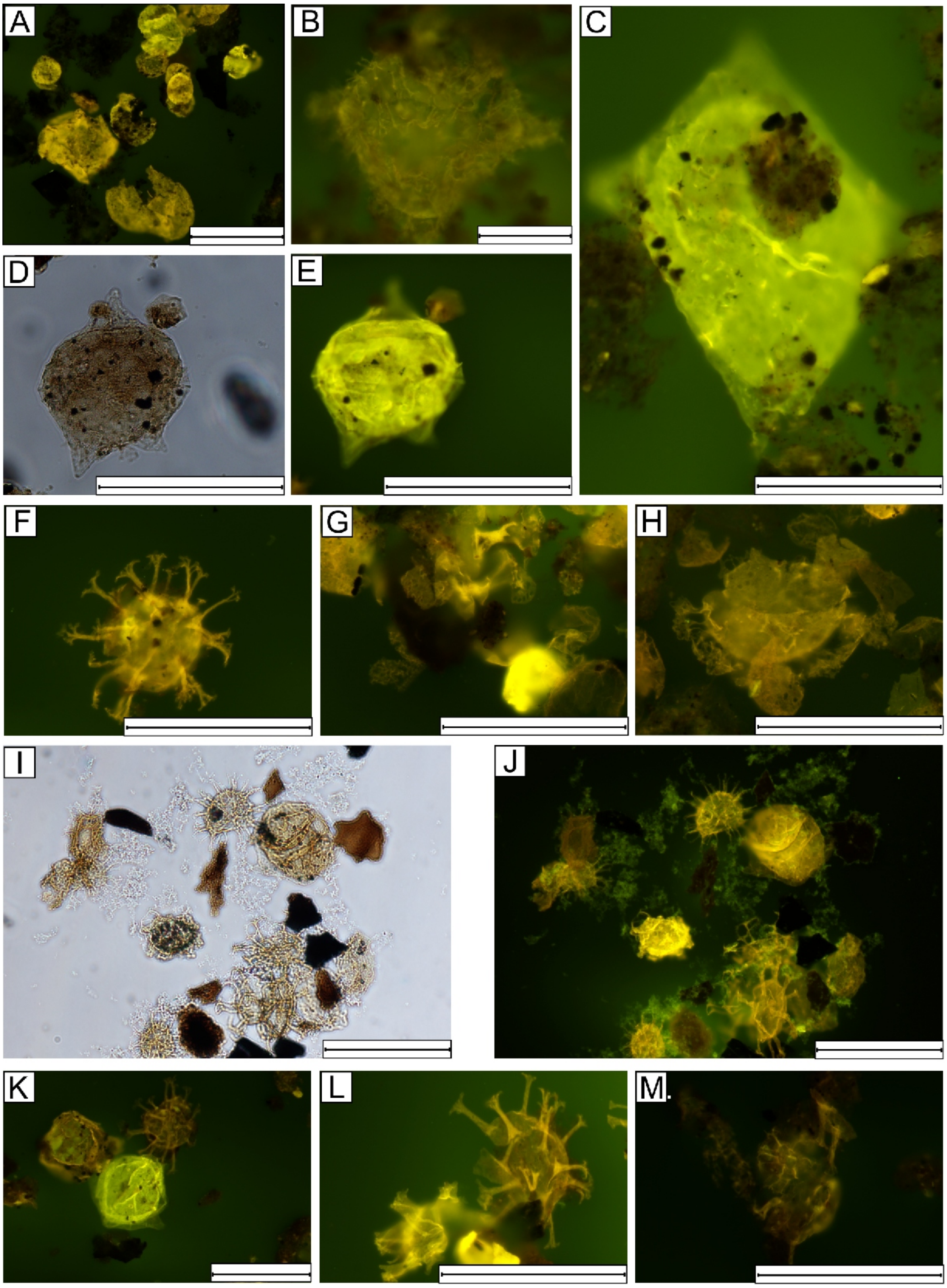

Fig. 7. Palynomorphs and sporomorphs from the Menilite (A–H, K–M) and Slon beds (I–J)

