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Lower and Middle Jurassic organic-rich rocks of W and SW Ukraine, NE Romania and S Moldova: occurrence, hydrocarbon generation zones and mineralogy

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ABSTRACT

The organic-rich strata of the Lower and Middle Jurassic occur within separate blocks, covering significant areas of the Carpathian Foredeep basement, the Bârlad Depression and the Dobrogean Foredeep. The deposition of these strata resulted from an anoxic event, leading to the formation of a globally important source rock level. The Rock-Eval pyrolysis results for the Middle Jurassic rocks in the Ukrainian part of the Carpathian Foredeep basement indicated a total organic carbon content of 1.61–14.98 wt%, with type II/III kerogen at an early mature to mature stage. Based on time-temperature index calculations, the top of the oil window occurs at the depth of 2200 m in the Ukrainian part of the Carpathian Foredeep basement, and at 1000 m in both the Bârlad Depression and the Dobrogean Foredeep. The former achieved thermal maturity during the Neogene period, while the latter two entered the oil window towards the end of the Cretaceous period. Across the entire study region, the Lower and Middle Jurassic strata are situated in the oil window over extensive areas, suggesting their potential inclusion in the petroleum system. Hydrocarbons generated by the Toarcian–Bathonian organic-rich rocks may contribute to the formation of Mesozoic–Miocene accumulations in the Carpathian Foredeep basement and some oil fields in the Carpathian flysch sequence. In the Bârlad Depression and the Dobrogean Foredeep, hydrocarbon occurrences may be expected in the Upper Jurassic reservoir rocks, potentially sourced by the Middle Jurassic black shales.s

Introduction

The study area includes the territory of the Lower and Middle Jurassic black shales situated in the Carpathian Foredeep basement (Ukraine, Romania), the Bârlad Depression (Romania) and the Dobrogean Foredeep (Ukraine, Moldova; Fig. 1). During the Jurassic period, this region constituted a shallow epicontinental sea of the sub-Mediterranean realm (Pieńkowski et al. 2008). Being a part of the Central European Basin System (CEBS), it developed in the peri-Tethyan realm, showing frequent connections with the Tethys Ocean, which developed during the breakup of Pangea in the Early and Middle Jurassic times (Golonka et al. 2009). The occurrence of Jurassic organic-rich rocks is widespread in the world, and their deposition was greatly influenced by climate variations, sea level changes and anoxic events (Hesselbo et al. 2000). During the Aalenian–Bathonian times, global warming of 5 to 10 °C and a three to five times higher carbon dioxide level was experienced compared to the present (Xie et al. 2006). The warm and humid climate of the Early and Middle Jurassic, along with greenhouse effects, is evidenced by the abundance of floral remains and coal deposits in the Central European region (Chandler et al. 1992).

The main Jurassic oceanic anoxic event occurred during the Toarcian, leading to the deposition of organic-rich shales in marine settings (Kemp et al. 2005; Hesselbo et al. 2007). Oil and gas shales from this period are widespread at the middle and high latitudes of the European platform, such as the Cleaveland Basin in the UK and the Boulonnais in northern France (Mattioli et al. 2004), as well as on the northern margin of Africa, including the Tunisian basins. Moreover, Lower–Middle Jurassic black shales are reported from the Qaidam Basin in northwestern China (Qiang et al. 2002), the Jaisalmer Basin in India (Srivastave and Ranawat 2015) and central Saudi Arabia (Moshrif 1987). The Toarcian organic-rich rocks are estimated to generate 25% of the global hydrocarbon reserves (Soua 2014). The Middle Jurassic organic-

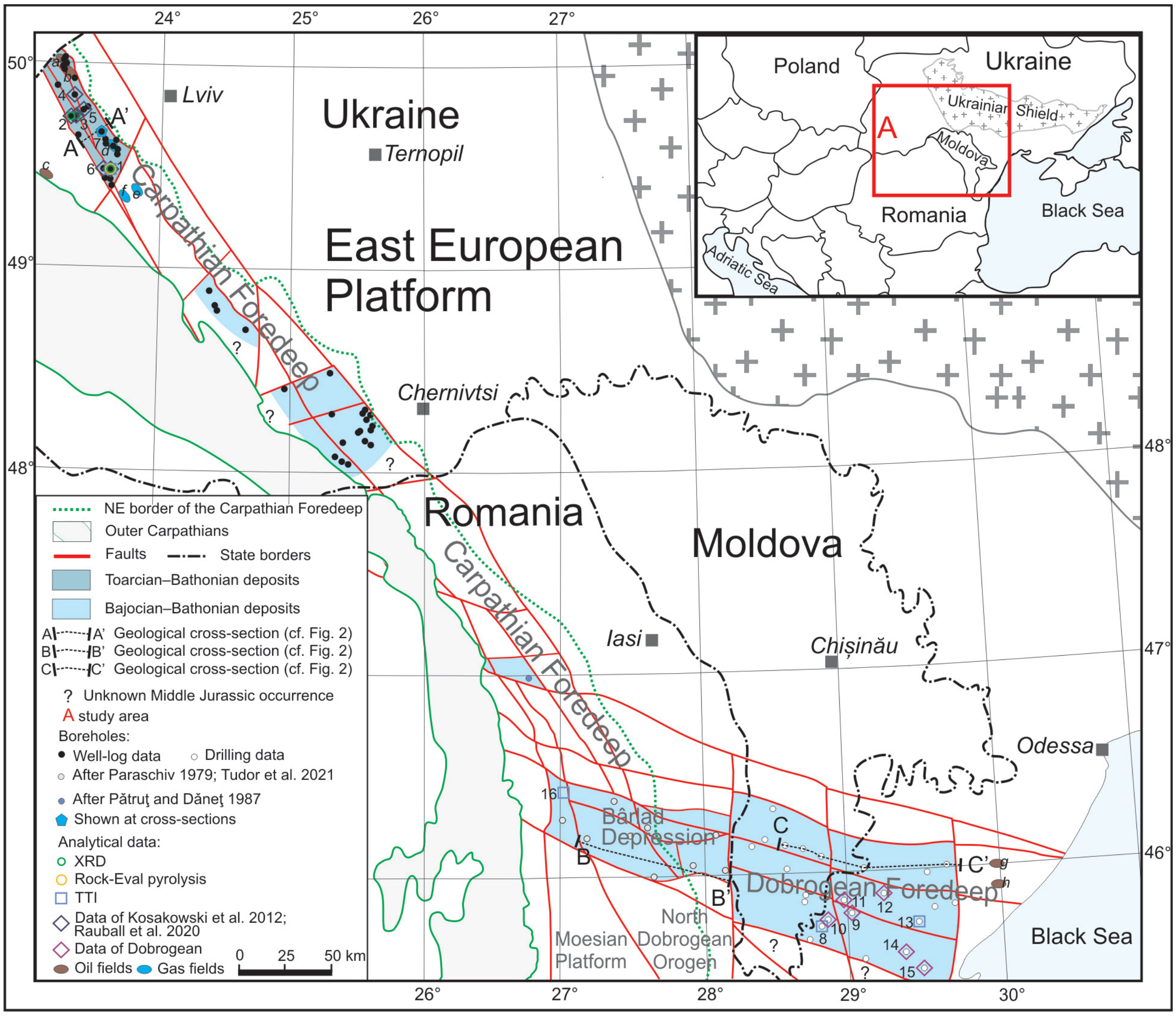

Fig. 1. Location map showing the distribution of the Lower–Middle Jurassic strata in W and SW Ukraine, NE Romania and S Moldova (geological structure after Peltz and Cazaban 1968; Bogayets et al. 1986; Popescu et al. 2016; Gnidets et al. 2003; Gerasimov et al. 2005; Pawlewicz 2007). Boreholes: 1 – Mosty-2, 2 – Korolyn-2, 3 – Korolyn-6, 4 – Bortyatyn-1, 5 – Chornokuntsi-1, 6 – Podiltsi-1, 7 – Romaniv-1, 8 – Kalcheva-3, 9 – Chervonoarmiysk-1, 10 – Chervonoarmiysk-2, 11 – Orikhiv-1, 12 – Orikhiv-3, 13 – Stari Troyany-1, 14 – Furmaniv, 15 – Kiliya-1, 16 – location of the section point for the burial diagram in the deep-seated western part of the Bârlad Depression. Oil and gas fields: *a* – Kokhanivka, *b* – Orkhovychi, *c* – Volya-Blazhivska, *d* – Rudky, *e* – Bilche-Volytsya, *f* – Letnia, *g* – Sarata, *h* – Zhovtoyarsk. Abbreviations: XRD – X-ray diffraction, TTI – time-temperature index.

