

PETROLEUM GENERATION AND EXPULSION IN THE LOWER PALAEOZOIC PETROLEUM SOURCE ROCKS AT THE SW MARGIN OF THE EAST EUROPEAN CRATON (POLAND)

Dariusz BOTOR^{1*}, Jan GOLONKA¹, Justyna ZAJĄC¹, Bartosz PAPIERNIK¹ & Piotr GUZY¹

¹AGH University of Science and Technology, Faculty of Geology, Geophysics and Environmental Protection, al. Mickiewicza 30, 30-059 Kraków, Poland; e-mails: botor@agh.edu.pl, jgonlonka@agh.edu.pl, papiern@geol.agh.edu.pl, [zajac@agh.edu.pl](mailto;zajac@agh.edu.pl), guzy@agh.edu.pl

* Corresponding author

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Abstract: In this work, 1-D numerical modelling of petroleum generation and expulsion processes in the Upper Ordovician and Lower Silurian source rocks was carried out in over sixty wells along the SW margin of the East European Craton (EEC) in Poland. Lower Palaeozoic sediments were subjected to rapid burial in the Palaeozoic and then were uplifted in several phases, but with the predominance of the late Variscan tectonic inversion. The thermal maturity of organic matter in the Lower Palaeozoic strata indicates the advancement of the generation processes from the phase of low-temperature thermogenic processes in the NE part of the Baltic and Podlasie-Lublin basins to the overmature stage along the zone adjacent to the Teisseyre-Tornquist Zone (TTZ). The results of modelling of generation and expulsion show that these processes took place mainly in the Devonian and Carboniferous periods and in the westernmost part (along the TTZ), even in the latest Silurian. The hydrocarbon expulsion took place with a small – delay after generation. During the Mesozoic and Cainozoic, generation processes practically were not resumed or intensified. Nevertheless, it was found that zones with an increased shale gas potential can occur only in a relatively narrow belt on the SW slope of the EEC, parallel to the edge of the TTZ. The most promising seem to be Caradocian, Llandovery and the Wenlock between the Lębork IG-1 and Kościerzyna IG-1 wells in the Baltic Basin, and the Wenlock source rocks in the Podlasie-Lublin Basin between the Okuniew IG-1, Łopiennik IG-1 and Narol IG-1 wells. Most of the hydrocarbons were subjected to expulsion and possible migration. As a result, there was a large dispersion of the hydrocarbons generated. The chance of preservation of these hydrocarbons in the source rocks is small.

Key words: 1-D maturity modelling, shale gas, petroleum source rocks, Lower Palaeozoic.

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INTRODUCTION

The source rocks of hydrocarbons are basic elements in any petroleum system and the first necessary factor for the creation of an oil or natural gas reservoir, either conventional or unconventional (e.g., Magoon and Dow, 1994; Jarvie *et al.*, 2006; Botor *et al.*, 2017a, b, c). In the last decade, source rocks also have been recognized as potential reservoirs for unconventional hydrocarbons (e.g., Jarvie *et al.*, 2006). The hydrocarbon potential of the source rock is controlled mainly by the amount of organic carbon (total organic carbon, TOC, as % by weight), the thermal maturity of the organic matter and the genetic type of the kerogen. At the same time, the parameters mentioned above are the most important in the assessment of the potential of unconventional deposits (Jarvie *et al.*, 2006). The Lower

Palaeozoic source rocks of the East European Craton (EEC) are seemingly monotonous, fine-grained sediments (shales, claystones and mudstones). They are in fact very diverse in terms of sedimentology (e.g., Lis, 2010; Porębski *et al.*, 2013) and the organic matter of the sediments also shows significant variation (e.g., Pletsch *et al.*, 2010; Więclaw *et al.*, 2010a, 2012; Kosakowski *et al.*, 2016).

The main exploration target for unconventional hydrocarbons is in Ordovician and Silurian black shales along the SW margin of the EEC in Poland (Poprawa, 2010; Kiersnowski and Dyrka, 2013; Botor, 2016; Karnkowski and Matyasik, 2016; Botor *et al.*, 2017c). These contain diversified amounts of organic matter and their thermal maturity is in the range of the main and late phases of hydrocar-

bon generation processes (Kosakowski *et al.*, 1998, 1999; Botor *et al.*, 2002; Klimuszko, 2002; Skręt and Fabiańska, 2009; Kosakowski *et al.*, 2010, 2016; Pletsch *et al.*, 2010; Poprawa, 2010; Więclaw *et al.*, 2010a, 2012; Botor *et al.*, 2017a, b, c). The study area is situated on the SW margin of the EEC, in the NE part of Poland (Fig. 1). So far, the modelling of hydrocarbon generation processes in the Lower Palaeozoic deposits has been carried out only in selected zones of the EEC area (Kosakowski *et al.*, 1998, 1999, 2010; Karnkowski, 2003a, b; Poprawa *et al.*, 2010; Wróbel and Kosakowski, 2010; Botor, 2016, 2018; Radkovets *et al.*, 2018). This modelling did not, in many cases, give clear conclusions, mainly owing to deficiencies in the measurement data, especially in the field of thermal maturity of the kerogen and its distribution in individual source rocks. To evaluate further the development of hydrocarbon generation as well as the expulsion processes in the Lower Palaeozoic source rocks of the EEC in NE Poland, first the modelling of the history of burial and thermal history was accomplished; this is presented in the companion paper by Botor *et al.* (2019). Then these results were applied to the modelling of hydrocarbon generation and expulsion in this study. However, the work was based mainly on wells of the Polish Geological Survey (Państwowy Instytut Geologiczny) and only a few new, industry wells were used (from Orlen Upstream and the Polish Oil and Gas Company). This paper focuses on the Caradocian, Llandovery and Wenlock source

rocks as the most prospective, unconventional hydrocarbon accumulations (Poprawa, 2010; Podhalańska *et al.*, 2016). The Alum Shale (late Cambrian to earliest Ordovician) is of limited value as a target because of its insignificant thickness in Poland (Pletsch *et al.*, 2010; Więclaw *et al.*, 2010a; Kosakowski *et al.*, 2016). This paper is an attempt to reconstruct the generation processes, not only in selected small regions, but on the scale of the entire SW area of the EEC in Poland. The present account in English is a significantly modified version of the book chapter (Botor *et al.*, 2017c), available so far only in the Polish language.

METHODS

Hydrocarbon generation and expulsion modelling was carried out using one-dimensional PetroMod ver. 11 software (Hantschel and Kauerauf, 2009) for over 60 wells in the study area. First, burial and thermal maturity modelling was performed, as discussed in the companion paper (Botor *et al.*, 2019). In these models, thermal maturity was calculated by means of the “Easy%R_o” method (Sweeney and Burnham, 1990). Once the thermal evolution of all sedimentary layers was established with confidence by comparing measured and calculated maturity and present-day corrected borehole temperatures (Botor *et al.*, 2019), the modelling of hydrocarbon generation and expulsion from organic-rich source rocks could be achieved on the basis of

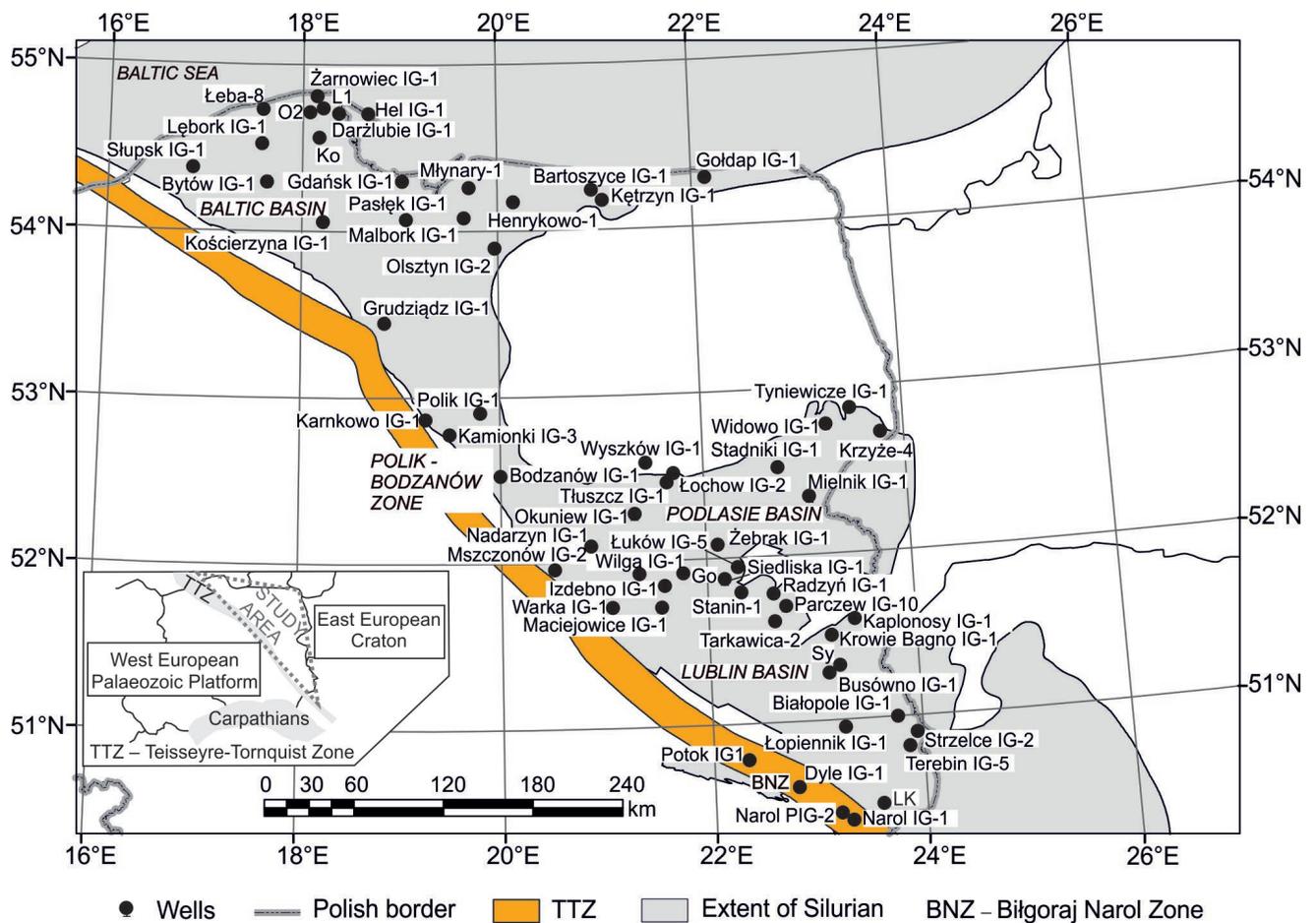


Fig. 1. Location map showing analysed boreholes. TT – Teisseyre-Tornquist Line.

Arrhenius law kinetics, using source-rock-specific kinetic data sets, which are sufficiently representative for the source rocks under investigation (Hantschel and Kauerauf, 2009). Published thermal maturity data were used for the calibration of the 1-D models (Nehring-Lefeld *et al.*, 1997; Grotek, 1998, 1999, 2005, 2006, 2015, 2016; Grotek *et al.*, 1998; Kosakowski *et al.*, 1998, 2010; Matyasik, 1998; Swadowska and Sikorska, 1998; Botor *et al.*, 2002; Skręt and Fabiańska, 2009; Pletsch *et al.*, 2010). Additionally, the present-day temperature in boreholes was applied (Plewa, 1991, 1994; Górecki *et al.*, 2006a, b). In some wells, also porosity and density data were used for calibration (Modliński, 2007; Paczeńska *et al.*, 2007; Modliński *et al.*, 2011; Karcz, 2015). This 1-D modelling of hydrocarbon generation and expulsion processes was performed by assuming the presence of low-sulphur marine-algae kerogen (type IIB) in the Ordovician–Silurian source rocks and applying the Pepper and Corvi (1995) kinetic model. The initial hydrogen index was set at 500 mg of hydrocarbons per gram of TOC in all the analysed source rocks. The original TOC content was assumed as follows: 2.0% in the Caradocian and 2.0% in the Llandovery as well as 1.0% in the Wenlock in the Baltic Basin and 1.6% in the Podlasie-Lublin Basin. The values of kerogen transformation, generation potential and expulsion shown on the maps are average values for each depth interval of occurrence of the source rocks analysed. Detailed descriptions of the modelling procedures are given in Botor *et al.* (2013). Further details of the methods are given by e.g., Hantschel and Kauerauf (2009).