A – bisaccate pollen grains, sample N 952; **B** – *Wetzelliela articulata*, sample N 962; **C** – *Rhombodinium freienwaldense*, sample N 960; **D**, **E** – *Cerodonium wardenense*, N 971; **F** – *Hystrichosphaeridium tubiferum*, sample N 1000; **G** – *Areosphaeridium diktyoplokum*, sample N 1000; **H** – *Glaphyrocysta* sp., N 1000; **I** – dinoflagellate cysts from the Slon Beds in transmitted light, sample A 1042j; **J** – dinoflagellate cysts from the Slon Beds in UV light, sample A 1042j; **K**, **L** – dinoflagellate cyst with two styles of fluorescence, sample N 971; **M** – *Reticulatosphaera actinocoronata* and *Areosphaeridium diktyoplokum*, sample N 971; scale bar on all photographs – 100 µm

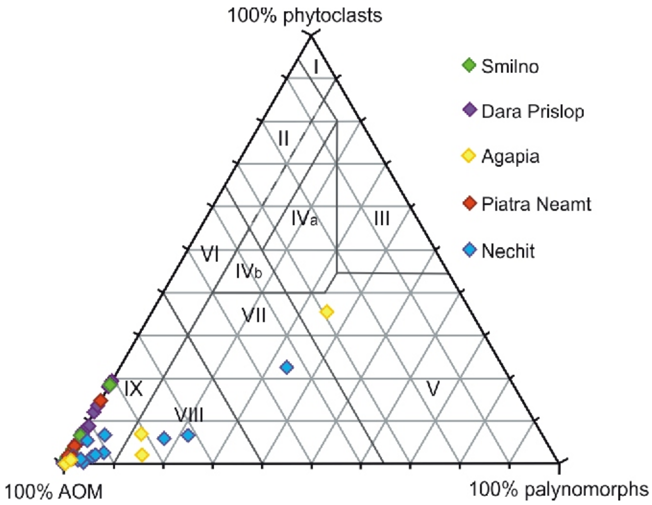

Fig. 8. AOM-palynomorph-phytoclast ternary plot (after Tyson, 1989)

Palynofacies fields: I – highly proximal shelf or basin, II – marginal dysoxic-anoxic basin, III – heterolithic oxic shelf ("proximal shelf"), IV – shelf to basin transition, V – mud-dominated oxic shelf ("distal shelf"), VI – proximal suboxic-anoxic shelf, VII – distal dysoxic-anoxic "shelf", VIII – distal dysoxic-anoxic shelf, IX – distal suboxic-anoxic basin

also be characteristic of stratified shelf seas. This interpretation is supported by geochemical investigations (Kotarba et al., 2013; Sachsenhofer et al., 2015; Bojanowski et al., 2018). Water column stratification might have been induced by basin isolation and subsequent freshwater influx, as in the case of the modern Black Sea (e.g., Murray et al., 1989). As a result, zones with reduced salinity and large-scale freshwater algae growth might have occurred in proximal settings within the basin.

Notably, freshwater algae have been identified in the Romanian sections. In the Vrancea Unit (Romania), the green algae *Botryococcus* sp. and *Pediastrum* sp. and the prasinophytes *Pterospermella* sp. and *Campenia* sp. are found, whereas in Smilno (Slovakia) only the prasinophyte *Tasmanites* sp. occurs. *Botryococcus* comprises a group of colonial green algae abundant in freshwater and brackish settings (e.g., lakes, seas, ponds; Metzger and Largeau, 2006; Mendonça Filho et al., 2012). Mobile mats with thicknesses of up to several centimetres and areas of hundreds of square metres may be formed during *Botryococcus* blooms (Guy-Ohlson, 1992). As a typical freshwater colonial form, *Botryococcus* may be transported by rivers to basins (Caratini et al., 1983; Guy-Ohlson, 1992), where it can co-occur with marine palynomorphs. Representatives of the extant genus *Pediastrum* mainly occur in freshwater environments: they are rarely found in brackish-water basins and these sparse appearances are related to freshwater influxes at river mouths (Komárek and Jankovská, 2001). *Pterospermella* sp. is also interpreted as an indicator of reduced surface salinity (Mudie, 1992; Below and Kirsch, 1997). In the framework of the Menilite Beds, these algae have also been identified in Poland, in the western part of the Silesian Unit.