rich rocks occur in the Upper Indus Basin in Pakistan (Ali et al. 2019), the Kurdistan region in northern Iraq (Abdula et al. 2015), the Danish Basin in Denmark (Nielsen et al. 2003), the Oseberg South area in the northern North Sea (Løseth et al. 2009), and the Shushan Basin in Egypt (Elsaqqa et al. 2023). These rocks represent important hydrocarbon source rocks in many regions of the world. In particular, the deposits of this age have charged the Cretaceous and Cenozoic reservoirs in Iraq, Kuwait and Iran, forming one of the world's most prolific petroleum systems (Pitman et al. 2004; Verma et al. 2004; Aqrawi and Badics 2015) with over 250 billion barrels of proven recoverable hydrocarbons (Abeed et al. 2011).

The objective of this paper is to study the globally important Lower and Middle Jurassic source rock complex in the peri-Tethian area of Ukraine, Romania and Moldova. This contribution presents the lateral extent of the Toarcian–Bathonian organic-rich rocks within this part of the CEBS, and explores their mineral composition, the spatial and depth-related changes in their thermal maturity, and the potential role of these deposits in the region's petroleum systems.

Geological setting

The study area covers the territory where the Lower and Middle Jurassic strata occur in the Carpathian Foredeep basement (Ukraine and Romania), the Bârlad Depression (Romania) and the Dobrogean Foredeep (Moldova, Ukraine), extending from the Ukrainian–Polish border to the Black Sea (Figs 1, 2). These deposits appear within subsided blocks and are absent in uplifted blocks. The deposits vary considerably in thickness and depth, indicating that their formation has been influenced by the tectonic movements of separate blocks.

In the Ukrainian part of the Carpathian Foredeep basement (Fig. 2A), the Jurassic strata occur within the Stryi Jurassic depression. They rest on the Proterozoic sequence and are overlain by the Miocene molasse. The Jurassic succession comprises the Lower, Middle and Upper Jurassic

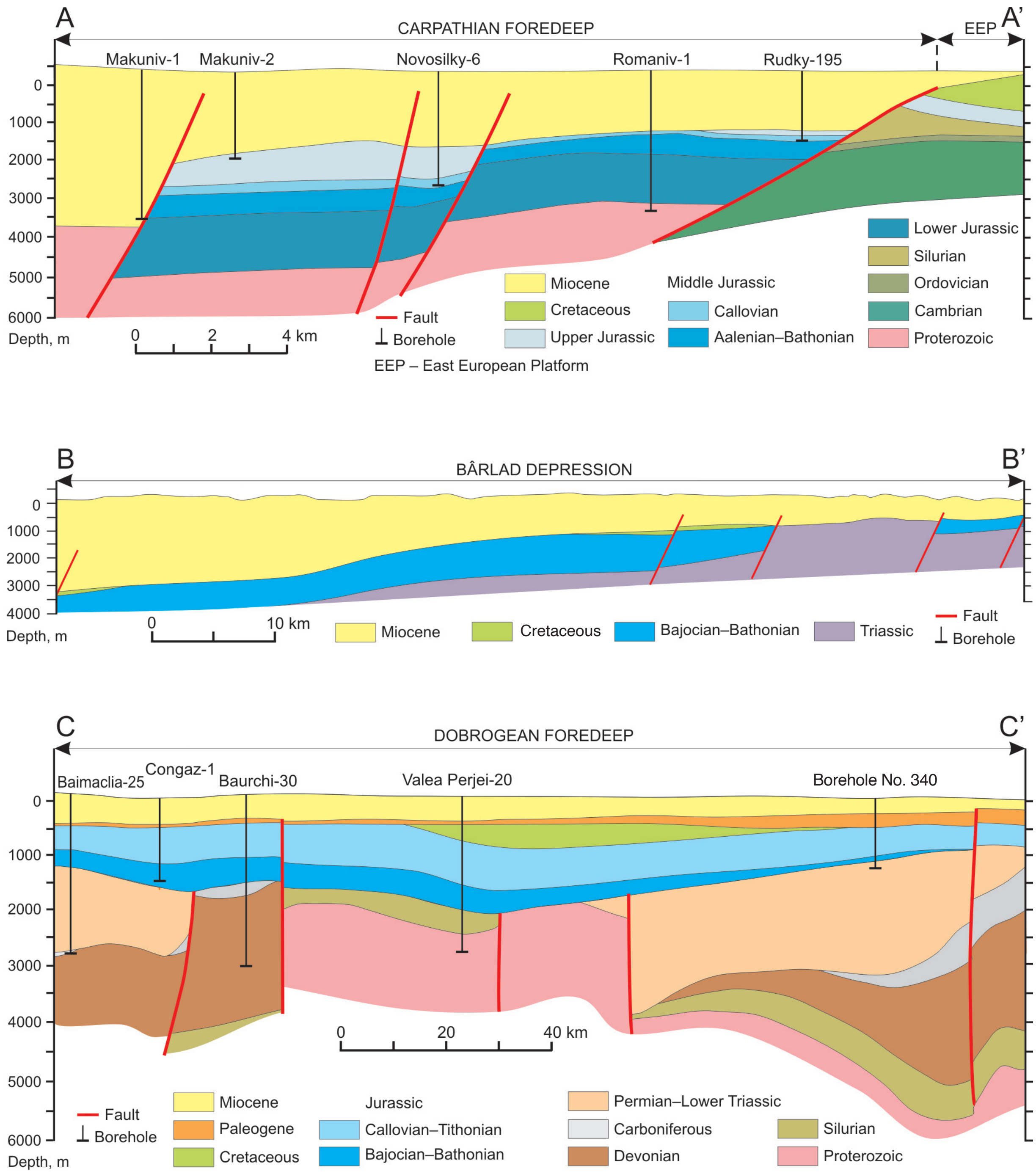

Fig. 2. Geological cross-sections (see Fig. 1 for locations): **A–A'** – through the Carpathian Foredeep (modified after Gerasimov et al. 2005), **B–B'** – through the Bârlad Depression (modified after Peltz and Cazaban 1968), **C–C'** – through the Dobrogean Foredeep (modified after Gnidets et al. 2003).

series, with a total thickness exceeding 3000 m (Gerasimov et al. 2005; Zhabina et al. 2017). In the Bârlad Depression (Fig. 2B), Jurassic rocks lie on the Triassic sequence and are also overlain by the Miocene molasse (Peltz and Cazaban 1968; Pawlewicz 2007). At some locations the Upper Jurassic sediments are overlain by Cretaceous beds. The Jurassic succession is represented here by the Middle and Upper Jurassic series, with a total thickness exceeding 2000 m. In the Dobrogean Foredeep (Fig. 2C), the Jurassic rocks in different blocks rest on Proterozoic, Silurian, Devonian, Carboniferous and Triassic sequences, and are overlain by Cretaceous, Paleogene and Neogene strata (Bogayets et al. 1986; Gnidets et al. 2003). The Jurassic succession includes the Middle and Upper Jurassic series, and at some locations also the Lower Jurassic series, with a total thickness reaching 3000 m.

