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Integrated stratigraphy of the Llandovery-Wenlock Boundary in the Łopianka–2 outcrop of the Sudeten Mountains, southwest Poland

SIGITAS RADZEVIČIUS, PAWEŁ RACZYŃSKI, ANDRIUS GARBARAS, ANNA CICHON-PUPIENIS AND TOMAS ŽELVYS

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Stable carbon isotopic composition of organic matter ($\delta^{13}C_{org}$) analyses were carried out along the 7-m-thick section of Lower Graptolitic Shales at the Llandovery/Wenlock boundary, outcropping on Łopianka Mountain (the Łopianka-2 outcrop) in the Bardo Mountains of the Central Sudetes, Southwest Poland (eastern part of the European Variscides Belt). This study presents the first attempt to establish integrated biostratigraphical and chemostratigraphical records for Silurian strata in the Bardo Mountains. Graptolite assemblages indicate the presence of *centrifugus* and *murchisoni* biozones at the Telychian-Sheinwoodian boundary and mid-Wenlock in the investigated interval, thus continuous graptolitic succession. The succession of graptolite biozones in the upper Wenlock section could not be determined due to the collapsed strata. The isotopic signature of δ13Corg showed a positive excursion which is referred to as the Ireviken or early Sheinwoodian Carbon Isotope Excursion (ESCIE). The $\delta^{13}C_{org}$ values of the Ireviken interval begin to rise higher than the first occurrence of *Cyrt. bohemicus* and does not coincide with the base of the *murchisoni* Biozone. Due to the fact that coupled carbon isotope chemostratigraphy and graptolite biostratigraphy for Silurian strata is a new approach in this region, this may serve as a standard for the Llandovery/Wenlock boundary in the area of the Saxothuringian Zone of the Central European Variscides. □ *Graptolite, Carbon isotopes, Silurian, Telychian – Sheinwoodian boundary, Łopianka Mountain, Bardo Mountains*

Sigitas Radzevičius ✉ [sigitas.radzevicius@gf.vu.lt] Institute of Geological Sciences, University of Wrocław, Pl. Maksa Borna 9, Wrocław 50-205, Poland; Department of Geology and Mineralogy, Vilnius University, M. K. Čiurlionio 21/27, 03101 Vilnius, Lithuania; Paweł Raczyński [pawel.raczynski@uwr.edu.pl] Institute of Geological Sciences, University of Wrocław, Pl. Maksa Borna 9, Wrocław 50-205, Poland; Andrius Garbaras [andrius.garbaras@ftmc.lt] Department of Nuclear Research, Center for Physical Sciences and Technology, 10221 Vilnius, Lithuania; Anna Cichon-Pupienis [anna.cichon-pupienis@gamtc.lt] Laboratory of Bedrock Geology, Nature Research Centre, Akademijos str. 2, 08412 Vilnius, Lithuania; Tomas Želvys [tzelvys@gmail.com] Department of Geology and Mineralogy, Vilnius University, M. K. Čiurlionio 21/27, 03101 Vilnius, Lithuania;
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Stable carbon isotopes ($\delta^{13}C$) are often used as a chemostratigraphical tool, and are mostly used alongside biostratigraphical data. The first studies of $\delta^{13}C$ variability in Silurian rocks started in the end of the last century (e. g. Corfield *et al.* 1992; Samtleben *et al.* 1996; Wenzel & Joachimski 1996; Kaljo *et al.* 1997). The increasing availability of $\delta^{13}C$ data made it possible to create a generalized variability of $\delta^{13}C$ for the Silurian in recent decades, with widely recognized positive $\delta^{13}C$ excursions being in the early and late Aeronian, early Telychian (Valgu), Telychian-Sheinwoodian boundary (Ireviken), late Homerian (Mulde), early Ludfordian (Linde), late Ludfordian (Lau) and Silurian-Devonian boundary interval (Klonk) (e.g. Cramer *et al.* 2011; Melchin *et al.* 2020). These Silurian positive $\delta^{13}C$ excursions are mostly obtained by $\delta^{13}C$ measurements from bulk carbonates.