By contrast, representatives of the genus *Tasmanites* are euryhaline, eurythermal marine algae found in a broad diversity of environments (Guy-Ohlson, 1988). Consequently, these algae can be regularly recovered from widely differing sedimentary facies (Guy-Ohlson, 1988). There are also reports that link *Tasmanites* sp. to eutrophic waters with regular and ample nutrient supply (Vigran et al., 2008).

The presence of these algal genera points to the existence of zones with significantly reduced salinity during the deposition of the Menilite Beds. Comparatively large variability in organic matter composition, and the presence of typical freshwater algae (*Pediastrum* sp., *Pterospermella* sp.) and algae tolerant of low-salinity settings (*Botryococcus* sp.), were observed in the marginal Outer Carpathians (Vrancea Unit). In contrast, the internal Outer Carpathians (Dukla and Grybów units) are characterized by low organic matter variability and the presence of only *Tasmanites* sp., which has a higher tolerance to salinity than *Botryococcus* sp., *Pediastrum* sp. and *Pterospermella* sp. The presence of those diagnostic algae and sedimentological observations suggest that the deposition of the Menilite Beds in the Vrancea Unit occurred in a marginal zone at a relatively short distance from the shoreline, above storm wave base. However, the deposition of the Menilite Beds at Dara Prislop and Smilno occurred farther from the shore in a more central part of the basin where salinity was higher than in its marginal zones.

Poor preservation of reworked palynomorphs (*Areosphaeridium diktyoplokum*, *Cerodinium wardenense*) may be a consequence of physical deterioration, as the result of transport processes during redeposition and the growth of pyrite crystals within. Some authors have found *Areosphaeridium diktyoplokum* in Lower Oligocene deposits (e.g., Maier, 1959; Śliwińska et al., 2012). However, poor preservation of *Areosphaeridium diktyoplokum* and co-occurrence with *Cerodinium wardenense* indicate that they are probably redeposited. UV analysis of samples from Nechit reveal differences in the fluorescence colours of palynomorphs and sporomorphs (Fig. 6K), testifying to the reworking of some palynomorphs and sporomorphs from older rocks and the presence of two distinct SOM groups with different thermal maturities (Figs. 6K and 7K, L). Additionally, samples without reworked specimens are dominated by peridinioid cysts (mainly Wetzelielloidae; Fig. 6B, C), whereas samples N 971 and N 1000, in which reworked palynomorphs were identified, are dominated by gonyaulacoid cysts (Figs. 6F, 7G, K–M). Indeed, sample N 971 contains three types of palynomorph fluorescence (yellow, orange, dark orange; Fig. 7). However, contrary to the general paradigm of UV fluorescence interpretation (Hartkopf-Fröder et al., 2015), a case was identified where older cysts (*Cerodinium wardenense*) are characterized by lighter fluorescence colours than younger cysts (e.g., *Reticulatosphaera actinocoronata*) characteristic of the Oligocene. This variability may be linked to reworking from rocks with internally diverse thermal histories. Similar fluorescence variability was observed in sample N 1000, where Eocene dinoflagellate cysts were observed; furthermore, different UV light colours were noted among sporomorphs as well (Fig. 6K). The reworking of organic particles from older rocks is likely to have caused much greater variability of SOM than in samples devoid of reworked material.