The Jurassic organic-rich rocks in the study area are recognized in the top portion of the Lower Jurassic and in most of the Middle Jurassic series (Fig. 3). In the basement of the Ukrainian Carpathian Foredeep, these rocks are represented by the Kokhanivka Formation, spanning the Toarcian–Bathonian periods (Zhabina et al. 2017). They are mainly represented by organic-rich mudstones, and to a lesser extent by organic-rich limestones and sandstone layers. In the Bârlad Depression, the studied stratigraphic interval covers only the Bajocian and Bathonian strata (Peltz and Cazaban 1968). In the lower part of the Bajocian, the rocks are represented

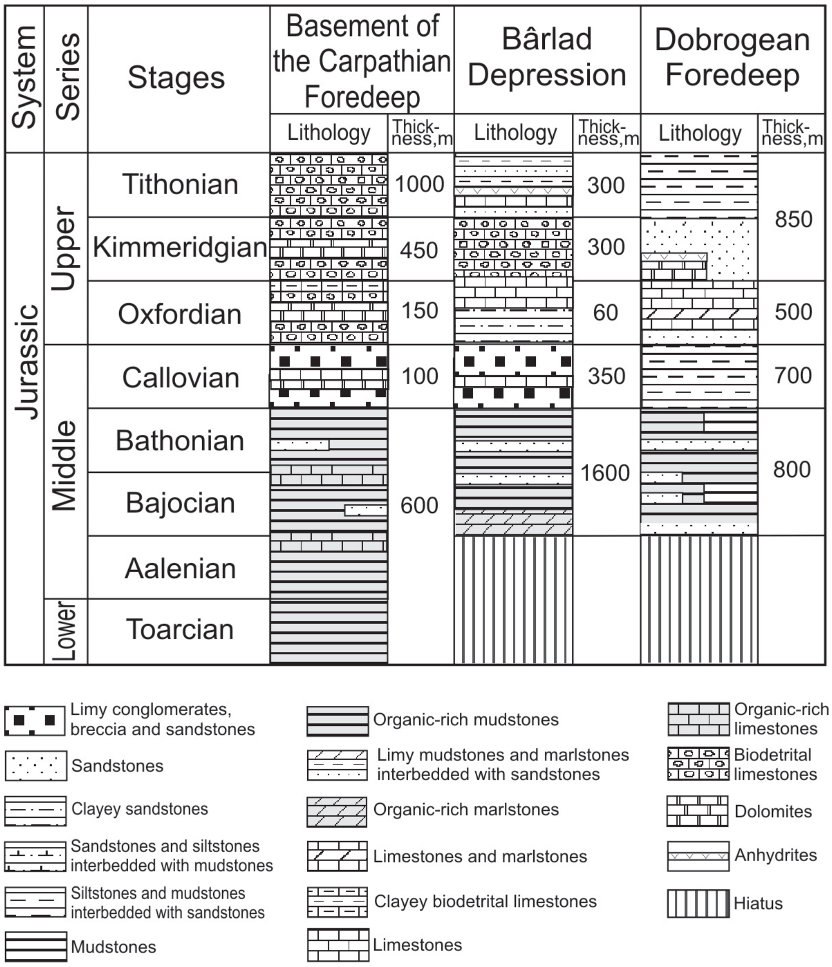

Fig. 3. Stratigraphic scheme of the Jurassic succession of the Carpathian Foredeep, the Bârlad Depression and the Dobrogean Foredeep (modified after Peltz and Cazaban 1968; Bogayets et al. 1986; Zhabina et al. 2017).

by organic-rich marlstones, while the upper part of the Bajocian and the entire Bathonian comprise organic-rich mudstones interbedded with sandstone layers. In the Dobrogean Foredeep, as well as the above-described adjacent Romanian territory, the studied sequence includes the Bajocian and Bathonian strata with the thickness of up to 800 m (Bogayets et al. 1986). These strata predominantly consist of organic-rich mudstones interbedded with sandstone layers and organic-lean mudstones.

Methods and data

The location map (Fig. 1) illustrates the lateral extent of the Lower and Middle Jurassic black shales in the study area, along with boreholes that penetrated these strata. Additionally, structural and thickness maps (Figs 4, 5) were constructed, based on the analysis of well-log and drilling data obtained from 102 boreholes spanning the entire study area. The data utilized for this analysis were sourced from the archives of the Institute of Geology and Geochemistry of Combustible Minerals of the Ukrainian National Academy of Sciences (IGGCM NAS), complemented by information from sections published by Peltz and Cazaban (1968), Popescu et al. (2016), Gnidets et al. (2003), Gerasimov et al. (2005) and Pawlewicz (2007).

Total organic carbon (TOC) and thermal maturity measurements of the Middle Jurassic black shales from six boreholes (Bortyatyn-1, Chornokuntsi-1, Mosty-2, Korolyn-2, Korolyn-6, Podiltsi-1) in the basement of the Ukrainian Carpathian Foredeep (Koltun et al. 1998; Kosakowski et al. 2012; Rauball 2020) and six boreholes (Kiliya-1, Furmaniv-3, Orikhiv-1, Orikhiv-3, Chervonoarmiysk-1, Chervonoarmiysk-2) in the Ukrainian and Moldovan parts of the Dobrogean Foredeep (Bogayets et al. 1986; Ivanova 2012) were integrated. The burial and thermal history of the Lower and Middle Jurassic black shale sequence in the Ukrainian part of the Carpathian Foredeep (Mostivska-2 borehole), the Bârlad Depression (the location of the section point for the burial diagram is shown in Fig. 1) and the Dobrogean Foredeep (Stari Troyany-1, Kalcheva-3 and Skhidna Sarata-1 boreholes) were reconstructed using burial history plots (Waples 1980, 1998). Present-day temperatures of boreholes were obtained from the archives of the IGGCM NAS. The burial history was reconstructed using data for defined events (deposition, erosion, non-deposition) along with stratal thicknesses, lithologies and temperatures. The burial and thermal history of the Mostivska-2 borehole was correlated and calibrated with Rock-Eval T_{max} data.

Nine rock samples of the Lower and Middle Jurassic black shales from three boreholes (Korolyn-2, Korolyn-6 and Mosty-2) in the Ukrainian part of the Carpathian Foredeep, which cover a wide range of present burial depths (2509–3926 m), were investigated for mineral composition, using X-ray diffraction (XRD). This study was performed in the laboratory of the IGGCM NAS in Lviv, employing an ADP-2.0 diffractometer and following standard methods (Moore and Reynolds 1997). The XRD system operated under the conditions of 34 kV, 14 mA, Mn-filtered Fe radiation, at