GEOLOGICAL SETTING

The study area is located on the SW slope of EEC, where the following geological units can be distinguished: the Baltic Basin in the northern part and the Podlasie-Lublin Basin in the south (Fig. 1). Between them, there is the Polik-Bodzanów zone, the geological history of which is not known in detail (Narkiewicz, 2007). In the study area, a sedimentary sequence of a significant thickness on the Precambrian crystalline basement, includes strata from late Precambrian to Cainozoic in age. The thickness of the Ediacaran to Lower Palaeozoic section increases towards the west. The Neoproterozoic to Early Palaeozoic geological development was closely related to tectonic processes occurring west of the EEC margin that is presently in the Trans-European Suture Zone (TESZ; Torsvik *et al.*, 1990, 1993, 1996; Oliver *et al.*, 1993; Meissner *et al.*, 1994; Nikishin *et al.*, 1996; Tanner and Meissner, 1996; Maletz *et al.*, 1997; Šliaupa *et al.*, 1997; McCann, 1998; Poprawa *et al.*, 1999, 2018; Poprawa, 2006a, b, 2017; Nawrocki and Poprawa, 2006; Radkovets, 2015). The geological history of the study area is summarized in detail by Botor *et al.* (2019). The following elements are of greatest importance to the objectives of the present account: 1) rifting in the Late Neoproterozoic to early Cambrian (Nikishin *et al.*, 1996; Šliaupa *et al.*, 1997, 2006; Poprawa *et al.*, 1999; Lassen *et al.*, 2001; Poprawa and Paczeńska, 2002; Poprawa, 2017; Krzywiec *et al.*, 2018); 2) a passive margin phase and later development of flexural foredeep in the late Ordovician to Silurian (Nikishin *et al.*, 1996; Šliaupa *et al.*,

1997, 2006; Poprawa *et al.*, 1999, 2018; Poprawa, 2006a, b, 2017; Paczeńska, 2006; Podhalańska and Modliński, 2006; Mazur *et al.*, 2016, 2018; Poprawa *et al.*, 2018); 3) a Devonian to Carboniferous subsidence phase (Żelichowski, 1987; Porzycki and Zdanowski, 1995; Narkiewicz *et al.*, 1998, 2007, 2011, 2015; Skompski, 1998; Waksmundzka, 1998; 2005, 2010; Matyja, 2006; Narkiewicz, 2007; Narkiewicz and Narkiewicz, 2008; Krzywiec, 2009; Radkovets, 2016; Krzywiec *et al.*, 2017a, b; Tomaszczyk and Jarosiński, 2017); 4) Variscan compression in the latest Carboniferous (Żelichowski, 1987; Narkiewicz *et al.*, 1998, 2011, 2015; Narkiewicz, 2007; Krzywiec, 2009; Krzywiec *et al.*, 2017a, b; Tomaszczyk and Jarosiński, 2017); 5) the Permian–Mesozoic development of the Polish Basin (Kutek and Głazek, 1972; Dadlez *et al.*, 1995; Kutek, 2001; Lamarche *et al.*, 2003); 6) the tectonic inversion of the Polish Basin in the Late Cretaceous–Palaeogene (Krzywiec, 2002; Lamarche *et al.*, 2003; Mazur *et al.*, 2005; Scheck-Wenderoth *et al.*, 2008; Krzywiec, 2009); 7) the development of the thin Cainozoic sedimentary cover of poorly consolidated (Kutek and Głazek, 1972; Dadlez *et al.*, 1995; Kutek, 2001; Krzywiec, 2002; Lamarche *et al.*, 2003; Mazur *et al.*, 2005; Scheck-Wenderoth *et al.*, 2008; Krzywiec, 2009).

PETROLEUM SYSTEMS AND SOURCE ROCKS

In the study area of the Polish part of the EEC, petroleum systems are related only to the Palaeozoic strata. However, in the Mesozoic basement of the Carpathian Foredeep, the Mesozoic reservoirs could have been sourced by the Jurassic source rocks as well as by bacterial gas from the Miocene strata (Kotarba *et al.*, 2011). The Lopushna oil field, occurring in the Mesozoic strata of the EEC, forming the basement of the Ukrainian part of the Carpathian Foredeep, covered by the Carpathian Overthrust, was sourced by the Oligocene source rocks of the Carpathian flysch (Radkovets *et al.*, 2016).

In the Baltic Basin, Cambrian quartzose sandstones with a thickness in the range of 60–120 m are the major reservoir horizons of conventional oil and gas accumulations (Górecki *et al.*, 1992; Karnkowski, 1993; Domżański *et al.*, 2004; Pletsch *et al.*, 2010). All the petroleum traps in the Cambrian sandstones are fault-related anticlines or cross-fault structures, which were active during the Caledonian and the Variscan vertical movements (Stolarczyk, 1979; Górecki *et al.*, 1992; Witkowski, 1993; Domżański *et al.*, 2004; Pletsch *et al.*, 2010). In the Lublin Basin, both carbonate reservoirs in the Devonian and sandstone reservoirs in the Carboniferous are known (Karnkowski, 1993, 2007; Helcel-Weil *et al.*, 2007; Pletsch *et al.*, 2010). Structural, fault-related traps formed mainly during the late Variscan tectonic inversion (Pletsch *et al.*, 2010).

On the SW slope of the EEC area, possible source rocks horizons occur in the Ediacaran to lower Cambrian strata (Paczeńska *et al.*, 2005) and in the upper Cambrian–Tremadocian Alum Shale (Kosakowski *et al.*, 2016), in the Upper Ordovician (mainly Caradocian) and in the Lower Silurian (Llandovery and Wenlock) strata (Pletsch *et al.*, 2010;

Poprawa, 2010; Więclaw *et al.*, 2010a; Kosakowski *et al.*, 2016; Podhalańska *et al.*, 2016; Radkovets *et al.*, 2017a). In the Lower Palaeozoic strata in the Polish part of the Baltic and Podlasie-Lublin basins, oil-prone, type-II kerogen occurs (Pletsch *et al.*, 2010; Poprawa, 2010; Więclaw *et al.*, 2010a, 2012; Kosakowski *et al.*, 2016; Podhalańska *et al.*, 2016). In the Lublin Basin, Devonian and Carboniferous source rocks also were identified (Botor *et al.*, 2002; Karnkowski, 2007; Pletsch *et al.*, 2010; Radkovets *et al.*, 2017b). In the Devonian, thin source-rock horizons containing type II kerogen predominate, with low TOC values of usually below 1% (Pletsch *et al.*, 2010; Radkovets *et al.*, 2017b). The source rocks in the Carboniferous are dominated by gas-prone coal-bearing sediments, containing an average of 2% TOC (Porzycki and Zdanowski, 1995; Botor *et al.*, 2002; Pletsch *et al.*, 2010).

Thermal maturity in the EEC

The thermal maturity of the Ediacaran to Mesozoic strata of the SW margin of EEC was investigated using reflectance of vitrinite and vitrinite-like particles, Rock-Eval Tmax, conodont colour alteration index (CAI) and Thermal Alteration Index (Drygant, 1993; Kanev *et al.*, 1994; Nehring-Lefeld *et al.*, 1997; Zdanaviëiûtë and Bojesen-Koefoed, 1997; Kosakowski *et al.*, 1998, 1999, 2016; Swadowska and Sikorska, 1998; Grotek, 1999, 2006; Botor and Kosakowski, 2000; Skręt and Fabiańska, 2009; Pletsch *et al.*, 2010; Poprawa, 2010; Więclaw *et al.*, 2010a, 2012; Stempień-Safek, 2011). The thermal maturity measurements of the Lower Palaeozoic formations are generally conducted on vitrinite-like organic particles, such as zooclasts, graptolites, alginates and bitumen, which leads to higher uncertainty than the usual analysis using vitrinite. The assumed relationship between graptolite reflectance and the equivalent vitrinite reflectance implies that the reflectance of graptolites increases faster than the reflectance of vitrinite (Petersen *et al.*, 2013).

The Ediacaran to Lower Palaeozoic strata of the SW margin of EEC revealed a systematic zonation (NE to SW) in the degree of diagenesis along the entire margin. The thermal maturity of the Lower Palaeozoic strata on the western slope of the EEC, as well as the depth of burial, generally increases from the east and NE to the west and SW. This increase is followed by a change in the properties of the source rocks, which changes their maturity from the immature or poorly mature rocks, through generation of the hydrocarbons window, to the dry gas window or even to overmaturity. This change occurs along the TTZ (Drygant, 1993; Kanev *et al.*, 1994; Nehring-Lefeld *et al.*, 1997; Zdanaviëiûtë and Bojesen-Koefoed, 1997; Kosakowski *et al.*, 1998, 1999, 2010, 2016; Swadowska and Sikorska, 1998; Grotek, 1999, 2006; Botor and Kosakowski, 2000; Zdanaviëiûtë, 2005; Pletsch *et al.*, 2010; Poprawa, 2010; Więclaw *et al.*, 2010a, 2012; Radkovets *et al.*, 2018).

In the Polish part of the Baltic Basin, the measured values of thermal maturity of the Lower Palaeozoic formations change from about 0.5–0.6% R_o on the eastern side to almost 5% R_o on the western side along the TTZ (Kanev *et al.*, 1994; Nehring-Lefeld *et al.*, 1997; Zdanaviëiûtë and

Bojesen-Koefoed, 1997; Kosakowski *et al.*, 1998, 1999, 2010, 2016; Swadowska and Sikorska, 1998; Grotek, 1999, 2006; Botor and Kosakowski, 2000; Zdanaviëiûtë, 2005; Pletsch *et al.*, 2010; Poprawa, 2010; Więclaw *et al.*, 2010a, 2012). Measurements of vitrinite-like reflectance (Swadowska and Sikorska, 1998; Grotek, 1999, 2006, 2009, 2016) are confirmed by pyrolytic Tmax values from Rock-Eval analysis (Kosakowski *et al.*, 1998, 1999, 2010, 2016; Botor and Kosakowski, 2000; Pletsch *et al.*, 2010) and Conodont CAI (Nehring-Lefeld *et al.*, 1997) as well as illitization of smectite (Środoń and Clauer, 2001; Kowalska *et al.*, 2017). In the Cambrian deposits, thermal maturity increases regularly with increasing depth from 0.63% R_o at a depth of 1,906 m in the eastern part of the Baltic Basin to 4.92% at a depth of 4,934 m in the western part (e.g., Słupsk IG-1). The least thermally mature (R_o below 1.0%), Cambrian deposits occur at shallow depths (1,537–2,591 m) in the eastern part of the basin, mainly in Lithuania (Zdanaviëiûtë, 2005). Thermal maturity increases towards the west, reaching the value of 1.5% R_o at the Łeba High. This increase is similar to one occurring within the much shallower (1,316–1,857 m) Cambrian deposits in the B block of the Baltic Sea, where it ranges from 1.0 to 1.2% R_o (Grotek, 2006). Towards the south and west edges of the basin, the thermal maturity continues to grow, reaching values from 1.5 to 2.5% R_o (Swadowska and Sikorska, 1998; Grotek, 2006). The very high degree of maturity is also characterized by dispersed organic matter in block A, located in the area of the TTZ, where already at a depth of 1,940 m the value of R_o reaches 2.3% (Swadowska and Sikorska, 1998; Grotek, 2006). The Tmax values from Rock-Eval pyrolysis analysis range from about 420 to 535 °C, while the PI values from 0.01 to 0.49 (Kanev *et al.*, 1994; Kosakowski *et al.*, 1998, 1999, 2016; Botor and Kosakowski, 2000; Zdanaviëiûtë, 2005; Pletsch *et al.*, 2010; Więclaw *et al.*, 2010a, 2012). Their values, like R_o , increase from east to west.

In the Lower Palaeozoic strata of the central part of the Podlasie Basin, the thermal maturity is ca. 0.9–1.1% R_o , reaching 1.3% R_o in the western part. In the eastern part of the Lublin Basin, the thermal maturity changes from east to west from 0.6–0.7% R_o to ca. 1.5–2.0% R_o (Grotek, 2005, 2016). The highest thermal maturity is in the Lopiennik IG-1 well (2.7–3.4% R_o). In the Biłgoraj-Narol area (the SW part of the Lublin Basin along the TTZ; Fig. 1), despite the small depth of occurrence of the Lower Palaeozoic, the thermal maturity is high (above 2.0% R_o ; Nehring-Lefeld *et al.*, 1997; Swadowska and Sikorska, 1998; Wróbel *et al.*, 2008; Grotek, 2009, 2015, 2016; Pletsch *et al.*, 2010; Poprawa, 2010; Kosakowski *et al.*, 2013; Karcz and Janas, 2016).