ASSESSMENT OF THE HYDROCARBON POTENTIAL OF THE MENILITE BEDS

The high thermal maturity observed in samples from the Dara Prislop and Smilno sections (Slovakia) suggests that hydrocarbons were probably generated or thermally decomposed from these parts of the Menilite Beds. The results support biomarker analysis, which suggests that the Menilite Beds in inner tectonic units are overmature (Kotarba et al., 2007; Kosakowski et al., 2018). The organic matter from the Vrancea Unit Menilite Beds is between 2.5 and 6.5 (Fig. 6B) on the Spore Colour Index Chart (Fischer et al., 1980), which indicates that it is either immature or in the initial oil window (Waples, 1985; Marshall, 1990). The high hydrocarbon potential of the Menilite Beds is reflected in the presence of abundant AOM (of marine/algal origin) and of algae (*Tasmanites* sp., *Botryococcus* sp., *Campenia* sp.), especially in the Vrancea area. All these algae – but in particular *Botryococcus* sp. and *Campenia* sp. – have an ability to synthesise and accumulate lipid substances, including hydrocarbon precursors (Metzger and Largeau, 2006). However, UV light analysis of exposures of Romanian Menilite Beds show very bright fluorescence colours, indicative of a low degree of thermal maturation. My interpretation agrees with previous geochemical investigations (Wendorff et al., 2017). This may imply that parts of the Menilite Beds with high potential for hydrocarbon generation may exist in deep structures within the Romanian Outer Carpathians, where the top of the Menilite Beds in the Vrancea Unit is located at an average depth of 2.5 km (Dicea, 1995; Popescu and Anastasiu, 2017). Kosakowski et al. (2018) envisaged a similar situation in the Boryslav-Pokuttya Unit in Ukraine, where the organic matter from exposures of the Menilite Beds is generally immature. However, the Menilite Beds buried between 3 and 6 km in the Boryslav-Pokuttya Unit are located in the oil and gas windows. This may be a sign that hydrocarbons were also generated from the Menilite Beds in the Vrancea area. To test this idea, palynofacies studies and UV light analysis should be performed on core samples collected from Menilite Beds, which exist at depth in this region.

DEPOSITIONAL ENVIRONMENT OF THE MENILITE BEDS

Studies of the Outer Carpathians have previously interpreted the flysch-like deposition of the Outer Carpathians succession as having mainly occurred in deep-marine settings (Książkiewicz, 1975; Unrug, 1979; Van Couvering et al., 1981; Leszczyński and Uchman, 1991; Kotlarczyk et al., 2006; Barwicz-Piszkorz and Rajchel, 2012; Oszczypko et al., 2015). In this framework, the Menilite Formation, in particular, was supposedly deposited in the distal parts of turbidite systems and in a pelagic environment on a continental slope, submarine ridges, and an abyssal plain associated with the development of submarine fans through time (Kotlarczyk and Leśniak, 1990; Puglisi et al., 2006; Prekopová and Janočko, 2009; Kotlarczyk and Uchman, 2012).

In contrast to this view, Baldi (1980), Van Couvering et al. (1981), and Rögl (1998) suggested that the deposition of the Menilite Beds took place in a basin isolated from the open ocean. As a consequence of this setting, periodically anoxic conditions developed, and black bituminous shales were deposited in deep-marine depositional settings (Kotlarczyk et al., 2006; Kotlarczyk and Uchman, 2012). Kotlarczyk and Uchman (2012) related anoxia at the sea bottom primarly to water-column stratification and partially to upwelling. Ichthyological analysis of the Outer Carpatian showed that different ecological groups of meso- and bathypelagic fish existed, indicating a depth in the 200–2000 m interval (Kotlarczyk et al., 2006; Kotlarczyk and Uchman, 2012). However, in the Polish (Skole Unit) and Romanian (Vrancea Unit) Outer Carpatians, several shallow-water fish taxa (e.g., flatfish), and taxa tolerant of brackish water fluxes have also been identified (Paucă, 1931; Kotlarczyk et al., 2006; Baciu et al., 2016). Based on foraminiferal assemblages, Olszewska (1985) interpreted the Menilite Beds to have been deposited between the sublittoral and the upper bathyal zones. Moreover, in the past two decades some authors have distinguished shallow and shelf areas during the deposition of the Menilite Beds, primarily on the basis of purely sedimentological investigations (Micłăuş et al., 2008, 2009; Dziadzio et al., 2016; Dziadzio, 2018; Dziadzio and Matyasik, 2018) and sedimentological observations in general geological papers (Watkinson et al., 2001; Jarmołowicz-Szulc and Jankowski, 2011).