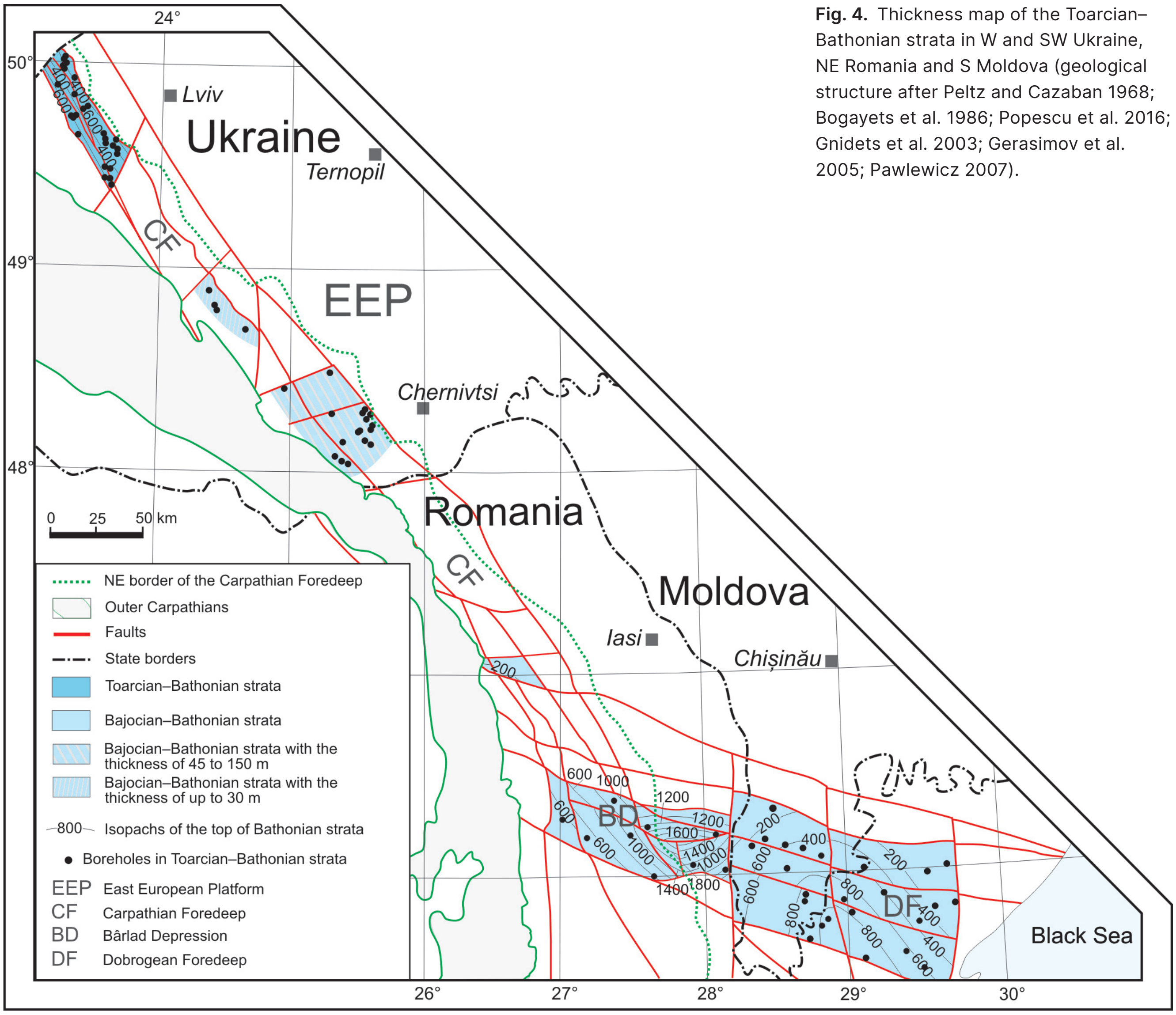

Fig. 4. Thickness map of the Toarcian–Bathonian strata in W and SW Ukraine, NE Romania and S Moldova (geological structure after Peltz and Cazaban 1968; Bogayets et al. 1986; Popescu et al. 2016; Gnidets et al. 2003; Gerasimov et al. 2005; Pawlewicz 2007).

0.025° 2Θ/step, with a counting time of 1.5 s. A mineralogical study of the samples was carried out on separate fractions (1–0.05, 0.05–0.005 and <0.005 mm) with a quantitative calculation for the whole rock. For this purpose, rock samples were disintegrated by prolonged exposure to distilled water with periodic gentle mechanical intervention (ibid.). The quantitative mineral composition in the coarser fractions (1–0.05 and 0.05–0.005 mm) was determined on the diffractograms of non-oriented powder preparations, using the Profex-8.4 program, based on the Rietveld method (Döbelin and Kleeberg 2015) of diffractogram profiles modelling.

Results and discussion

Occurrence and thickness of the Toarcian–Bathonian organic-rich strata

In the northwestern block of the basement of the Ukrainian Carpathian Foredeep, the thickness of the Toarcian–Bathonian sequence ranges from 400 to 600 m (Fig. 4). In the central and southeastern blocks, these formations are represented by Bajocian–Bathonian layers with the thickness of up to 30 and 45–150 m, respectively. The first two blocks are bounded from the northeast by the Storozhynets fault, and the southeastern block, due to the special tectonic structure of the Pokuttya basement ledge, by the Kosiv fault (Shcherba et al. 1987). On the territory of Romania, the Middle Jurassic black shales are of Bajocian–Bathonian age and occur mainly in the southeastern part of the Carpathian Foredeep basement and the Bârlad Depression, ranging from 200 to 1800 m in thickness (Peltz and Cazaban 1968; Popescu et al. 2016). The Bajocian–Bathonian organic-rich strata, with the thickness of 200 to over 800 m, are spread over almost the entire territory of the Dobrogean Foredeep (Bogayets et al. 1986; Gnidets et al. 2003).

In the Ukrainian part of the Carpathian Foredeep basement, the Lower–Middle Jurassic strata follow the dip of the East European Platform in the southwestern direction towards the Carpathians (Fig. 5). The depth of the Bathonian's top in the northwestern block varies from 1200 m in its northeastern part to over 4000 m in the southwestern part. In the central block, it changes from 1800 to over 2400 m, respectively, while in the southeastern block, it varies from 200 to over 2800 m , respectively. In the Romanian part of the Carpathian Foredeep basement, the Middle Jurassic strata dip also towards the Carpathians, with the depth of the present burial of the Bathonian's top ranging from 1400 to over