The interior of the EEC has yielded CAI values of 1–1.5 (for the Ordovician–Devonian rocks), indicative of palaeotemperatures between ca. 50 and 90 °C. Toward the SW margin of the EEC, the CAI values (at least for the Ordovician strata) gradually increase and reach level 5 (i.e. over 300 °C) in the TTZ (Drygant, 1993; Nehring-Lefeld *et al.*, 1997).

The Lower Palaeozoic strata in the Baltic Basin are covered by the Permian and Mesozoic strata that show much lower thermal maturity than the Lower Palaeozoic deposits, usually below 0.6% R_o (Grotek 1999, 2006; Poprawa *et al.*, 2010). Toward the SE, the Lower Palaeozoic strata

in the Podlasie and Lublin basins are covered by Devonian (Rock-Eval Tmax 426–466 °C) and Carboniferous (ca. 0.4–1.2% R_o) strata that show variable thermal maturity (Matyasik, 1998; Botor *et al.*, 2002; Poprawa and Paczeńska, 2004; Grotek, 2005; Pletsch *et al.*, 2010; Radkovets *et al.*, 2017a, b). However, the Permian–Mesozoic strata also show relatively low thermal maturity below 0.5% R_o (Grotek, 1998, 2005, 2006; Botor *et al.*, 2002; Pletsch *et al.*, 2010).

TOC and thickness of source rocks horizons of Lower Palaeozoic

In the study area, the oldest levels of source rocks are the upper Cambrian and lowest Ordovician (Alum Shale). However, their thickness is identified within the Polish borders as being from 0 to 10 m, rising to over 100 m below the Baltic Sea towards the NW, outside of Poland (Karnkowski, 1993; Kanev *et al.*, 1994; Buchardt *et al.*, 1998; Kosakowski *et al.*, 1998, 2016; Pletsch *et al.*, 2010). These source rocks contain up to 22% TOC (Buchardt *et al.*, 1998; Kosakowski *et al.*, 2010, 2016; Pletsch *et al.*, 2010; Poprawa, 2010; Więclaw *et al.*, 2010a; Wróbel and Kosakowski, 2010; Karcz and Janas, 2016). Their total thickness, however, is small in Poland, so for this reason they cannot be a significant collector of unconventional shale gas. Therefore, the authors have not presented their generation analysis in this work.

Above, there are the Caradocian source rocks represented by the Sasino Formation, developed as black, dark grey clays, often bituminous (Modliński and Podhalańska, 2010; Pletsch *et al.*, 2010; Podhalańska *et al.*, 2016). In the Baltic Basin, they have a high organic carbon content, on average at a level of 1–3% (Pletsch *et al.*, 2010). Their thickness of ca. 20–50 m allows consideration of them as an important horizon of source rocks. The Caradocian shales, despite their great thickness in the Podlasie-Lublin Basin, contain relatively small amounts of dispersed organic matter, usually below 1% and they should not be treated as a potential shale gas collector (Karcz and Janas, 2016; Podhalańska *et al.*, 2016).

The Silurian strata in the western part of the Baltic Basin are represented by siliciclastics of considerable thickness (e.g., Modliński and Podhalańska, 2010). Particularly, the Lower Silurian (Llandovery and Wenlock) source rocks are important (Poprawa, 2010). The lower Llandovery is represented by black bituminous claystones of the Jantar Member (Modliński and Podhalańska, 2010; Pletsch *et al.*, 2010; Karcz *et al.*, 2013; Karcz and Janas, 2016; Podhalańska *et al.*, 2016). The thickness of the Jantar Member reaches a maximum of 10–20 m. The TOC in the Jantar Member is locally up to 20% (in the Białowieża Formation (in the Podlasie Basin) which is the Jantar time equivalent), while the average in the Llandovery profiles ranges from 1 to 6% TOC. In the middle part of the Baltic Basin, the average content of organic matter in Llandovery ranges from 1.0 to 2.5%, while in the eastern and western parts, it is usually less than 1% (Pletsch *et al.*, 2010; Poprawa, 2010; Karcz *et al.*, 2013; Karcz and Janas, 2016). In the NW part of the Lublin region, the average content of organic matter reaches 2–3% in the Llandovery, while towards the south and

SE it decreases to below 1% TOC. The claystones of the Pasłęk Formation contain only ca. 1% TOC (Modliński and Podhalańska, 2010; Pletsch *et al.*, 2010; Karcz *et al.*, 2013; Karcz and Janas, 2016).

The youngest source rocks horizons are the Wenlock strata, with a thickness from 120 m in the east to over 1,000 m in the western part (along the TTZ) of the Baltic Basin. However, the net thickness of intervals enriched with organic matter is smaller, usually ca. 50–100 m (Poprawa, 2010; Karcz *et al.*, 2013; Podhalańska *et al.*, 2016). In the middle and western parts of the Baltic Basin, the average organic matter content is below 1% TOC, whilst in the eastern part of the basin, it increases to 1.3% TOC (Pletsch *et al.*, 2010; Karcz and Janas, 2016). The average content of organic matter also increases towards the SE from the Baltic Basin. In the Podlasie Basin, it ranges from 0.6 to 1.3% and in the Lublin Basin from 1 to 1.7% TOC (Klimuszko, 2002; Poprawa, 2010; Więclaw *et al.*, 2012; Karcz *et al.*, 2013; Karcz and Janas, 2016). Similar average TOC values are given by Podhalańska *et al.* (2016): 1.2% for the Podlasie Basin and 1.3% for the Lublin Basin.

Kerogen type in the Lower Palaeozoic source rocks

The kerogen type was identified by means of geochemical and microscopic methods. The hydrogen index (HI), based on pyrolysis Rock-Eval, fluctuates to a large extent across source rocks in the EEC. The hydrogen index for immature type II kerogen varies on average from 300 to 550 (Hunt, 1996), while in the Lower Palaeozoic shales on the EEC, the variability is even greater, from about 10 to 730 (Kanev *et al.*, 1994; Kosakowski *et al.*, 1998, 1999, 2016; Zdanavičiūtė and Lazauskiene, 2004; Zdanavičiūtė, 2005; Skręt and Fabiańska, 2009; Pletsch *et al.*, 2010; Więclaw *et al.*, 2010a, 2012; Karcz and Janas, 2016), although most HI values are below 450. The value of the hydrogen index and the amount of organic carbon determine the amount of hydrocarbons that can be generated in a given source rock. These two indices show very high variability in the Lower Palaeozoic shales analysed. Therefore, it is difficult to determine the type of kerogen that potentially generated hydrocarbons of the Lower Palaeozoic source rocks by analytical methods, because in areas where the discussed source rocks are characterized by an increased TOC, they also have very high maturity. In turn, where thermal maturity is lower, the organic carbon content is often negligible. This is a significant limitation for laboratory analysis and the proper estimation of the kerogen type. Nevertheless, most likely in the Cambrian, Ordovician and Silurian source rocks in the SW slope of the EEC, mainly low-sulphur type II kerogen of algae-marine origin and with high generation potential predominates (Kanev *et al.*, 1994; Zdanavičiūtė and Bojesen-Koefoed, 1997; Kosakowski *et al.*, 1998, 1999, 2010, 2016; Swadowska and Sikorska, 1998; Grotek, 1999; Zdanavičiūtė, 2005; Pletsch *et al.*, 2010; Więclaw *et al.*, 2010a, 2012; Karcz and Janas, 2016).

In the Lower Palaeozoic sediments, both syngenetic and epigenetic organic matter can be distinguished petrographically (Swadowska and Sikorska, 1998; Grotek, 1999, 2016), which is also confirmed by Rock-Eval pyrolysis (Kanev

et al., 1994; Botor and Kosakowski, 2000; Pletsch *et al.*, 2010; Więclaw *et al.*, 2010a; Karcz and Janas, 2016; Kosakowski *et al.*, 2016). Syngenetic organic matter is represented for the most part by amorphous material, mainly fluorescing. This material consists mainly of alginite and liptodetrinite. The inertinite content does not exceed 5%, and in zooclasts (including graptolites) ranges from 0 to 20%. The remaining part of the organic material is a largely liptinite (Swadowska and Sikorska, 1998; Grotek, 1999). In contrast, the epigenetic matter consists mainly of migrating bitumens (Calikowski, 1984; Swadowska and Sikorska, 1998; Grotek, 1999; Klimuszko, 2002). The saturated hydrocarbons dominate over aromatic ones, with an increased amount of resins and asphaltenes (Calikowski, 1984; Kanev *et al.*, 1994; Grotek, 1999; Klimuszko, 2002; Zdanavičūtė, 2005; Pletsch *et al.*, 2010).

RESULTS AND DISCUSSION

In this section, the results for burial and thermal history evolution are only briefly mentioned, as they were discussed in detail in companion papers (Botor *et al.*, 2017c, 2019), then they are followed by a discussion of maturation, hydrocarbon generation and expulsion through time. The following discussion focus on how these processes led to variations in the present-day oil/gas potential of the source rocks in the study area of the EEC.

The organic matter in the Lower Palaeozoic strata in the eastern part of the Baltic, Podlasie and Lublin basins is immature to generate hydrocarbons (R_o below 0.6%). In the middle part of the Baltic and Podlasie-Lublin basins, kerogen in the Lower Palaeozoic deposits has reached the early and main phases of liquid hydrocarbon generation, characterized by reflectance values of vitrinite ca. 0.6 to 1.0% R_o (Kosakowski *et al.*, 1998, 1999, 2010; Swadowska and Sikorska, 1998; Grotek, 1999, 2006; Zdanavičūtė, 2005; Pletsch *et al.*, 2010; Botor, 2016). In a westerly direction, towards the central part of the Baltic Basin and on the Łeba High, the maturity of organic matter increases up to ca. 1.3% R_o , which indicates the main and late phases of oil condensate generation (Kosakowski *et al.*, 1998, 1999, 2010; Swadowska and Sikorska, 1998; Grotek, 1999; Botor and Kosakowski, 2000; Karnkowski, 2003; Grotek, 2006, 2009, 2016; Botor, 2016). Maximum values of thermal maturity (up to 4.9% R_o) were obtained in the Lower Palaeozoic deposits in the western part of the Baltic Basin near the TTZ. These values pass through the phase of dry thermogenic gas up to the over-maturity phase (Kosakowski *et al.*, 1998, 1999, 2010; Swadowska and Sikorska, 1998; Botor and Kosakowski, 2000; Karnkowski, 2003b; Grotek, 2006, 2009, 2016; Botor, 2016; Botor *et al.*, 2017). In the southern and SE part of the Podlasie-Lublin region, the Lower Palaeozoic organic matter has reached a thermal maturity of more than 1.2% R_o , with maximum values of more than 2.0% R_o in the zone between the Łopiennik IG-1, Narol IG-1 and LK wells (Swadowska and Sikorska, 1998; Grotek, 2008, 2009, 2011, 2015, 2016; Wróbel *et al.*, 2008), which indicates the late phase of liquid hydrocarbon generation and the transition to the gas window phase.

Development of hydrocarbon generation processes over time in the light of burial and thermal history

The results of burial and thermal history modelling, calibrated with thermo-chronological data (Botor *et al.*, 2017a, b, 2018, 2019; Kowalska *et al.*, 2017, 2019) showed that Lower Palaeozoic strata occurring in the area of the EEC in Poland reached the maximum palaeotemperature already in the Palaeozoic, the majority in the period being from the end of Silurian to Early Carboniferous (in the Baltic Basin and Late Carboniferous / Early Permian (in the Podlasie-Lublin Basin). In the Mesozoic and Cainozoic, the Lower Palaeozoic strata in general were cooled. Possible additional heating of the Lower Palaeozoic deposits occurred only in zones of increased Mesozoic burial between the Nadarzyn IG-1 and Maciejowice IG-1 wells (Botor *et al.*, 2017c). As a result, in the Mesozoic, there was no additional transformation of kerogen into hydrocarbons in the greater part of the area analysed. Thus, the main period of maturation of organic matter and generation of hydrocarbons in the Ordovician and Silurian source rocks was between the end of the Silurian and the Carboniferous. The generation of hydrocarbons ended as a result of the Variscan inversion (Figs 2–5).

In the most western part of the Baltic Basin between the Słupsk IG-1 and Kościerzyna IG-1 wells, the source rocks analysed entered the hydrocarbon generation phase in the oil window at the end of the Silurian. In the Devonian, there was a further development of these processes, and there was entry into the gas window and along of the TTZ (Słupsk IG-1), also entry into the overmature phase (Fig. 2). In the central part of the Baltic Basin, the source rocks analysed entered the hydrocarbon generation phase in the oil window in the Early Devonian (Gdańsk IG-1) or Late Devonian (Paślęk IG-1) (Fig. 3). The gas window was reached in the Late Devonian or Early Carboniferous in parts of the source rocks (Fig. 3). In an easterly direction, the advancement of generation processes definitely decreased and to the east from the region between the Henrykowo-1 and Olsztyn IG-2 wells, the potential source rocks are too immature to have generated hydrocarbons. They did not even reach the beginning of the oil window.