A few studies suggested that salinity decreased during the deposition of the Menilite Beds (Kotlarczyk and Kaczmarska, 1987; Melinte, 2005; Garecka, 2012; Studencka et al., 2016). Carbonate units are characterized by the presence of brackish calcareous nannoplankton, bivalves, and the freshwater fish *Barbus* sp., as well as marine fish (Melinte, 2005; Kotlarczyk et al., 2006; Garecka, 2012; Studencka et al., 2016). Considerable freshwater influxes are also inferred in the present study. Freshwater algae – i.e., *Botryococcus* sp., *Pediastrum* sp. and *Pterospermella* sp. – were recognized in the Piatra Neamt, Agapia, Nechit, and Dara Prislop sections. However, freshwater algae were not observed in the Dynów Marl, probably because the samples were dominated by structureless AOM. Sachsenhofer et al. (2015) suggested that the MTTC ratio (0.32–0.85) indicates reduced and enhanced salinity during Bituminous Marls deposition.

Changes in sedimentary conditions within the Carpathian basin(s) in the Oligocene were driven by oscillations in relative sea level, in turn resulting from eustatic changes (Popov et al., 2010) and/or transformation of the basin as a result of regional tectonism (Jankowski and Probulski, 2011). According to these hypotheses, changes in the tectonic regime and an ongoing, multistage reconstruction of the basin resulted in a constantly variable basin morphology. The deposition of the Menilite Beds took place during the reorganization of the seaway and the extensional stage of basin development, indicated by the presence of synsedimentary faults (Jankowski and Probulski, 2011). The oldest part of the Menilite Beds was deposited in the earliest Rupelian (Olszewska, 1985). At this time, the Paratethys experienced a eustatic sea level rise followed by isolation of the basin (Baldi, 1980; Haq, 1981; Van Couvering et al., 1981; Rögl, 1998; Popov et al., 2010). Substantial freshwater input and a reduced supply of saline water drove the formation of low-salinity regions in the marginal parts of the Paratethys.

Tyson et al. (1979) suggested that stratified water columns develop in basins with high freshwater supply, reducing the efficiency of vertical mixing processes and driving the development of anaerobic conditions at the sediment-water interface. Limestone and marl horizons containing calcareous nannoplankton may represent the terminal stage in the development of anaerobic conditions (Tyson et al., 1979). In the Menilite Basin, the equivalent limestone and marl deposits are represented by the Tylawa Limestones and the Dynów Marl. Prevalent anoxic conditions during the deposition of the Dynów Marl drove an elevated concentration of fine AOM, within which specific sedimentary organic partciles cannot be identified, even under UV light. Considerable support for this notion comes from Bojanowski et al. (2018) and focuses on the coccolith limestones from the Menilite Beds: here, samples with abundant AOM are barren with regard to palynomorphs. Tyson et al. (1979) suggested that coccolith blooms are induced by high nutrient supply to the euphotic zone and shallower water depths, leading to a cyclic lithological pattern: clay (oxygenated bottom water conditions), to bituminous/oil shale (anaerobic/intermittently anaerobic bottom water conditions), to coccolith limestone (convective mixing which stimulates coccolith blooms). In this framework, the preservation of laminations in marls and limestones is indicative of hemipelagic or pelagic deposition, and large accumulations of pyrite within AOM point to deposition during the acme of anoxic conditions (Tyson et al., 1979). Sachsenhofer et al. (2015) suggested that organic matter from the Bituminous Marls in the Tazlau section, which is located ~8 km from Nechit, contains a large amount of chrysophyte algae, probably dinoflagellatas. However, based on this biomarker analysis (without specifying the species) it is not possible to determine depositional settings.