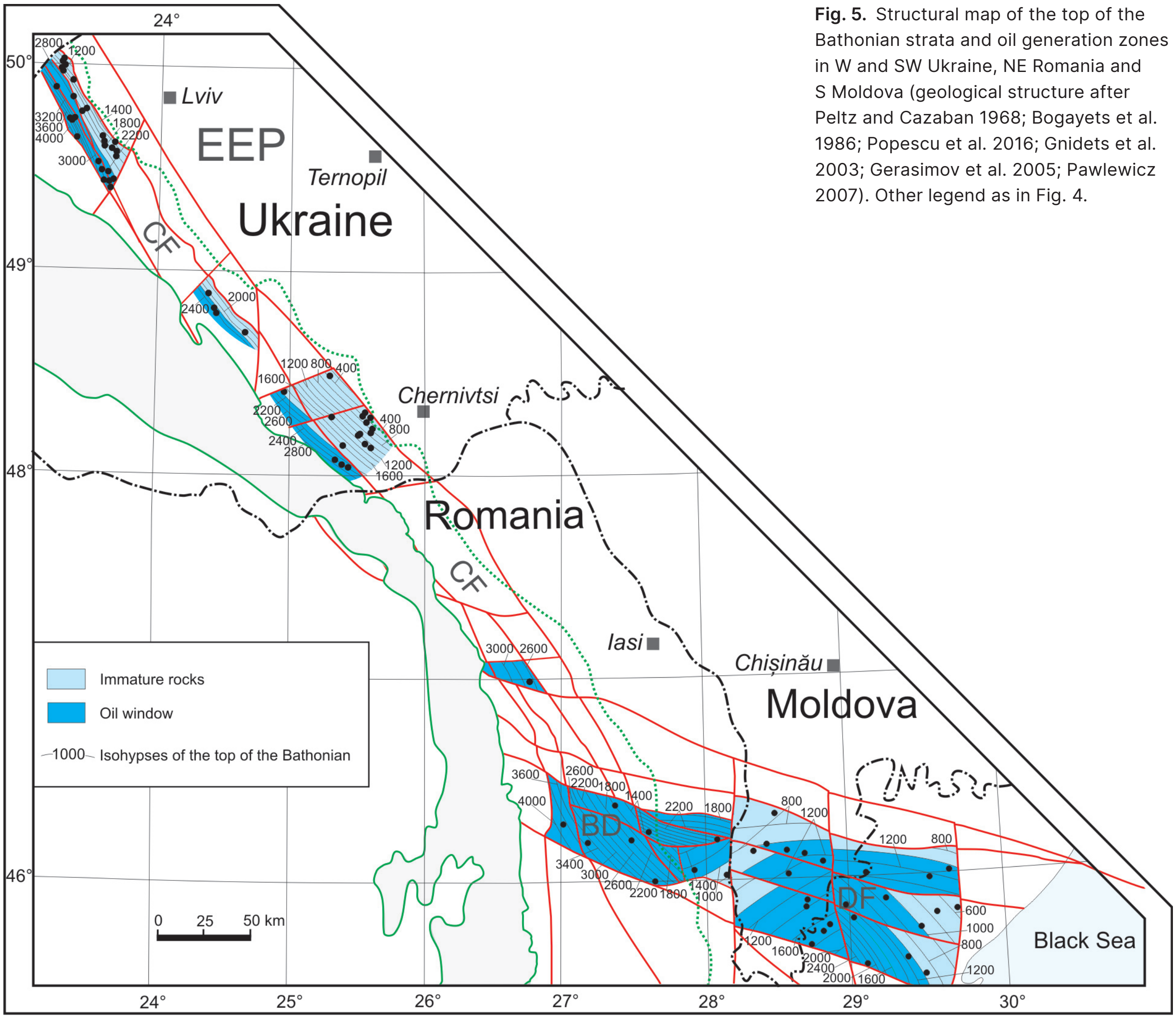

Fig. 5. Structural map of the top of the Bathonian strata and oil generation zones in W and SW Ukraine, NE Romania and S Moldova (geological structure after Peltz and Cazaban 1968; Bogayets et al. 1986; Popescu et al. 2016; Gnidets et al. 2003; Gerasimov et al. 2005; Pawlewicz 2007). Other legend as in Fig. 4.

3600 m. In the Bârlad Depression and the Dobrogean Foredeep, the depth of occurrence of the Middle Jurassic strata varies significantly in different blocks, and the depth of the Bathonian's top varies between 600 and 2400 m.

Thermal maturity and hydrocarbon generation zones

The Toarcian–Bathonian black shales in the study region were investigated in the most detail in the Ukrainian part of the Carpathian Foredeep basement. In this region, the Toarcian–Bathonian succession, which is up to 600 m thick, is predominantly composed of organic-rich mudstones and siltstones. The Rock-Eval pyrolysis results (Koltun et al. 1998) on core samples from the Mostivska-2 borehole, ranging between 2364 and 2555 m in depth, show TOC content from 1.61 to 12.79 wt% (Table 1). The hydrogen index (HI) values vary from 73 to 320 mg HC/g TOC and, taking into account the low maturation level of the rocks, indicate type II/III kerogen. T_{max} ranges from 434 to 438 °C, showing that the rocks at these depths occur in the top part of the oil window. The studies by Kosakowski et al. (2012) and Rauball et al. (2020) display similar results. Kosakowski et al. (2012) investigated the Middle Jurassic rocks from the Bortyatyn-1, Chornokuntsi-1, Mosty-2, Korolyn-2, Korolyn-6 and Podiltsi-1 boreholes within the depth range of 2096–3523 m. They reported TOC content of up to 8.3 wt%, T_{max} ranging from 412 to 454 °C (average 437 °C), vitrinite reflectance (Ro) values varying between 0.61 to 0.65% (average 0.61%), and HI values ranging from 8 to 229 mg HC/g TOC. Lower Jurassic strata from the Rudky-300 borehole within the depth range of 3241–3905 m indicate a higher maturation level, corresponding to Ro values of 1.74 to 1.99%. The data by Rauball et al. (2020) for the Korolyn-2, Korolyn-6 and Mosty-2 boreholes within the depth range of 2361–3708 m showed TOC content reaching 14.98 wt% (average 4.19 wt%) in the Middle Jurassic rocks. HI values typically around 100 mg HC/g TOC, reaching up to 242 mg HC/g TOC, indicate the predominance of type III kerogen. Thermal maturity was measured by T_{max} (432–448 °C) and Ro (0.69–0.90%). The above-mentioned studies show that the Middle Jurassic succession in the Ukrainian part of the Carpathian Foredeep basement consists of organic-rich rocks with type II/III kerogen, exhibiting good petroleum potential at an early mature to mature stage.

In Romania, the Middle Jurassic rocks are considered as one of the main source rock strata in the Bârlad Depression and the Moesian Platform (Popescu 1995; Pene 1996). The Bajocian–Bathonian succession primarily comprises black

Table 1. Rock-Eval pyrolysis results of the Middle Jurassic core samples from the Mostivska-2 borehole (Koltun et al. 1998)

Depth, m	T_{max}, °C	S_1, mg HC/g rock	S_2, mg HC/g rock	S_3, mg CO_2/g rock	PI	TOC, wt%	HI, mg HC/g TOC	OI, mg HC/g CO_2
2364	438	0.03	1.19	0.37	0.02	1.61	73	22
2522	434	0.42	31.62	0.37	0.01	12.79	247	2
2555	435	0.12	14.40	0.19	0.01	4.49	320	4

T_{max} – temperature of maximum of S_2 peak, S_1 – amount of free hydrocarbons (gas and oil) in the sample, S_2 – amount of hydrocarbons generated through thermal cracking of non-volatile organic matter, S_3 – amount of CO_2 produced during pyrolysis of kerogen, PI – production index, TOC – total organic carbon, HI – hydrogen index, OI – oxygen index

shales with TOC content of 1.2–2 wt% and type II/III kerogen. The total petroleum system is composed of Middle Jurassic rocks (Pene 1999). In the Dobrogean Foredeep, a significant part of the Bajocian–Bathonian sequence consists of organic-rich mudstones with TOC content ranging from 0.6 to 1.3 wt% (Kiliya-1, Furmaniv-3, Orikhiv-1, Orikhiv-3, Chervonoarmiysk-1 and Chervonoarmiysk-2 boreholes; Bogayets et al. 1986). The Ro values of the Bajocian–Bathonian rocks were measured in the Chervonoarmiysk-1 (0.61%) and Chervonoarmiysk-2 (0.60–0.62%) boreholes, indicating that these strata occur in the oil window (Ivanova 2012).

Burial and maturation history modelling of the Toarcian–Bathonian rocks

In the Ukrainian part of the Carpathian Foredeep basement, the modelling was performed for the Mostivska-2 borehole (Fig. 6A). The Toarcian–Bathonian organic-rich rocks here, at the depth of around 2500 m, reach the time-temperature index (TTI) value of 11, showing that the maturation level of these strata corresponds to the upper part of the oil window. The measured value of T_{max} for these rocks is 434–438 °C, matching with the calculated TTI value. This confirms that the burial history reconstruction for the strata studied in this borehole was correct. Furthermore, this modelling shows that the Toarcian–Bathonian rocks of the Carpathian Foredeep basement (Mostivska-2 borehole) remained immature until the Neogene, and only the deposition of the Miocene molasse sequence caused their rapid burial and maturation to the level of the beginning of the oil window (Fig. 6A). In the Karolina-2 and Karolina-6 boreholes, these strata occur at greater depths (3421–3708 m) and reach the deeper part of the oil window. As shown by Rauball et al. (2020), these rocks have T_{max} and Ro values of 443 °C, 0.78–0.82% and 448 °C, 0.85–0.90%, respectively. The burial and maturation history modelling allows to estimate that the Toarcian–Bathonian strata of the Carpathian Foredeep basement enter the oil window at a depth of around 2200 m.