In a SE direction, the lateral variation of the generating processes is similar to that existing in the Baltic Basin, both in the Polik-Bodzanów Zone and in the Podlasie-Lublin area. In the Polik-Bodzanów Zone, entry into the oil and gas window took place in the Silurian – as was shown in the model for the Bodzanów IG-1 well (Fig. 4) – owing to the proximity of the TTZ. These processes reached the overmature phase in the Devonian or maybe in the Silurian. Further to the east, entry into the oil window occurred in the Middle Devonian in the area of the Okuniew IG-1 borehole (Fig. 4), and the gas window could have been reached in the Carboniferous, depending on the amount of burial (Fig. 4). Even further to the east, entry into the oil window occurred in the Carboniferous (Parczew IG-10), no longer reaching the phase of generating condensate or dry gas in the window (Fig. 5). The development of these processes in the SE part of the Lublin area was somewhat different, where the entry into the oil window already had occurred in the Early Devonian – as was shown in the model for the Łopiennik

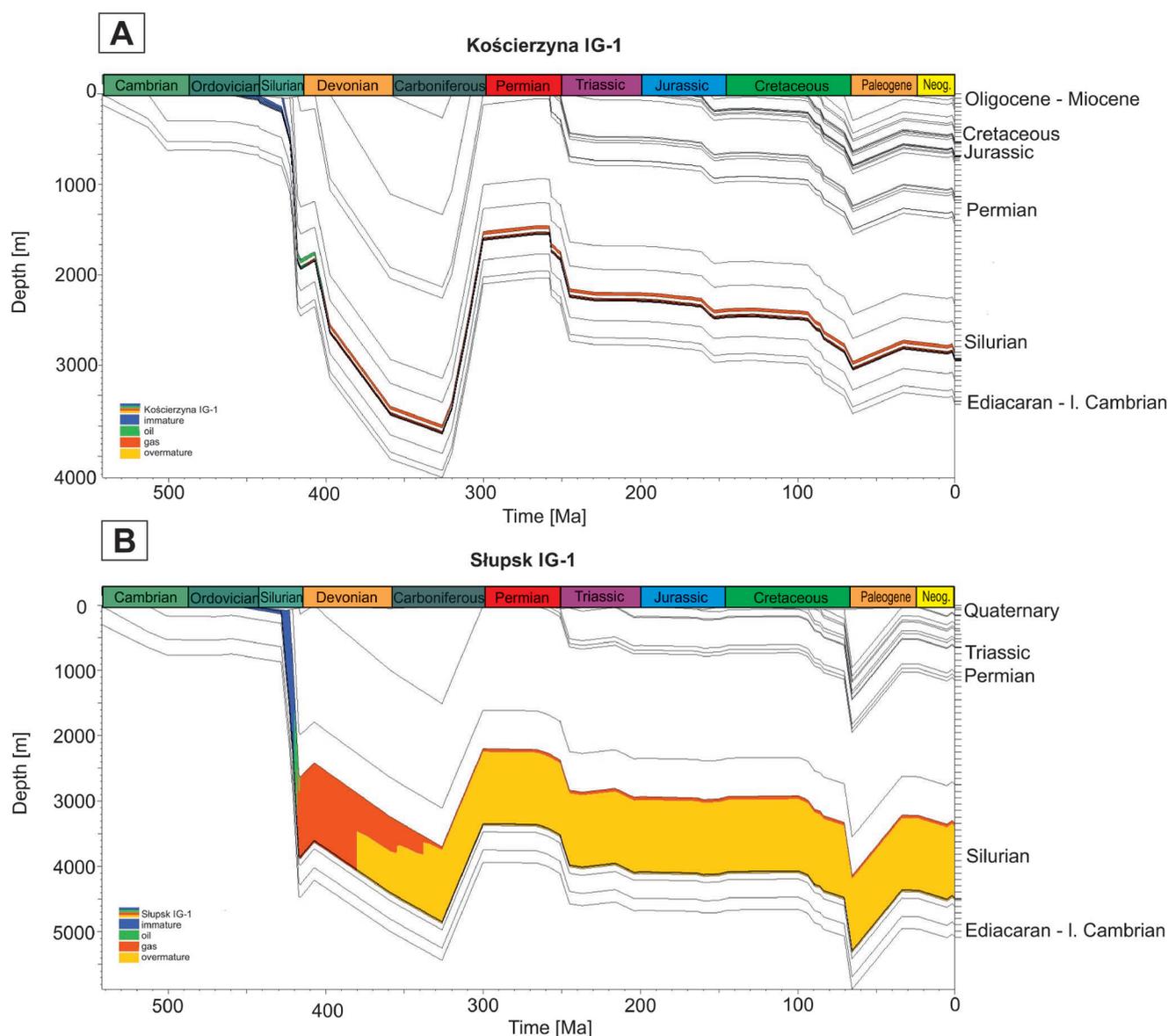


Fig. 2. Petroleum generation history in wells: Kościerzyna IG-1 (A) and Słupsk IG-1 (B). Based on Botor *et al.* (2017c), modified.

IG-1 well (Fig. 5). The gas window phase was obtained in the Late Devonian. In the Late Carboniferous, Lower Palaeozoic source rocks reached the overmature phase (Fig. 5).

Therefore, in the entire SW margin of the EEC, generating processes developed in a similar manner; in the direction from the NE to the TTZ, their intensity decreases and the beginning of subsequent stages of advancement was even later, from the end of the Late Silurian to the Late Carboniferous. In the Baltic Basin, hydrocarbon generation processes in the Lower Palaeozoic source rocks developed from the end of the Silurian to Early Carboniferous (Figs 2–3). In the Podlasie-Lublin Basin, however, they continued into the Late Carboniferous (Figs 4–5). These processes ended together with the Variscan tectonic inversion and were not resumed during the Mesozoic era. The generation processes were accompanied by the expulsion of hydrocarbons. This expulsion removed most of the hydrocarbons from the source rocks during the time of generation or with a small delay (Botor, 2016; Botor *et al.*, 2017c).

Kerogen type

Another problem in correctly determining hydrocarbon potential is the use of currently measured values of geochemical parameters (e.g., TOC, HI). This causes significant errors in the estimation of these geochemical parameters, resulting in the incorrect assessment of hydrocarbon resources. Therefore, the initial source rock and the original values, preceding the thermogenic hydrocarbon generation processes (from the immature stage), should be used to model the processes of hydrocarbon generation and to calculate the amount of gas and oil that was formed (e.g., Mann and Zweigel, 2008).

These primary parameters include the amount and quality of the kerogen contained in a given source rocks. Significant changes in the amount of hydrocarbons generated are observed with the introduction of different values of the TOC and the HI value. The amount of hydrocarbons generated changes directly with the accepted values of both HI

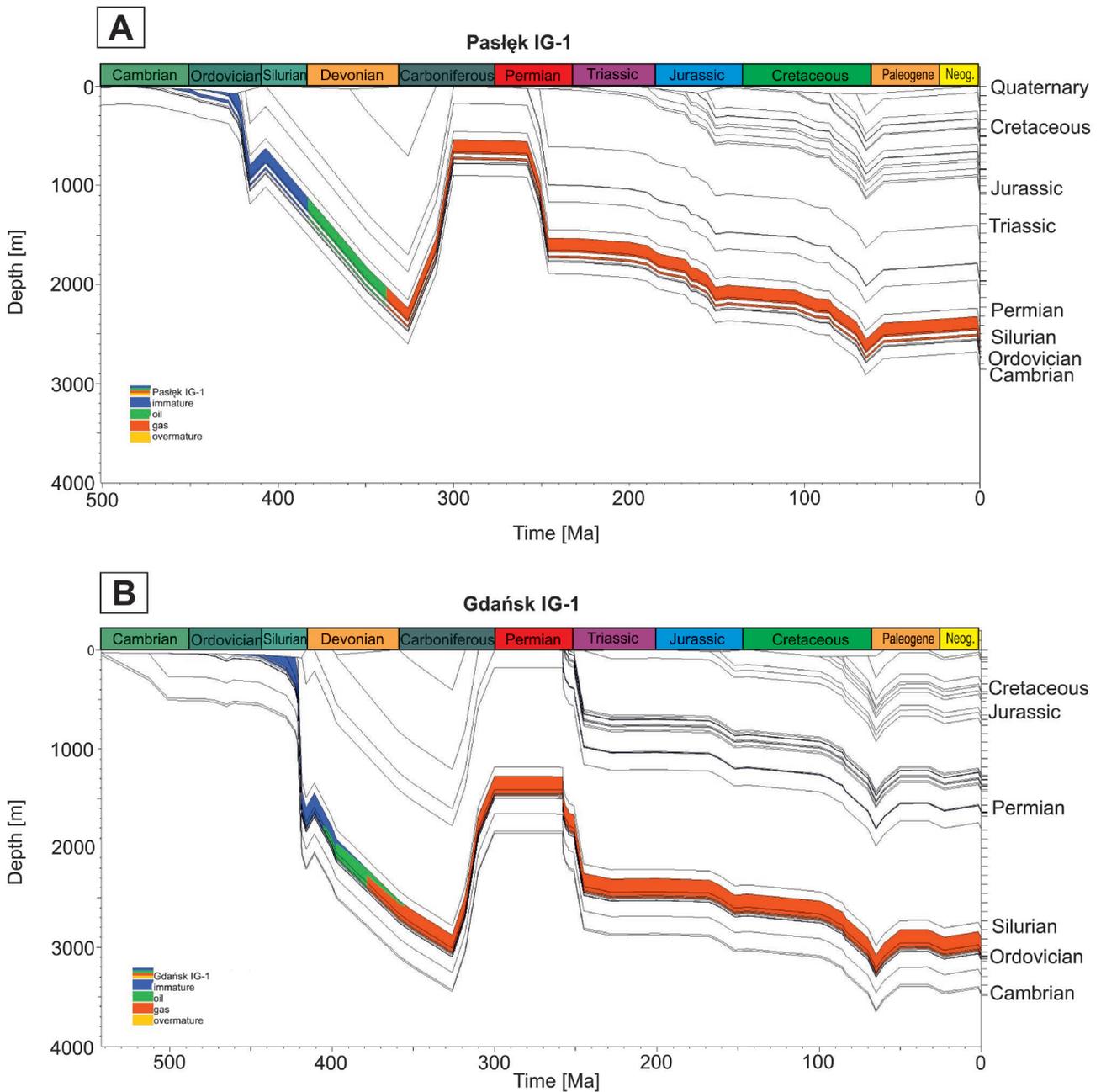


Fig. 3. Petroleum generation history in wells: Pasłek IG-1 (A) and Gdańsk IG-1 (B). Based on Botor *et al.* (2017c), modified.

and TOC. In particular, the amount of organic carbon in the Lower Palaeozoic shales shows a large spread (from 0 to 20% onwards), although the average values are well below 2.0%. Oil-prone type II of kerogen (algae-marine) widely accepted for organic matter in the Lower Palaeozoic shales is in fact a mixture of various components including both phytoplankton, zooplankton and bacterial components deposited mainly in marine environments (e.g., Hunt, 1996).

The Lower Palaeozoic sediments are dominated by the typical low-sulphur type II of kerogen, whereas the share of terrestrial type III (humic gas-prone) is excluded, because there were no higher terrestrial plants at that time that could produce biomass of this type (Hunt, 1996). Occurrence of the I type of kerogen seems to be poorly documented (Paczeńska

et al., 2005; Więclaw *et al.*, 2010a, 2012; Kosakowski *et al.*, 2016). Nevertheless, even within the type II of kerogen, it is possible to differentiate the original organic matter, which is visible in the variation of the hydrogen index. If we exclude the variability associated with the increase in thermal maturity in the thermally immature kerogen, the original HI depends primarily on the initial characteristics of the organic matter deposited and its facies diversity (e.g., Mann and Zweigel, 2008).