The stable carbon isotope data from organic ($\delta^{13}C_{org}$) material are rare from Silurian rocks (e.g. Vandenbroucke 2013; Sullivan *et al.* 2018; Cichon-Pupienis *et al.* 2021; Hartke *et al.* 2021). This is probably related to the specific method of preparation of samples for $\delta^{13}C_{org}$ analysis and content (concentration) of organic matter in rocks. On the other hand, some diachroneity can be presented in $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ records. Positive carbon excursions based on $\delta^{13}C_{org}$ data may start earlier than those based on $\delta^{13}C_{carb}$ (e.g. Biebesheimer *et al.* 2021). This creates inaccuracies in the design of high-resolution

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integrated stratigraphy. However, positive carbon isotopes excursions of the Telychian shales were determined based on $\delta^{13}C_{org}$ data from sparsely studied material from the Kallholn in Sweden (Walasek *et al.* 2018) and the Sommerodde in Denmark (Hammarlund *et al.* 2019; Loydell *et al.* 2023). These excursions are well integrated with graptolite biostratigraphy data, making $\delta^{13}C_{org}$ data important as it provides additional knowledge for high resolution stratigraphy.

The purpose of this paper is to present new data on the distribution and changing of Silurian graptolites combined with stable carbon isotope record from organic material ($\delta^{13}C_{org}$), from the Bardo Moutains (Sudetes), in an attempt to integrate new biostratigraphical data with $\delta^{13}C_{org}$ material. Integrated stratigraphy data allows us to determine the age of the Łopianka – 2 outcrop geological section. This is the first such study conducted in the Bardo mountains region and could be used as a standard or supporting Silurian geological section there. On the other hand, the Silurian geological section of the Łopianka – 2 outcrop could be a candidate for use among Standard Auxilliary Boundary Stratotypes (Head *et al.* 2023) for the Llandovery/Wenlock boundary in globally or a reference section for Saxothuringian Zone of Sudetes.

Geological background

The European Variscides Belt spreads from southern Portugal in the West to Central Poland in the East (Mazur *et al.* 2006). There are several zones distinguished in the Central European Variscides Belt. The Sudeten Mountains are in the East part of the European Variscides Belt. The Bardo Mountains (in Polish: Góry Bardzkie) are located in the Central Sudetes (Żelaźniewicz & Aleksandrowski 2008) and are assigned to the Saxothuringian Zone (Porębska & Sawłowicz 1997) of the Central European Variscides (Fig. 1A). Saxothuringia is a part of the Armorican Terrane Assemblage (Franke 2000) or a vestige of the Saxo-Thuringian Ocean (Franke *et al.* 2017). However, the Armorican Terrane Assemblage, with narrow oceans between terranes, has been located in the South part of the Rheic Ocean near Gondwana during the Silurian (Franke *et al.* 2017).

There are several Silurian outcrops in the Bardo Mountains which are well documented by Wyżga (1987). The most complete Silurian geological section is in the Żdanów outcrop (in German Herzogswalde) (Fig. 1B). It comprises lydites, siliceous and clayey shales with phosphate concretions, and tuff interbeds, ranging in age from the early Llandovery *Parakidograptus acuminatus* Biozone to