The modelling for the Middle Jurassic strata of the Bârlad Depression (Fig. 6B) was performed for its deep-seated western part. For the case where the top of the Bajocian–Bathonian

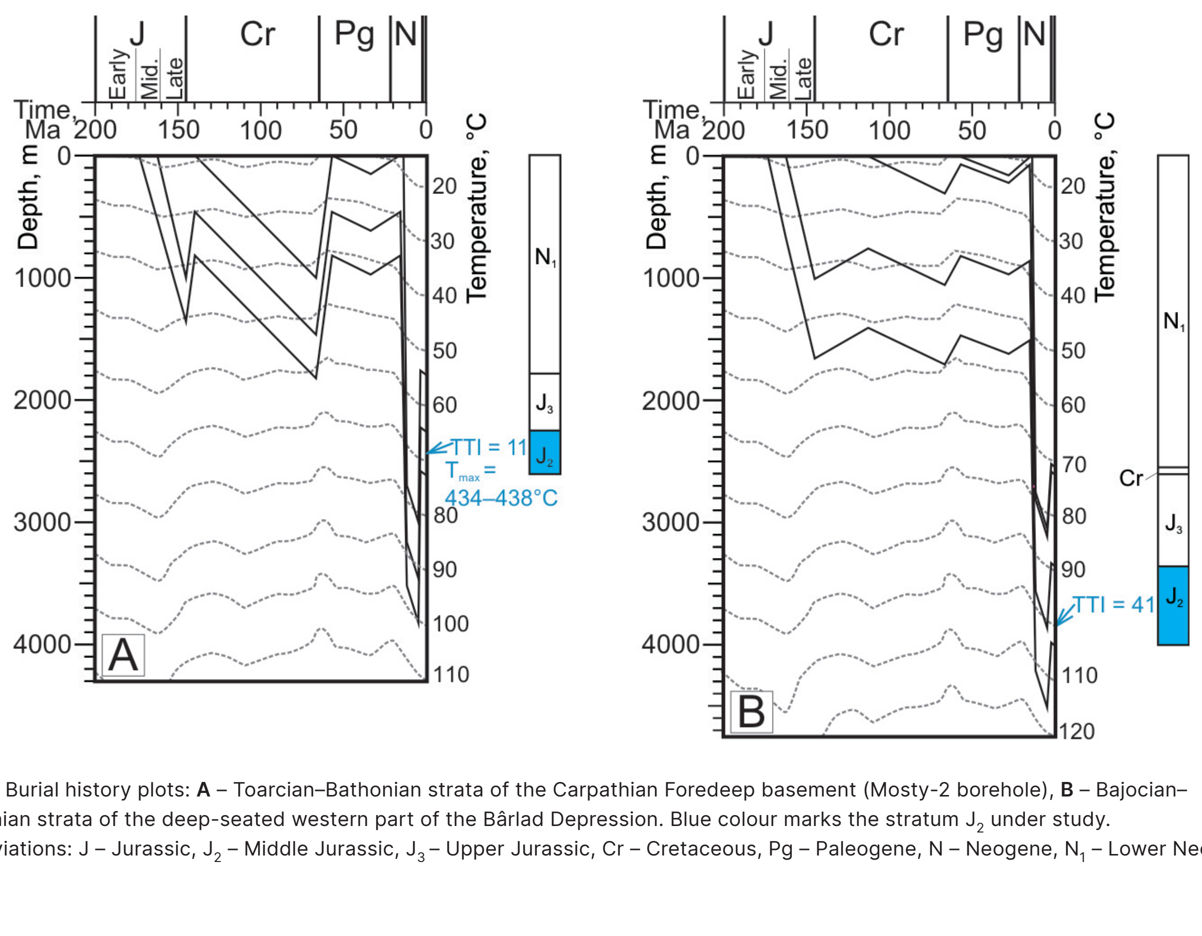

Fig. 6. Burial history plots: **A** – Toarcian–Bathonian strata of the Carpathian Foredeep basement (Mosty-2 borehole), **B** – Bajocian–Bathonian strata of the deep-seated western part of the Bârlad Depression. Blue colour marks the stratum J_2 under study. Abbreviations: J – Jurassic, J_2 – Middle Jurassic, J_3 – Upper Jurassic, Cr – Cretaceous, Pg – Paleogene, N – Neogene, N_1 – Lower Neogene.

sequence is at the depth of 3600 m, these rocks reach the TTI value of 41, equivalent to the middle part of the oil window.

For the modelling of the burial and maturation history of the Middle Jurassic rocks in the Dobrogean Foredeep, the Stari Troyany-1 (Fig. 7A) and Kalcheva-3 boreholes (Fig. 7B) were chosen, with the depths of the Bathonian's top being 980 and 1700 m, respectively. The burial history plot of the Stari Troyany-1 borehole shows that the Bajocian–Bathonian sequence here reaches the TTI value of 6 and hence just enters the oil window. Therefore, the top of the oil window in this region was defined at a depth of ca 1000 m. In the Kalcheva-3 borehole, these strata reach the TTI value of 25, corresponding to the deeper part of the oil window close to the peak of oil generation.

While the Lower and Middle Jurassic black shales of the Carpathian Foredeep basement reached their present maturation level in the Neogene, the Bajocian–Bathonian rocks of the Bârlad Depression and the Dobrogean Foredeep entered the oil window at the end of the Cretaceous.

As shown by the biomarker studies of Kosakowski et al. (2012) and Rauball et al. (2020), despite the good petroleum potential of the Middle Jurassic rocks of the Carpathian Foredeep basement and their occurrence in the oil window, these rocks have not been a source for the Kokhanivka and Orkhovychi heavy oil fields in the overlying Upper Jurassic carbonate reservoirs. Geochemical investigations of gases from the overlying Mesozoic–Miocene strata in their deeper occurrence, e.g. Bilche-Volytsia, Rudky and Letnia fields (Kotarba and Koltun 2011), show that they represent a mixture of thermogenic and biogenic hydrocarbons. Isotopic studies of kerogen from the Middle Jurassic rocks (Kosakowski et al. 2012) indicate that they can be considered as a source for this thermogenic component. Geochemical studies of oils from the adjacent Carpathian flysch sequence (Więcław et al. 2012) reveal that apart from the main oil family generated within this succession, there are oils of mixed origin (Volya-Blazhivska-31 borehole), containing hydrocarbons originating from the Oligocene flysch rocks and the platform basement.

Hydrocarbon accumulations in the Mesozoic reservoir rocks in the Bârlad Depression could originate from the Middle Jurassic strata, which are considered to be one of the main source rock strata in the area (Pawlewicz 2007; Pene 1999). Reservoir rocks in the Bârlad Depression occur in the Jurassic and Paleozoic sections at the depth of around 4200 m, and are represented by the quartzitic sandstones with an average porosity of 4–9% and permeability of 1–5 mD (Ionescu 1994).

The Middle Jurassic organic-rich rocks in the Dobrogean Foredeep occur in the oil window over a significant part of the study area (Fig. 4). Numerous oil and gas shows have been reported from the Middle Jurassic sequence during the drilling of several boreholes (Bogayets et al. 1986), suggesting possible hydrocarbon generation within these strata. The frequent oil and gas shows in the overlying Upper Jurassic organic-lean sequence indicate that this sequence