Kerogen transformation

The extent of advanced hydrocarbon generation processes is best represented by the transformation ratio (TR), expressed as %, showing the degree of thermogenic trans-

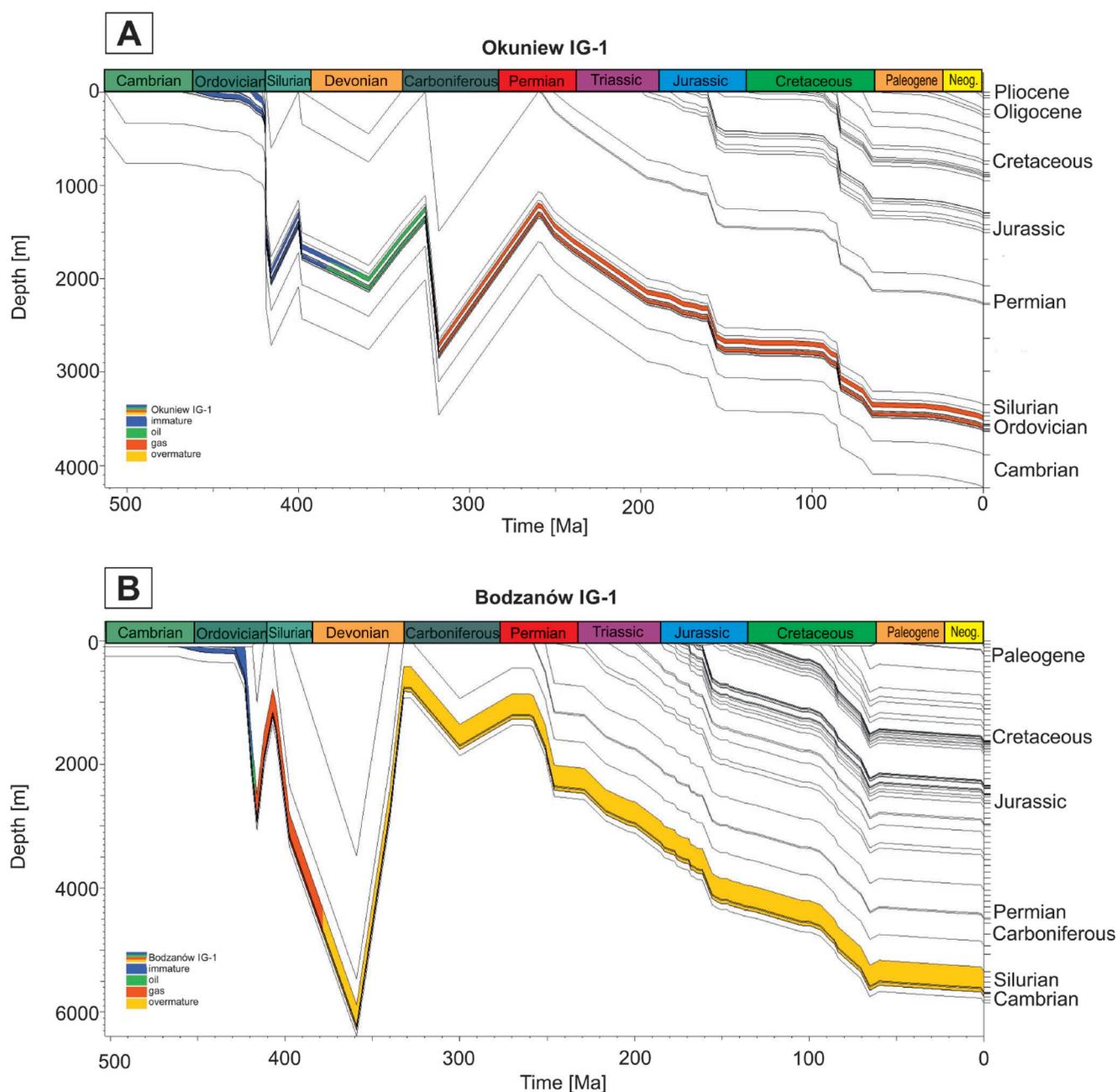


Fig. 4. Petroleum generation history in wells: Okuniew IG-1 (A) and Bodzanów IG-1 (B). Based on Botor *et al.* (2017c), modified.

formation of kerogen, which goes through the processes of hydrocarbon generation (e.g., Botor and Kosakowski, 2000). In the Ordovician–Lower Silurian source rocks, in the Baltic Basin, the maximum TR was reached at the end of the Early Carboniferous, while in the Lublin area at the end of the Late Carboniferous. In all basins, however, there is a significant increase in the intensity of hydrocarbon generation processes already in the Devonian, and in the most western part even with the end of the Silurian. In the Baltic Sea, in the Caradocian, the TR reached values from below 5% in the area between the Gołdap IG-1 and Bartoszyce IG-1 wells up to almost 100% in the area between the Kościerzyna IG-1 and Słupsk IG-1 wells (Fig. 6A). In the Llandovery source rocks, the TR reached values ranging from nearly zero percent in the area of the Bartoszyce IG-1

and Kętrzyn IG-1 wells to almost 95–100% in the area of the Kościerzyna IG-1 and Słupsk IG-1 boreholes (Fig. 6B).

In the Wenlock, TR values ranged from near-zero percent in the area of Gołdap IG-1 and Bartoszyce IG-1 wells to almost 100% in the area of the Kościerzyna IG-1 and Słupsk IG-1 wells (Fig. 6C). In the Podlasie-Lublin area, in the Caradocian source rocks, the TR reached values below 1% in the area between Krzyże-4, Mielnik IG-1 and Stadniki IG-1 wells up to almost 100% near the area between the Bodzanów IG-1 and Polik IG-1 wells as well as in the area between the Łopiennik IG-1 and Narol IG-1 boreholes (Fig. 6A). In the Llandovery and Wenlock, the TR attained a similar range of values and spatial distribution (Fig. 6B, C), which is due to similar depths of the three source rocks investigated. Thus, the TR indices increase westwards in all

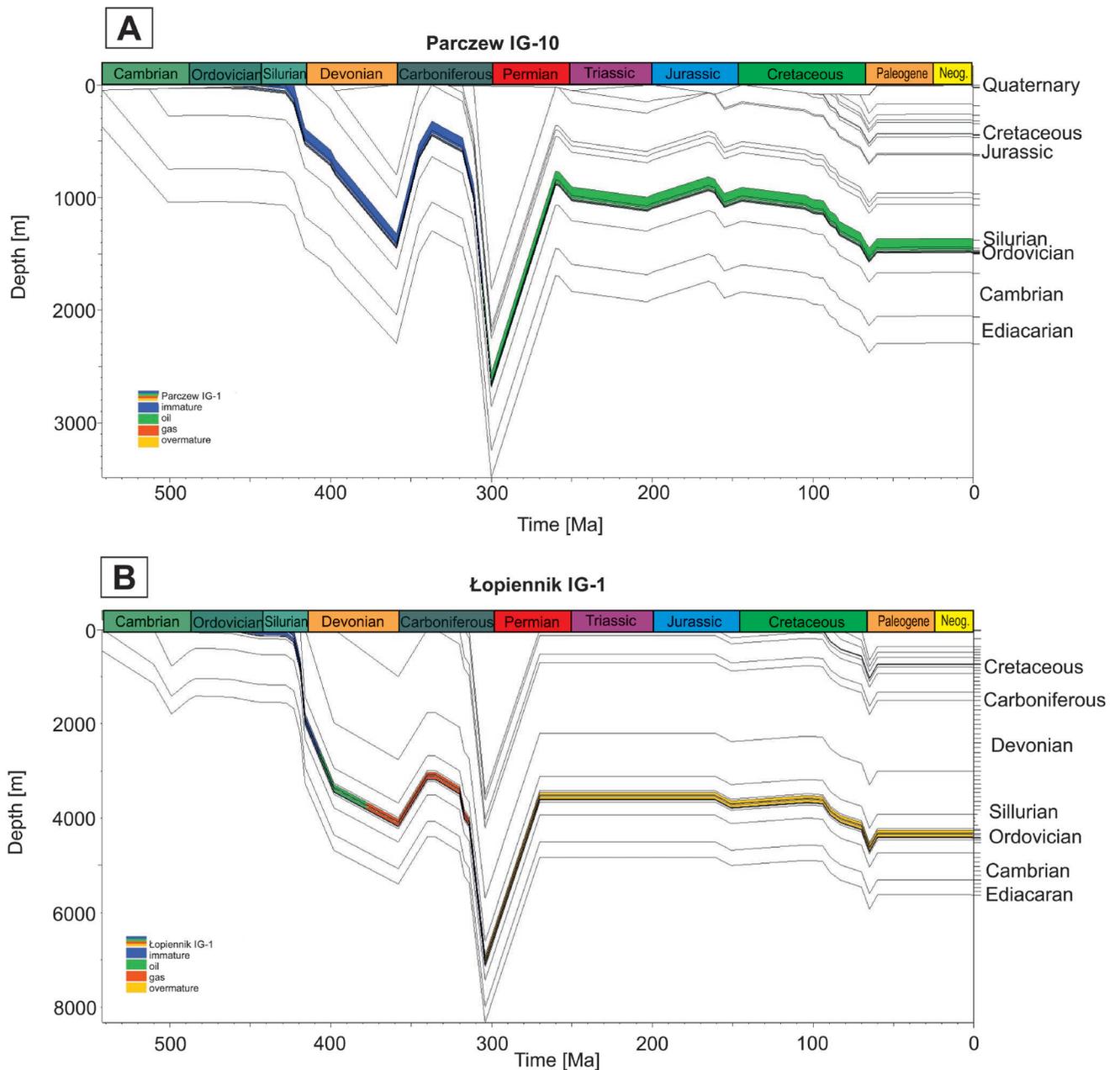


Fig. 5. Petroleum generation history in wells: Parczew IG-10 (A) and Łopiennik IG-1 (B). Based on Botor *et al.* (2017c), modified.

source rocks analysed throughout the entire SW region of the slope of the EEC (Fig. 6).

Generation potential

For the generation potential, only average regional data were used on the basis of data available in the public domain. Thus, the characteristics presented are qualitative, not quantitative, showing regional trends in individual source rocks in particular oil basins. The hydrocarbon generation potential of the Caradocian source rocks for gas generation, calculated for the end of the Early Carboniferous, reached values close to zero in the area of the Gołdap IG-1 and Bartoszyce IG-1 wells in the Baltic Basin and the Mielnik IG-1 and Stadniki IG-1 wells in the Podlasie Basin (Fig. 7A).

However, in the Baltic Basin, the generation potential increases to ca. 30–40 mg hydrocarbons/g TOC for the area between the Olsztyn IG-2, Henrykowo-1 and Młynary-1 wells (3–15 mg hydrocarbons/g TOC) westwards. In the Baltic Basin, the highest values were calculated in the area between the Słupsk IG-1 and Kościerzyna IG-1 wells (about 90–100 mg hydrocarbons/g TOC) (Fig. 7A). In the Podlasie-Lublin region, the generation potential calculated at the end of the Late Carboniferous also increases towards the SW, reaching a maximum value (over 90 mg hydrocarbons/g TOC) in the area between the Łopiennik IG-1 and Narol IG-1 wells (Fig. 7A).

The potential of the Caradocian source rocks for oil generation, calculated for the end of the Early Carboniferous, reached values close to zero in the area between the Gołdap