The palynofacies and sedimentology of the exposures selected in Slovakia and Romania indicate that the Menilite Beds were deposited in distinct sedimentary settings. Hummocky cross-stratification structures in the Vrancea Unit (Micläuş et al., 2009, and the present study) suggest a relatively shallow-water depositional environment. However, hummocky cross-stratification can be also found in other depositional environments, such as turbidite sequences (e.g., Mulder et al., 2009; Tinterri and Muzzi Magalhaes, 2011), shoreface-offshore (e.g., Handford, 1986), fan-delta (e.g., DeCelles and Cavazza, 1992; Uroza and Steel, 2008), river-delta (e.g., Leithold and Bourgeois, 1984; Plint and Norris, 1991), fluvial (Browne and Plint, 1994), lacustrine (Eyles and Clark, 1986), and even pyroclastic deposits (e.g., Fisher, 1990). Moreover, a variety of hummocky cross-stratification formation mechanisms exist: they can be caused by oscillatory, combined, or unidirectional flows, related to the oscillation of the wave-base location of the surface or internal waves (e.g., Myrow et al., 2002; Pomar et al., 2012). Therefore, interpretation of hummocky cross-stratification type structures should be combined with other sedimentological and/or palaeoecological records. For this reason, these structures together with cuticle fragments and fragmented and reworked palynomorphs are interpreted here as an indicator for relatively shallow-water environments, probably oscillating around storm wave-base. Additionally, the presence of cuticle fragments in samples above hummocky cross-stratification points towards a relatively short distance from the source area. Cuticle fragments were also identified in Tazlau section, which is located a short distance from Nechit (Sachsenhofer et al., 2015). At the same time, simultaneous water column stratification and anaerobic sediment-water interfaces prevailed in some parts of the basin (e.g., Bojanowski et al., 2018). In the Vrancea Unit, cherts are present. The origin of these cherts may be explained by the diagenetic transformation of diatomites – specifically, transformation of the opal within the diatom frustules (Kaczmarska and Kilarski, 1979; Krhovský, 1981b; Vetö and Hetényi, 1991). Diatomites are predominantly composed of freshwater species, such as *Melosira islandica* (Krhovský, 1981a). It is likely that the composition of diatomites is related to coastal blooms in regions where salinity was reduced. The presence of freshwater algae (*Botryococcus* sp., *Pediastrum* sp., *Pterospermella* sp.) support these conclusions.

Meyers (1997) compared the conditions of marine nearshore zones to lacustrine settings. Their common characteristics include a large supply of continental organic matter (e.g., phytoclasts, cuticle fragments, sporomorphs), relatively high sedimentation rates, and reduced salinity. Similar conditions could have potentially occurred during the deposition of the Menilite Beds, allowing for the appearance of algae typically characteristic of brackish environments and the development of depositional cycles in relatively shallow parts of the basin susceptible to depth changes, with a constant supply of terrestrial nutrients and clastic material.

In the Dara Prislop exposure, stacked fining-upwards sequences with load structures and sole marks were observed, potentially indicative of deposition during turbidite activity along horst slopes. The samples from Dara Prislop and Smilno are characterized by a low diversity of organic matter (Table 3 and Fig. 6A, C). This may be associated with fast depositional rates, resulting from turbidity current sedimentation and the preferential preservation of components resistant to mechanical degradation (e.g., opaque wood). In the Slovakian sections (Smilno, Dara Prislop), only a few terrestrial organic particles were identified (cuticle particles in DP 4A and spores in DP 10). Additionally, the presence of *Tasmanites* sp., which may occur in waters with higher salinity than *Botryococcus* sp. and *Pediastrum* sp., suggests the Dara Prislop and Smilno (Slovakia) sections were probably located farther from the source area of organic matter influx than the Vrancea area. It should be emphasized, though, that the source areas might be different as well. That is why palynofacies and sedimentological analysis should be carried out in a larger area of the Outer Carpathians. This will allow for constraints on the sediment transport directions during deposition of the Menilite Beds.