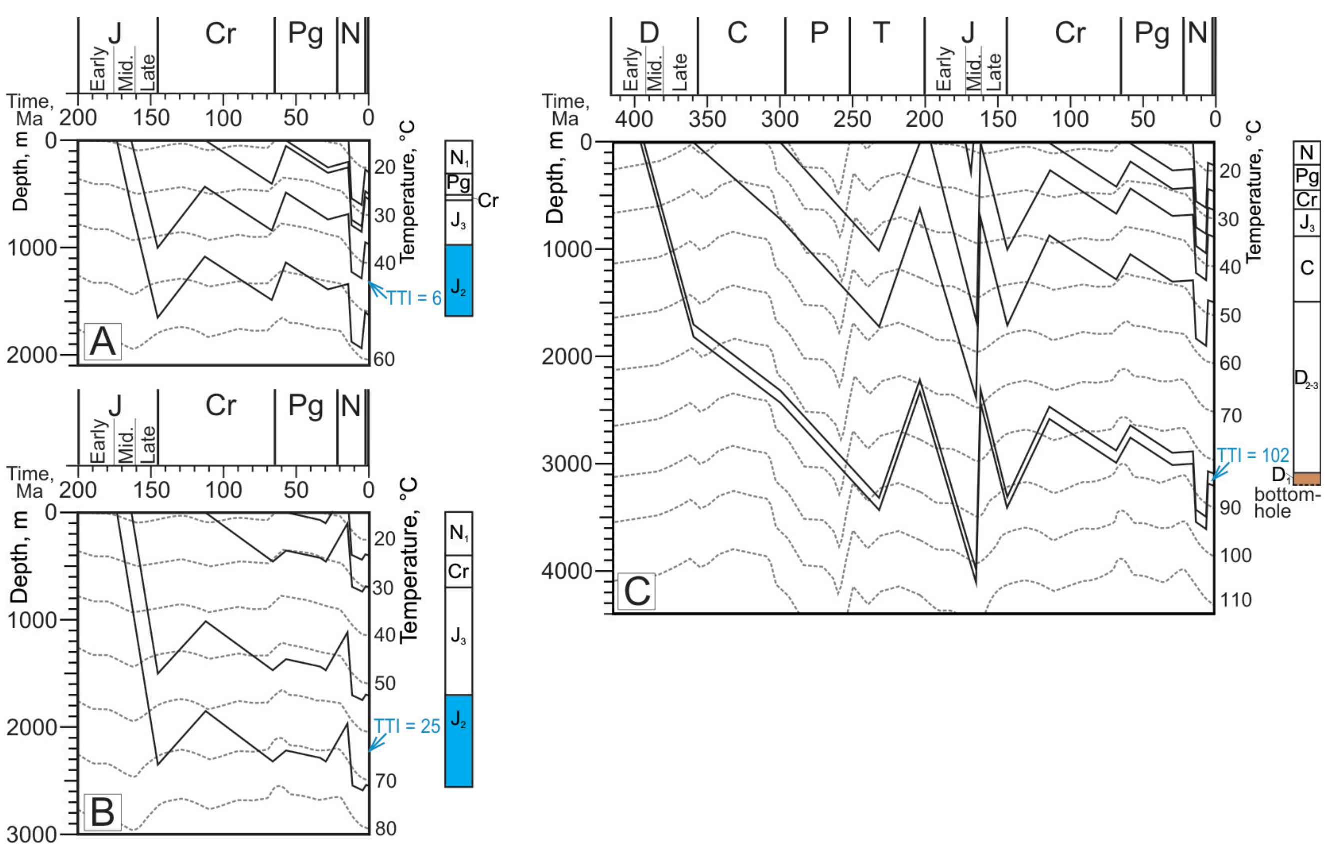

Fig. 7. Burial history plots for the Bajocian–Bathonian (A, B) and the Lower Devonian (C) strata of the Dobrogean Foredeep: **A** – Stari Troyany-1 borehole, **B** – Kalcheva-3 borehole, **C** – Skhidna Sarata-1 borehole. Blue and brown colours mark the strata under study, J_2 and D_1, respectively. Abbreviations: D – Devonian, D_1 – Lower Devonian, D_{2-3} – Middle–Upper Devonian, C – Carboniferous, P – Permian, T – Triassic, J – Jurassic, J_2 – Middle Jurassic, J_3 – Upper Jurassic, Cr – Cretaceous, Pg – Paleogene, N – Neogene, N_1 – Lower Neogene.

may contain hydrocarbon accumulations. These terrigenous-carbonate rocks exhibit favourable reservoir properties (open porosity 8–10%) and are covered with clay-lithology beds, acting as seals (ibid.).

Two oil fields have been recently discovered in the Dobrogean Foredeep: Zhovtoyarsk and Sarata. They occur in the Middle Devonian reservoirs of the adjacent block eastwards from the study area (Fig. 1). The Middle Jurassic strata are lacking in this block. Therefore, the Lower Devonian (Lochkovian) organic-rich beds are supposed to source these hydrocarbon accumulations (Radzivil et al. 2012; Radkovets 2016; Radkovets et al. 2021). The modelling of the burial and maturation history of the Skhidna Sarata-1 borehole, located in this block, shows that the top of the Lower Devonian strata at the depth of 3270 m reached the TTI value of 102 (Fig. 7C), indicating that the maturation level corresponds to the lower part of the oil window. The black shales of the Lochkovian beds, which are supposed to source these oil fields, occur within the depth range of 3570–4260 m, showing oil generation from the base of the oil window. The Silurian strata in this block, widely represented by black shales, are made up of the biolithites with $CaCO_3$ content of over 95% (Radkovets 2015; Radkovets et al. 2021), and hence cannot be considered as source rocks.

The Middle Jurassic strata within the studied blocks occur in the oil window and can be considered as a source rock, while the Lower Devonian (Lochkovian) strata occur much deeper, reaching up to 12 km in the southern part of the study area, being evidently overmature. Thus, possible hydrocarbon occurrences within the studied blocks of the Dobrogean Foredeep can be expected in the Upper Jurassic strata, where the terrigenous-carbonate sequence could be the reservoir rocks for hydrocarbons, mostly sourced by the Middle Jurassic black shales.

Mineral composition of the Toarcian–Bathonian rocks

The XRD results indicate that the Toarcian–Bathonian organic-rich rocks are characterized by heterogenic mineral composition (Fig. 8; Table 2). Terrigenous minerals predominate over clay, and their content ranges from 41 to 85.6%. The most common terrigenous mineral is quartz, the content of which in the rock is from 27 to 49%, while sericite makes up 13–20%, mica 2–10% and chlorite 3–11%. Kaolinite is a mineral of the clay group; however, a significant amount of it was recorded in coarser fractions. Albite, calcite, siderite and pyrite were identified in individual samples. An admixture of witherite (ca 1%) was found in all samples.

In the sand and silt fractions of the rocks (1–0.05 and 0.05–0.005 mm, respectively), the coexistence of two micaceous minerals was revealed: sericite (hydrated, fine-scale muscovite) and mica, the structure of which can be attributed to muscovite $2M_1$. These minerals are characterized by re-