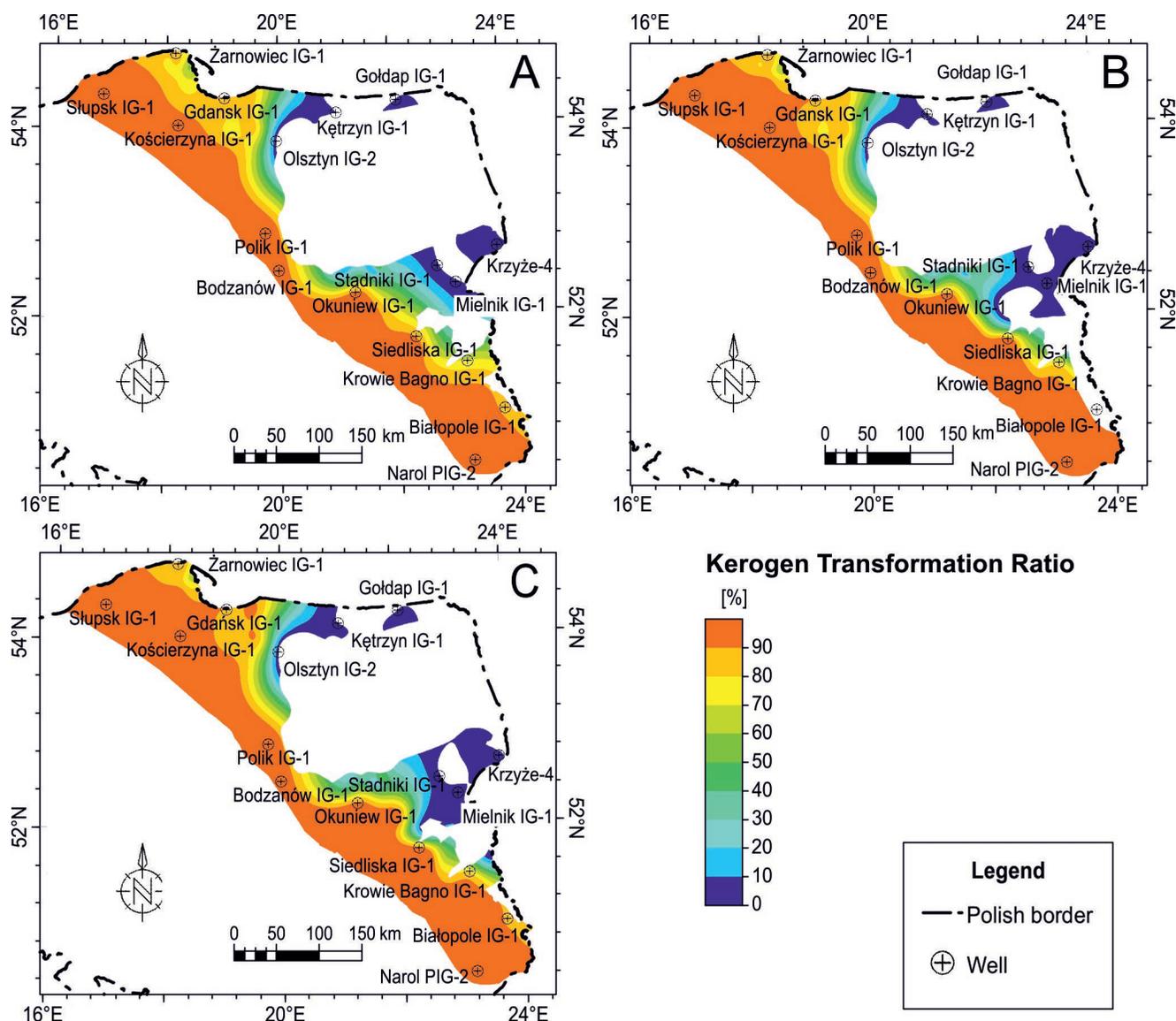


Fig. 6. Kerogen transformation in the Caradocian source rocks (A), in the Llandovery source rocks (B), in the Wenlock source rocks (C). Based on Botor *et al.* (2017c), modified.

IG-1, Bartoszyce IG-1, Stadniki IG-1 and Mielnik IG-1 wells (Fig. 8A). However, in the Baltic Basin the generation potential increases westwards, from about 170 mg hydrocarbons/g TOC on the area between the Olsztyn IG-2 and Henrykowo-1 wells to about 450–500 mg hydrocarbons/g TOC in the belt extending from the Malbork IG-1 well through the Gdańsk IG-1 well to the area between the Kościerzyna IG-1, Żarnowiec IG-1 and Łeba-8 wells (Fig. 8A). In the Podlasie-Lublin region, the generation potential calculated at the end of the Late Carboniferous grows westwards and southwestwards, reaching over 400 mg of hydrocarbons per gram of TOC (Fig. 8A).

The generation potential of the Llandovery source rocks for gas generation calculated at the end of the Early Carboniferous has reached values close to zero in the area between the Gołdap IG-1, Bartoszyce IG-1 and Stadniki IG-1, Mielnik IG-1 wells (Fig. 7B). However, the generation potential increases westwards from 2–12 mg hydrocarbons/g

TOC the area between the Olsztyn IG-2, Henrykowo-1 and Młynary-1 wells, to a value of about 30 mg hydrocarbons/g TOC in the Baltic Basin. The highest values (90–100 mg hydrocarbons/g TOC) were recorded in the area between the Słupsk IG-1 and Kościerzyna IG-1 wells (Fig. 7B). In the Podlasie-Lublin region, the generation potential calculated at the end of the Late Carboniferous also grows towards the west and SW, reaching more than 90 mg of hydrocarbons/g TOC in the area between wells Łopiennik IG-1, LK1 and Polik IG-1, Bodzanów IG-1 (Fig. 7B).

The generation potential of the Llandovery source rocks for oil generation calculated at the end of the Early Carboniferous has reached values close to zero in the area between the Gołdap IG-1, Bartoszyce IG-1 and Stadniki IG-1, Mielnik IG-1 wells (Fig. 8B). However, the generation potential increases westwards in a belt extending from Malbork through Gdańsk from ca. 100 mg hydrocarbons/g TOC in the area between the Olsztyn IG-2 and Henrykowo-1 wells,

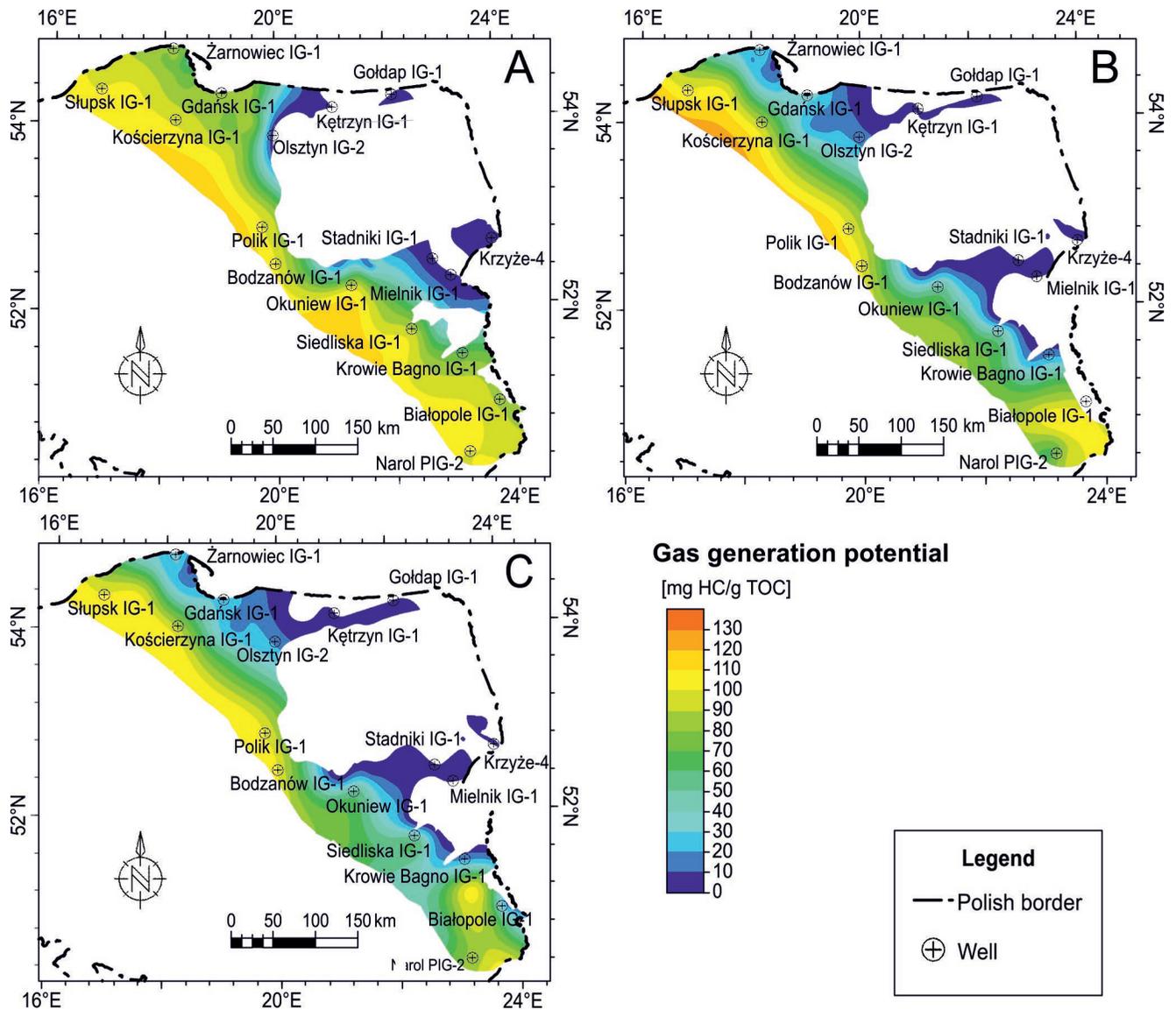


Fig. 7. Gas generation potential in the Caradocian source rocks (A), in the Llandovery source rocks (B), in the Wenlock source rocks (C). Based on Botor *et al.* (2017c), modified.

to 50–500 mg hydrocarbons/g TOC in the area between the Kościerzyna IG-1, Żarnowiec IG-1 and Łeba-8 wells (Fig. 8B). In the Podlasie-Lublin region, the generation potential calculated for the end of the Late Carboniferous grows to the west and SW, reaching more than 400 mg of hydrocarbons/g TOC in the area of the Łopiennik IG-1, LK1, Polik IG-1 and Bodzanów IG-1 wells (Fig. 8B).

In the Baltic Basin, the generation potential of the Wenlock source rock for gas generation calculated for the end of the Early Carboniferous has reached values close to zero in the area between the Goldap IG-1, Bartoszyce IG-1 and Stadniki IG-1, Mielnik IG-1 wells (Fig. 7C). However, the generation potential grows westwards from 1–10 mg hydrocarbons/g TOC in the area between the Olsztyn IG-2, Henrykowo-1 and Młynary-1 wells to about 15–30 mg hydrocarbons/g TOC in the area between Malbork IG-1 and Gdańsk IG-1 wells, in the Baltic basin. The highest values (80–100 mg hydrocarbons/g TOC) were recorded in the

area between the Kościerzyna IG-1 and Słupsk IG-1 wells (Fig. 7C). In the Podlasie-Lublin region, the generation potential calculated in the end of the Late Carboniferous grows in the west and SW, reaching more than 100 mg of hydrocarbons/g TOC in the area between the Łopiennik IG-1, LK and Polik IG-1, Bodzanów IG-1 wells (Fig. 7C).

The generation potential of the Wenlock source rocks for oil production, calculated for the end of the Early Carboniferous, reached values close to zero in the area between the Goldap IG-1, Bartoszyce IG-1 and Stadniki IG-1, Mielnik IG-1 wells (Fig. 8C). However, the generation potential increases westwards in the zone between the Malbork IG-1 and Gdańsk IG-1 wells from about 70 mg hydrocarbons/g TOC in the area of the Olsztyn IG-2 and Henrykowo-1 boreholes in the west, to values in the range of 440–490 mg hydrocarbons/g TOC in the area of Kościerzyna IG-1, Żarnowiec IG-1 and Łeba-8 wells (Fig. 8C). In the Podlasie-Lublin region, the generation potential calculated for the end