If the presence of several sedimentary basins, as opposed to a unitary basin, in the Outer Carpathians is assumed, the distribution of organic matter indicates the presence of Paratethys shallow-water zones with reduced salinity during the deposition of the Menilite Beds in the outermost basin, associated with freshwater influxes adjacent to river mouths. In the Dara Prislop and Smilno area, located in the inner basin (Dukla Basin), the low diversity of organic components indicates the absence of proximal terrestrial areas and considerable distances from the source area. However, Cergowa Beds type deposits pinpoint the presence of elevated areas, probably submarine highs.

The occurrence of several deep-water adjacent sub-basins, separated by subaqueous to subaerial elevations, would be reflected in repetitions in the nature of organic matter distribution (a decrease in organic matter component diversity and the disappearance of terrestrially derived particles with greater distance from the source area). Gągała et al. (2012) suggested that the minimum orogenic shortening of the Outer Carpathians was ~507 km. In this context, the width of the Outer Carpathians basin(s) was probably ~700 km. Therefore, the development of several deep-water basins seems unlikely.

In the semi-isolated, multiple sub-basin model, the Slovakian samples would expect a larger number and greater diversity of terrestrial particles (e.g., pollen, spores, resins, cuticle fragments, translucent wood) or reworked organic matter from submarine highs/land areas. Their absence may suggest that the deposition of the Menilite Beds took place in a large, more or less morphologically diverse basin (with potential submarine highs), with a centre in Slovakia and marginal elements in Vrancea. To test this model, integrated analysis (including sedimentological and palynofacies studies) should be carried out in units to the north of Dara Prislop and Smilno and to the west of the Vrancea Unit. This will allow a more precise reconstruction of basinal development and of the depositional settings of the Menilite Beds.

CONCLUSIONS

A detailed palynofacies analysis of 42 samples from the 23 exposures in the Menilite Beds of the Romanian (21 samples) and Slovakian (21 samples) Outer Carpathians was conducted. These samples represent various lithologies (siltstones, mudstones, limestones, marls). Additionally, two samples from the overlying Slon Beds (Agapia area in Romania) were investigated. The SOM content of the Menilite Beds from Dara Prislop, Smilno, and the Vrancea Unit areas provides insight into the regional depositional environment during the Oligocene. The following observations are of particular importance:

1. The palynofacies show great AOM abundances in all samples studied. In samples with reworked palynomorphs (e.g., dinoflagellate cysts and sporomorphs), a larger variability of organic matter components and reduced AOM content is noted.
2. UV-light analysis allowed for the identification of dinoflagellate cysts, sporomorphs, and freshwater algae (*Botryococcus* sp., *Campenia* sp., *Pediastrum* sp. and *Pterospermella* sp.), which are generally not visible in transmitted light. The presence of freshwater algae points to the existence of zones with reduced salinity during the deposition of the Menilite Beds. It is likely these zones were associated with freshwater influxes adjacent to river mouths.
3. Palynofacies results, organic matter observations under UV light, and sedimentological observations (e.g., hummocky cross-stratification) suggest that a relatively shallow-water (above storm wave base), brackish environment prevailed during deposition of those Menilite Beds presently exposed in the Vrancea Unit, which is among the marginal units of the Outer Carpathians.
4. The Menilite Beds deposits in Slovakia (Dukla Unit – internal Carpathian Unit) were deposited farther from the source area than those of the Vrancea Unit (external Carpathian Unit).
5. The presence of algae (*Tasmanites* sp., *Botryococcus* sp., *Campenia* sp.) characterized by the ability to synthesise and accumulate lipid substances, including hydrocarbon precursors, points to a high hydrocarbon potential. However, palynomorph colours in both transmitted and UV light are indicative of thermal immaturity in the Vrancea Unit. By contrast, the dark colours of palynomorphs and AOM from Dara Prislop and Smilno indicate that in the Slovakian exposures, organic matter in the Menilite Beds is characterized by a high degree of thermal maturity, related to the occurrence of the Carpathian Conductivity Anomaly zone in this region.

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