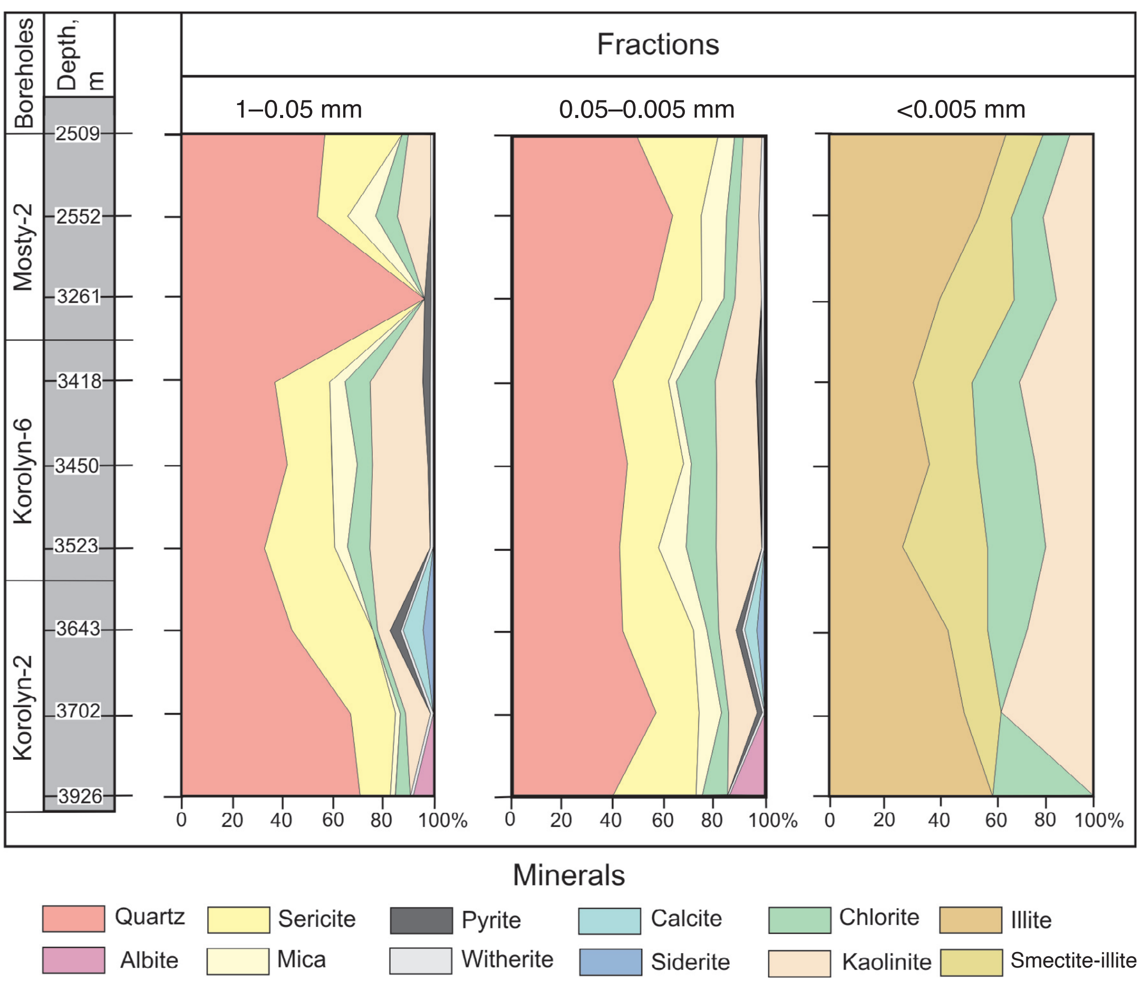

Fig. 8. Mineral composition of the Toarcian–Bathonian organic-rich rocks of the Carpathian Foredeep basement based on XRD results.

Table 2. Mineral composition of the Toarcian–Bathonian organic-rich rocks of the Carpathian Foredeep basement based on XRD results

Boreholes	Depth, m	Whole-rock mineral content, %											Mineral content of clay fraction, %			
		Q	Ab	Sr	Mi	Ch	K	Pr	Wt	Ca	Sd	TC	It	Ch	S/I	K
Mosty-2	2509	31.9	0	16.2	4.8	4.2	2.4	0	0.6	0	0	39.9	67	10	14	9
Mosty-2	2552	38.0	0	14.8	4.9	4.3	7.0	0	1.4	0	0	29.6	57	12	12	19
Mosty-2	3261	49.1	0	13.1	6.6	4.9	6.5	0	1.6	0	0	18.2	42	16	28	14
Korolyn-6	3418	36.4	0	13.2	10.1	5.4	10.1	1.5	0.8	0	0	22.5	32	18	22	28
Korolyn-6	3450	32.5	0	16.3	3.9	7.0	15.5	1.5	0.7	0	0	22.6	38	22	18	22
Korolyn-6	3523	26.7	0	19.6	7.9	6.3	17.3	0	0.8	0	0	21.4	28	22	32	18
Korolyn-2	3643	28.9	0	17.2	6.2	3.4	5.5	0	0.7	4.1	2.7	31.3	45	15	15	25
Korolyn-2	3702	43.4	0	26.5	2.5	0	7.2	0	0.8	0	0	19.6	51	0	14	35
Korolyn-2	3926	39.4	10.3	14.6	9.4	11.0	0	0	0.9	0	0	14.4	62	38	0	0

Q – quartz, Ab – albite, Sr – sericite, Mi – mica, Ch – chlorite, K – kaolinite, Pr – pyrite, Wt – witherite, Ca – calcite, Sd – siderite, TC – total clay, It – illite, S/I – smectite-illite

flexes among which 00*L* have the same values of interplanar distances (*d*). The presence of mica of polymorphic modification $2M_1$ was determined by the location of non-basal reflexes in the area of high angles using Profex software (Döbelin and Kleeberg 2015). Chlorite is mostly represented by its ferrous variety – chamosite.

Detailed investigations of the clay fraction (<0.005 mm) of the rocks in non-treated (air-dried) oriented samples, saturated with ethylene glycol, heated at 550 °C and washed with a solution of 15% HCl, showed that the clay fraction in all the studied samples contained illite and chlorite, which were identified by the characteristic maxima that do not change their positions after saturation with ethylene glycol and remain after heating the samples. Kaolinite was identified by a clear reflex of 0.71 nm. The mixed-layered mineral with predominantly illite layers – mixed-layered smectite-illite – was also present. It was reflected by small maxima from a side of low angles at the slope of reflex 001, characteristic of illite, which changes its position after saturation with ethylene glycol and disappears after heating at 550 °C. In the clay fraction of the rocks (14.4–39.9%), illite predominates, making up 32–67%. Mixed-layered smectite-illite varies between 12–32%, kaolinite 9–35% and chlorite 10–16% (Fig. 8). At the depth of 3926 m (Korolyn-2 borehole), kaolinite and mixed-layered smectite-illite disappear, and the clay fraction consists only of hydromica and chlorite.

The content of clay minerals in the Toarcian–Bathonian organic-rich rocks reflects their depth-dependence. The XRD analysis indicated smectite-illite and kaolinite in the rocks only above the depths of 3700 m, i.e. in immature and early mature rocks. Within the zones of advanced oil and gas generation window, the rocks contain only chlorite and illite. The lack of kaolinite and smectite-illite with swelling components, as well as the increased terrigenous content in black shales at the depth of over 3700 m, testify to high brittleness, which makes these rocks suitable for hydraulic fracturing.

Conclusions

The sedimentation of the Lower and Middle Jurassic strata resulted from a global anoxic event that caused the deposition of organic-rich rocks, which are often considered as important source rocks, in many parts of the world. In the study area, which includes the Carpathian Foredeep basement, the Bârlad Depression and the Dobrogean Foredeep, these strata have a laterally wide occurrence and comprise black shale sequences of high thickness.

In the Ukrainian part of the Carpathian Foredeep basement, the Toarcian–Bathonian is represented by the up to 600 m thick succession of organic-rich rocks, containing type II/III kerogen with good petroleum potential at an early mature to mature stage. The top of the oil window occurs at the depth of 2200 m. Although these rocks have not sourced hydrocarbons into the Kokhanivka and Orkhovychi heavy oil fields, occurring in the overlying Upper Jurassic carbonate reservoirs, as shown by previous biomarker studies, it can be assumed that the hydrocarbons generated by these rocks might have charged the Mesozoic–Miocene accumulations in the Carpathian Foredeep basement and some oil fields of the Carpathian flysch sequence.

In the Bârlad Depression, the Bajocian–Bathonian strata, which are up to 1800 m thick, are essentially made up of black shales with mainly type II/III kerogen occurring in the oil window. This succession is considered to be one of the main source rocks in the region. The coeval age sequence of the Dobrogean Foredeep, reaching the thickness of 800 m and being composed mainly of black shales, achieved maturation equivalent to the oil window over a significant area. The top of the oil window occurs at the depth of 1000 m. The existing Zhovtoyarsk and Sarata oil fields occur within the block, lacking Middle Jurassic strata and indicating the presence of mature Lower Devonian beds, being evidently sourced by the latter. Within the study part of the Dobrogean Foredeep, the Lower Devonian sequence is overmature, and hence the

Middle Jurassic strata remain the only potential source rocks. Thus, the overlying Upper Jurassic terrigenous-carbonate sequence may be considered as the prospective target for hydrocarbons exploration.

The XRD results indicate that the Toarcian–Bathonian organic-rich rocks are characterized by a heterogenic mineral composition. Terrigenous minerals predominate over clay, and their content ranges from 41 to 85.6%. The content of clay minerals reflects their depth-dependence. Smectite-illite and kaolinite occur in the rocks only up to the depths of around 3700 m, i.e. in immature and early mature rocks, while at greater depths, the content of clay minerals shows high brittleness of the rocks, which makes them suitable for hydraulic fracturing.

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