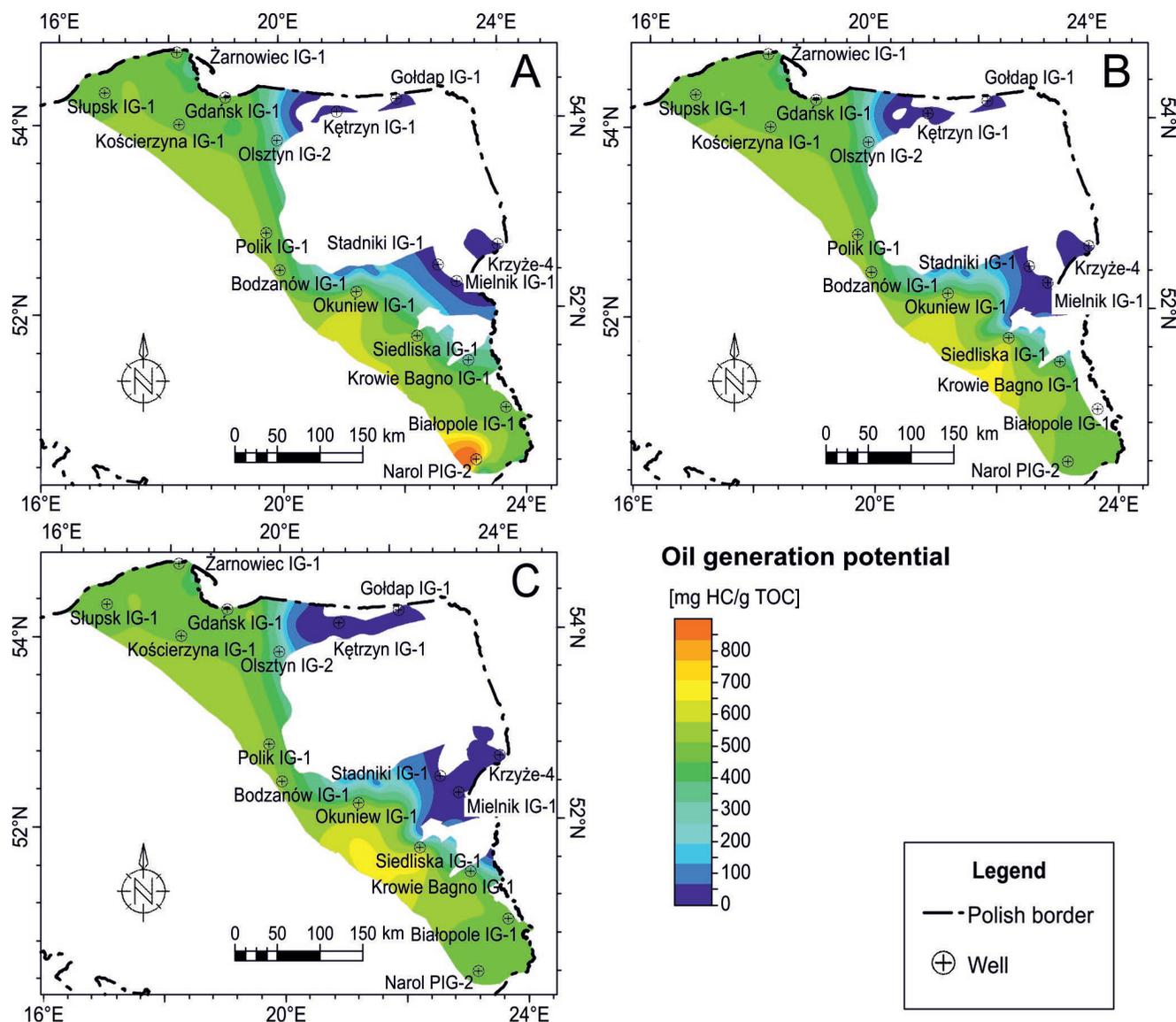


Fig. 8. Oil generation potential in the Caradocian source rocks (A), in the Llandovery source rocks (B), in the Wenlock source rocks (C). Based on Botor *et al.* (2017c), modified.

of Carboniferous grows in the west and SW, reaching more than 400 mg hydrocarbons/g TOC in the area between the Łopiennik IG-1, LK1 and Polik IG-1, Bodzanów IG-1 wells (Fig. 8C). The local maximum generation potential was also calculated in the zone between the Narol IG-1 and Dyle IG-1 wells (Fig. 8C).

Hydrocarbon expulsion

In the boreholes analysed, expulsion was commonly associated with the hydrocarbon generation processes with a slight delay (Botor, 2016, 2018; Botor *et al.*, 2017b). Expulsion did not occur only in the eastern part of the Baltic Basin (east of the Olsztyn IG-2 region) and the Podlasie-Lublin Basin (NE from the area between the Tłuszcz IG-2 and Mielnik IG-1 wells; Figs 9–10), where the source rocks did not reach an advanced stage of kerogen transformation. The time of expulsion varies from the latest Silurian (along the

TTZ) to the Late Devonian and/or Carboniferous in the central part of the analysed basins. The following description of the hydrocarbon expulsion rate with the division into natural gas and crude oil is only qualitative. The current stage of research allows only such an approach.

In the Baltic Basin, the rate of gas expulsion from the Caradocian source rocks reached values below 10 Mtons/km²/Ma in almost the entire research area (Fig. 9A). Only in the area between the Leba-8 and Słupsk IG-1 wells gas expulsion is higher (up to 15 Mtons/km²/Ma) (Fig. 9A). However, the rate of oil expulsion from Caradocian source rocks reached values not exceeding 1 Mtons/km²/Ma in the east of the research area (Fig. 10A). From the area between the Henrykowo-1 and Młynary-1 wells, it increases westwards. In the zone between the Żarnowiec IG-1 and Słupsk IG-1 wells, these values increase from about 20 to ca. 80 Mtons/km²/Ma. High values are also in the Kościerzyna IG-1 zone (60 Mtons/km²/Ma) (Fig. 10A).

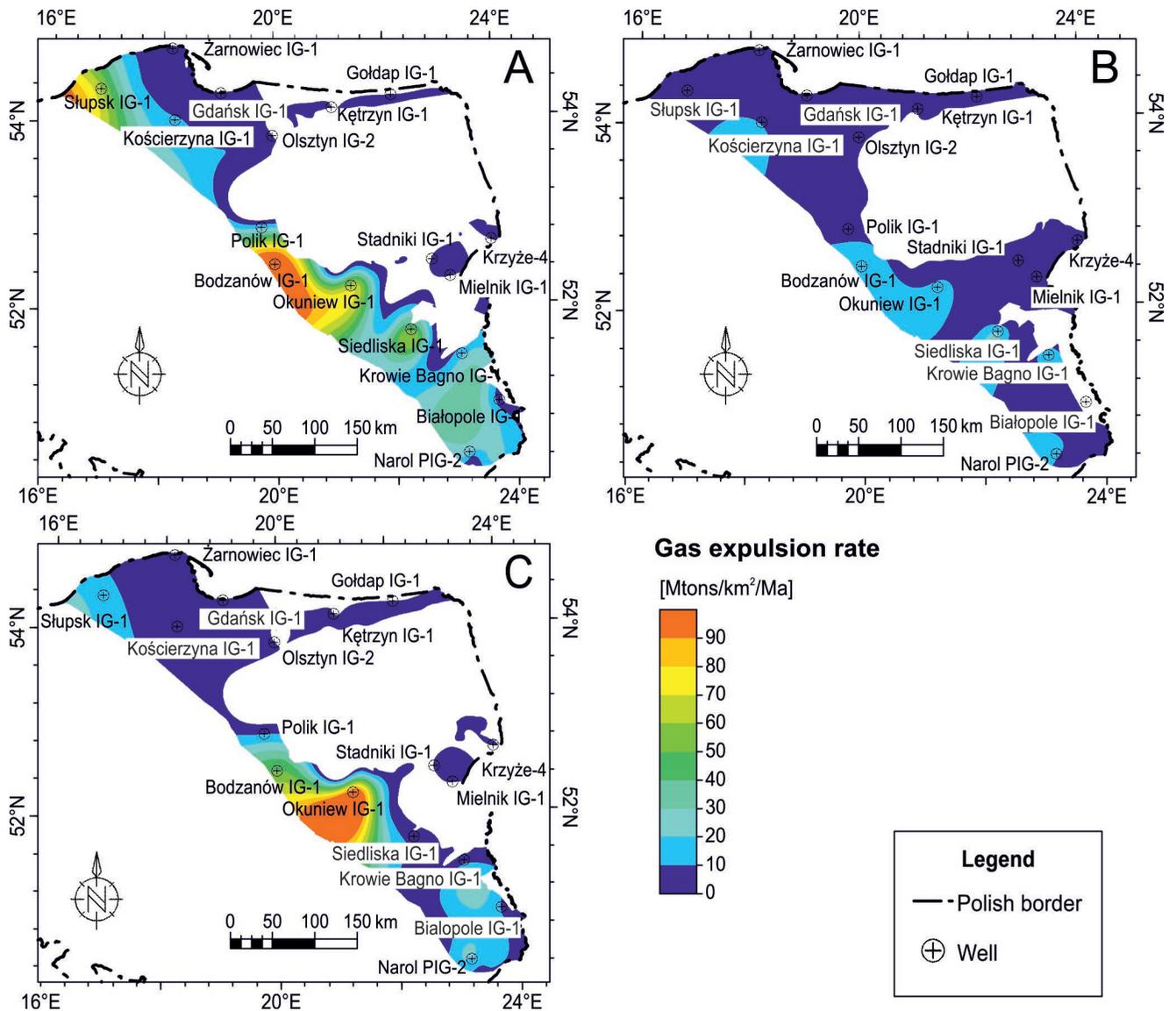


Fig. 9. Gas expulsion rate in the Caradocian source rocks (A), in the Llandovery source rocks (B), in the Wenlock source rocks (C). Based on Botor *et al.* (2017c), modified.

In the Polik-Bodzanów zone and in the Podlasie Basin, the rate of gas expulsion from the Caradocian source rocks reached values close to zero in the entire NE area in the zone between the Krzyże-4, Stadniki IG-1 and Mielnik IG-1 wells (Fig. 9A). Expulsion values increase in a westerly direction to 50–100 Mtons/km²/Ma in the area between the Okuniew IG-1 and Bodzanów IG-1 wells (Fig. 9A). Oil expulsion from the Caradocian source rocks reached values not exceeding 1 Mtons/km²/Ma in the east of the research area (Fig. 10A). In a westerly direction, however, the values rise to over 200 Mtons/km²/Ma in the area between the Okuniew IG-1 and Bodzanów IG-1 wells (Fig. 10A).

In the Lublin Basin, the rate of gas expulsion from the Caradocian source rocks reached low values in the NE part of the study area (Fig. 9A). The rates of the expulsion increase in a westerly direction to 20 Mtons/km²/Ma in the area between the Busówno IG-1, Łopiennik IG-1 and Narol IG-1 wells (Fig. 9A). Oil expulsion from the Caradocian source rocks reached values not exceeding 1 Mtons/km²/Ma

in the eastern part of the research area. In western and southern directions, however, the values increase to over 50 Mtons/km²/Ma in the Narol IG-1 region (Fig. 10A).

In the Baltic Basin, the rate of gas expulsion from the Llandovery source rocks reached values close to zero or not exceeding 1 Mtons/km²/Ma in the eastern part of the study area (Fig. 9B). On the other hand, west of the line between the Młynary-1 and Olsztyn IG-2 boreholes, the value of expulsion is higher than 1 Mtons/km²/Ma and increases further to the west. In the area between the Słupsk IG-1 and Kościerzyna IG-1 wells, it reaches 6–10 Mtons/km²/Ma (Fig. 9B). Oil expulsion from Llandovery source rocks reached values not exceeding 1 Mtons/km²/Ma in the eastern part of the study area (Fig. 10B), whereas from the area between the Gdańsk IG-1 and Malbork IG-1 wells, it increased westwards. In the zone between the Żarnowiec IG-1 and Słupsk IG-1 wells, these values increased to ca. 100 Mtons/km²/Ma. The highest values are in the region of

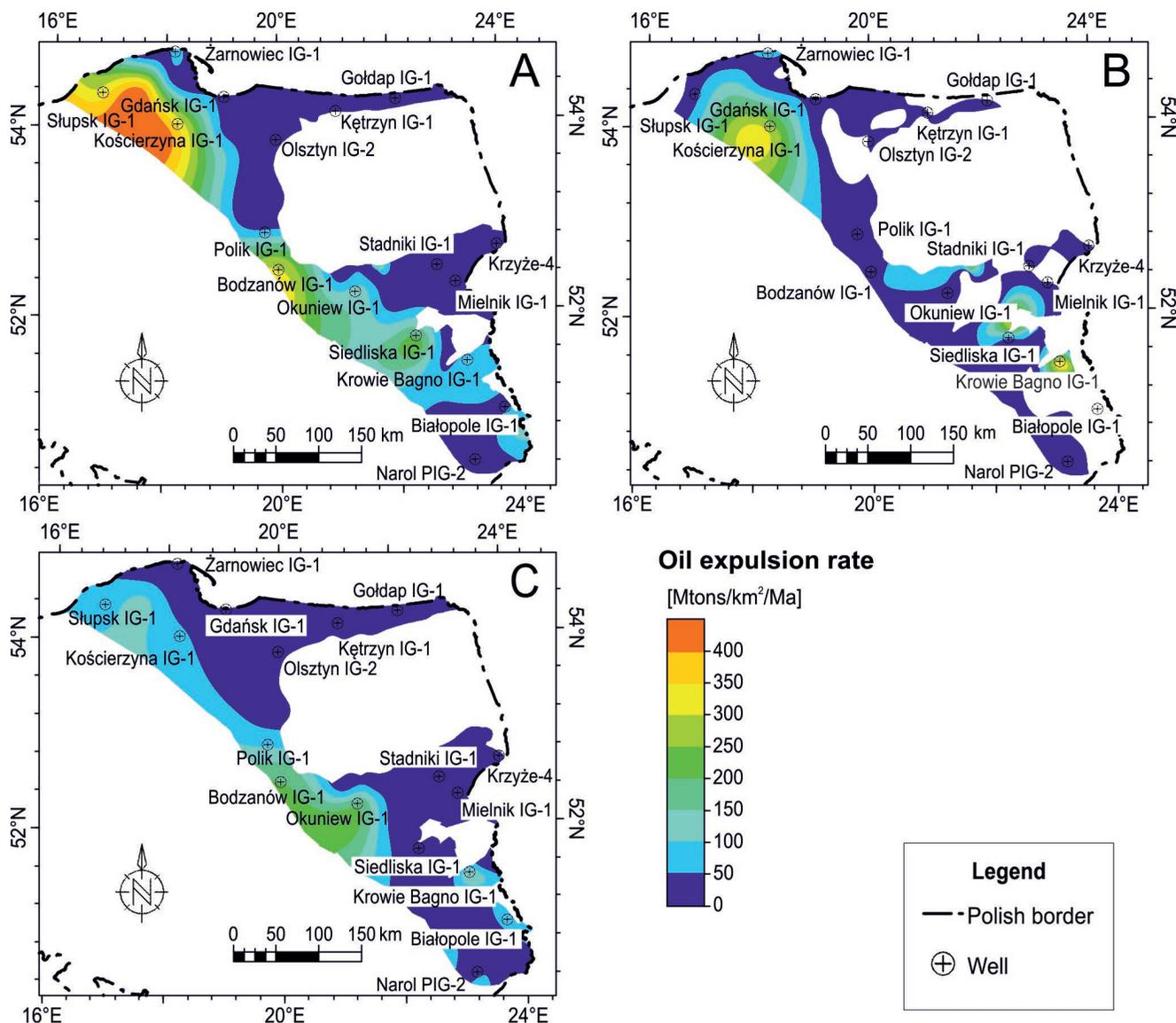


Fig. 10. Oil expulsion rate in the Caradocian source rocks (A), in the Llandovery source rocks (B), in the Wenlock source rocks. Based on Botor *et al.* (2017c), modified.

the Kościerzyna IG-1 well, exceeding 200 Mtons/km²/Ma (Fig. 10B).

In the Polik-Bodzanów zone and in the Podlasie Basin, the rate of gas expulsion from the source rocks reached values close to zero in the entire eastern area between the Krzyże-4, Stadniki IG-1 and Mielnik IG-1 wells (Fig. 9B). Expulsion values increased in the western direction to over 15 Mtons/km²/Ma in the area of the Bodzanów IG-1 well (Fig. 9B). The rate of oil expulsion from the Llandovery source rocks reached similar values, not exceeding 1 Mtons/km²/Ma in the eastern part of the research area (Fig. 10B). In the western direction, however, the values increased to 20–50 Mtons/km²/Ma in the area between the Okuniew IG-1 and Go wells with a maximum value of over 100 Mtons/km²/Ma in the area of the Łochów IG-2 well (Fig. 10B).

In the Lublin area, the rates of gas expulsion from the Llandovery source rocks had values close to zero in the entire eastern area (Fig. 9B). Expulsion rates increased to-

wards the SW to over 15 Mtons/km²/Ma, with local maxima in the area between the Dyle IG-1, Narol IG-1 and Siedliska IG-1 wells (Fig. 9B). The rates of oil expulsion from Llandovery source rocks also had values close to zero in the eastern area, while in a westerly direction these values rose to create local peaks in the area of the Siedliska IG-1 and Krowie Bagno IG-1 wells (Fig. 10B).

In the Baltic Basin, the rates of gas expulsion from the Wenlock source rocks have values close to zero or not exceeding 1 Mtons/km²/Ma in the eastern area (Fig. 9C). However, west of the area between the Malbork IG-1 and Gdańsk IG-1 wells, the expulsion value grows westwards, reaching 60 Mtons/km²/Ma in the area of the Słupsk IG-1 well. The oil expulsion from the Wenlock source rocks reached values close to zero or not exceeding 1 Mtons/km²/Ma in the eastern part of the research area (Fig. 10C). However, these values increase westwards from the area between the Gdańsk IG-1 and Malbork IG-1 wells (ca. 50 Mtons/km²/Ma). In the zone between the Żarnowiec IG-1 and

Słupsk IG-1 wells, these values increase from ca. 60 to 320 Mtons/km²/Ma. The highest values are in the area between the Słupsk IG-1 and Kościerzyna IG-1 wells (up to ca. 330 Mtons/km²/Ma (Fig. 10C).

In the Polik-Bodzanów zone and in the Podlasie Basin, the rate of gas expulsion from the Wenlock source rocks have values close to zero in the entire eastern area between the Krzyże-4, Stadniki IG-1 and Mielnik IG-1 wells (Fig. 9C). The expulsion rate increases in the westerly direction to about 100 Mtons/km²/Ma in the Bodzanów IG-1 well (Fig. 9C). The oil expulsion rates from the Wenlock source rocks have values not exceeding 1 Mtons/km²/Ma in the eastern area (Fig. 10C). In a westerly direction, however, the values increase to 300 Mtons/km²/Ma in the area of the Bodzanów IG-1 well (Fig. 10C).

In the Lublin area, the rates of gas expulsion from the Wenlock source rocks have values close to zero in the entire eastern area (Fig. 9C). The values of expulsion rate increase in a westerly direction to ca. 60–70 Mtons/km²/Ma in the region of the Siedliska IG-1 well (Fig. 9C). The rates of oil expulsion from the Wenlock source rocks have values not exceeding 1 Mtons/km²/Ma in the eastern area (Fig. 10C). In a westerly direction, however, the values rise to about 200 Mtons/km²/Ma in the area of the Siedliska IG-1 well (Fig. 10C).

Finally, it is worth mentioning that owing to the lack of detailed geochemical and hydrocarbon sorption data in the source rocks studied, the assessment of expulsion should be considered as ambiguous and not very precise. It only shows some general, regional trends of change and the zones of expulsion without any detailed quantification.

Implications for petroleum exploration

In summary, in the area of the Baltic Basin, the degree of kerogen transformation and the generation potential increase from the east (from the Gołdap IG-1 well) to the west (to the Słupsk IG-1 well) in a more or less regular manner. This conclusion applies to all source rocks analysed (Wenlock, Llandovery, and Caradocian). The eastern zone of the basin in the area from the Gołdap IG-1 well to the area around the Henrykowo-1 well shows negligible potential (close to zero), owing to the very low transformation of kerogen. However, in the area between the Henrykowo-1 and Młynary-1 wells, the generation potential clearly increases towards the west. However, in the westernmost area (the Słupsk IG-1 well) the Lower Palaeozoic source rocks analysed are overmature in relation to the generation of hydrocarbons.

In the Polik-Bodzanów Zone and in the Podlasie Basin, the kerogen transformation degree and generation potential increase also substantially from east (and NE) (Krzyże-4) to west (and SW) towards the area between the Okuniew IG-1 and Bodzanów IG-1 wells in all source rocks analysed. The eastern zone, in the area between the Krzyże-4 borehole to the Stadniki IG-1 and Mielnik IG-1 wells, shows negligible potential, owing to the low transformation of kerogen. In the area between the Wrotnów IG-1, Łochów IG2 and Tłuszcz IG-1 wells, the generation potential is clearly increasing towards the west and southwest, reaching a maximum in the area around the Bodzanów IG-1 well.

In the Lublin Basin, the thermal maturity and, as a result, the kerogen transformation rate of the Lower Palaeozoic source rocks varies. However, the trend of increasing transformation of kerogen toward the SW is preserved, as in the Baltic, Polik-Bodzanów and Podlasie areas. The main period of generation and expulsion of hydrocarbons from the Lower Palaeozoic source rocks was mainly in the Devonian and Carboniferous. The scope of development of these processes clearly increases towards the south and southwest, reaching a maximum in the area between the Łopiennik IG-1, LK, and Narol IG-1 wells. Only in the NW part of the Mazowsze-Lublin Trough, that is in the area between the Nadarzyn IG-1, Wilga IG-1, and Warka IG-1 wells, there could have been local Jurassic–Cretaceous reheating associated with significant burial due to the development of the Polish Basin (Botor *et al.*, 2002) and the assumed effects of hot solutions (Kozłowska and Poprawa, 2004). In this zone, there is no significant coalification jump between the Carboniferous and the Permian–Mesozoic strata. In this zone, at the present stage of research, it can be assumed that hydrocarbon generation could have taken place both in the Palaeozoic and in the Mesozoic. However, there is no drilled profile of the entire Lower Palaeozoic, which makes detailed recognition impossible. In addition, in inverted basins with a rift-related origin (like the Polish Basin), a significant increase in the thickness of sediments in the late stages of development may even have caused a cooling effect associated with a decrease in thermal flux (e.g., Ceriani *et al.*, 2006). As a result, maximum temperatures do not occur during the maximum burial. This issue requires further research.

In the entire SW part of the EEC, hydrocarbon expulsion (both natural gas and oil) occurred in the period from the latest Silurian to the Late Carboniferous, only slightly delayed in relation to generation, in one and sometimes in several phases, usually slightly distant from one another. The earliest (Late Silurian) phases of generation and expulsion occurred in the westernmost zone between the Słupsk IG-1, Kościerzyna IG-1, Bodzanów IG-1 and Łopiennik IG-1 wells (close to the TTZ). To the east of this zone, the phase of generation and expulsion took place in the Devonian and partly in the Carboniferous. The process of expulsion occurred almost simultaneously with the generation of hydrocarbons or slightly later. Hydrocarbon expulsion occurred mainly in the western zone of the basins (generally west of the line of wells Gdańsk IG-1, Malbork IG-1, Tłuszcz IG-1, and Krowie Bagno IG-1). However, detailed quantification of this expulsion requires further geochemical studies and additional studies on the sorption of hydrocarbons. The hydrocarbons generated were greatly scattered, owing to the tectonic multiphase inversion of the Palaeozoic basins, especially the Variscan and Late Cretaceous–early Palaeogene inversion (e.g., Krzywiec, 2009; Krzywiec *et al.*, 2017a, b), and as a result only a small fraction of them could have survived to the present day. The hydrocarbons that were subject to retention as a result of absorption in the kerogen themselves had the greatest chance of preservation.

The size and intensity of hydrocarbon seepages from the Polish Lower Palaeozoic shales was relatively low. On the entire SW slope of the EEC analysed, gas seepages indicate

the presence of dry gas with a high methane content, containing no nitrogen or other non-hydrocarbon components, except for in the eastern part of the Polish shale zone, where nitrogen is present (Poprawa, 2010). The seepages of gas and oil have a zonality that corresponds approximately to the zones of kerogen thermal maturity, which determined the development of hydrocarbon generation processes (Poprawa, 2010).

The Baltic Basin has a relatively simple tectonic structure, while that of the Lublin Basin is much more complicated, which is a disadvantageous factor in relation to the possibility of preservation of the hydrocarbon accumulations. The study area is characterized by a relatively high degree of lithological consolidation and rock brittleness (Poprawa, 2010; Podhalańska *et al.*, 2016). The Lower Palaeozoic source rocks on the SW margin of the EEC show a lower TOC and lower thermal maturity at the optimal exploitation depth interval i.e. 1.5–3.5 km, compared to the US equivalents (Jarvie *et al.*, 2006; Poprawa, 2010).

CONCLUSIONS

The above discussion on the generation and expulsion of hydrocarbons demonstrates that the maturity of organic matter in the Lower Palaeozoic strata indicates the advancement of generation from the phase of low-temperature thermogenic processes in the NE part of the Baltic and Podlasie-Lublin basins to the overmature stage along the zone adjacent to the TTZ. The results of the modelling of generation and expulsion processes showed that they took place mainly in the Devonian and Carboniferous periods and in the westernmost part (along the TTZ), even in the latest Silurian. This applies to both the Baltic area and the Polik-Bodzanów Zone, the Podlasie-Lublin area and the Biłgoraj-Narol area. During the Mesozoic and Cainozoic eras, generation processes essentially were not resumed or intensified.

The Lower Palaeozoic sediments were subject to intense subsidence and rapid burial in the Palaeozoic and then were uplifted, in several phases, but with the predominance of the late Variscan tectonic inversion. Unfortunately, the generation of hydrocarbons and their expulsion took place only in the Palaeozoic (latest Silurian to Carboniferous), at least 300 million years ago. Most of the hydrocarbons were subjected to expulsion and possible migration. As a result, there was a large dispersion of the hydrocarbons generated. The chance of preservation of these hydrocarbons in the source rocks is small. As a result, the calculated saturation of shale hydrocarbons is relatively low (e.g., Botor, 2016), which also is confirmed by the results of new drilling in the EEC area.

Nevertheless, it was found that zones with an increased shale gas potential can only occur in a narrow belt on the SW slope of the EEC, parallel to the edge of the TTZ. The most promising seem to be the Caradocian, Llandovery and Wenlock source rocks in the zone between the Lębork IG-1 and Kościerzyna IG-wells in the Baltic Basin and the Wenlock source rocks in the Podlasie-Lublin Basin between the Okuniew IG-1, Go, Łopiennik IG-1 and Narol IG-1 wells.

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