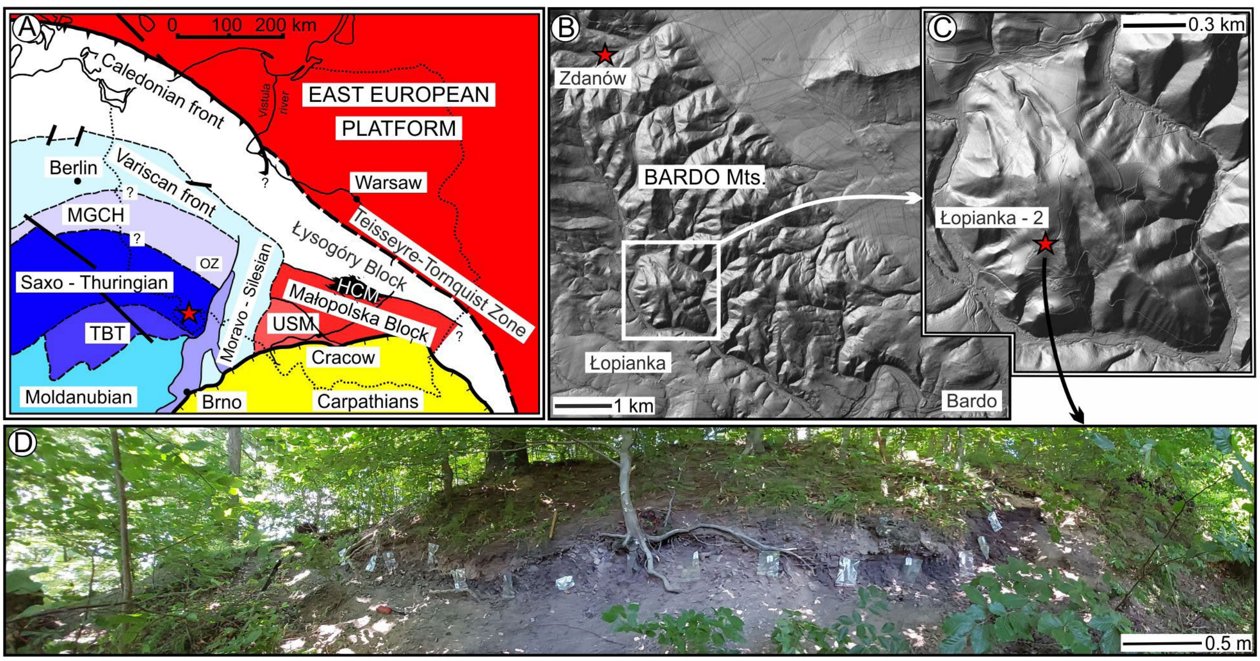

Fig. 1. A, simplified structural map of central Europe (Bełka *et al.* 2002) with the Bardo Mountains location. Abbreviations: HCM – The Holy Cross Mountains; MGCH – Mid German Crystalline High; OZ – Odra Zone; TBT – Tepla-Barrandian Terrane; USM – Upper Silesian Massif. B, LiDAR map of the fragment of the Bardo Mountains with the Zdanów outcrop and Łopianka mountain location. C, location of the Łopianka–2 outcrop (B,C from www.geoportal.gov.pl). D, general view of Silurian rocks in the Łopianka–2 outcrop.

the latest Pridoli *Istrograptus transgrediens* Biozone (Malinowska 1955a, Porębska & Koszowska 2001; Radzevičius *et al.* 2020). The total thickness of Silurian strata is about 50 m in Żdanów section. It is subdivided into Lower Graptolitic Shales, Green Shales, and the Upper Graptolitic Shales (Porębska 1980), or only the Graptolite Shales (Nowak, 2019). These deposits indicate a pelagic environment and developed on the floor of the Saxo-Thuringian Ocean (Franke *et al.*, 2017) or the Bardo Ocean (Racki *et al.* 2022).

The Silurian deposits form Łopianka mountain (in German Pinkeberg) have been studied for a century (Dahlgrün & Finckh 1924). There are two outcrops on Łopianka mountain (Wyżga 1987) called 'the first' and 'the second'. Both outcrops are characterized by the Lower Graptolitic Shales. The Silurian sequence is more complete in Łopianka–1 and commences with Ordovician sandstone that is overlapping Telychian and lower Wenlock shales. According to Wyżga (1987), Wenlock shales uncover in the Łopianka–2 section. There are few studies on geochemistry (Malinowska 1955b; Bauersachs *et al.* 2009) or sedimentation (Wyżga 1987; Kremer 2011) from the Łopianka outcrops. A few graptolite studies recorded *Spirograptus turriculatus*, *Streptograptus crispus*, *Monoclimacis griestoniensis*, *Oktavites spiralis* and *Cyrtograptus murchisoni* biozones in the Łopianka–1 outcrop (Malinowska 1955a, Wyżga 1987) and *lundgreni – praedeubeli* biozones in the Łopianka–2 outcrop (Porębska 1998). Thus, the Silurian deposits are known from Łopianka but detailed bio– and chemostratigraphical investigations have not yet been completed.

Material and methods

New material for geochemical, and palaeontological investigations comes from the Łopianka – 2 outcrop (N: 50°31'14.40' E: 16°40'17.76') which is located on Łopianka mountain (Fig. 1C) in the Central part of the Sudetes (SW Poland). Pelagic black, grey, greyish and greenish argillitic shales about 7 m thick (Fig. 2) of the Lower Graptolitic Shales (Wyżga 1987) are exposed there (Fig. 3). Bed layering is almost vertical. Samples for palaeontological, and $\delta^{13}C_{org}$ analyses were collected approximately every 0.1 m, about 0.5 kg each. It was not possible to collect samples from the *lundgreni – praedeubeli* interval, as this part of the outcrop is now collapsed and overgrown.

Standard methods were used for the $\delta^{13}C_{org}$ analysis (Radzevičius et al 2019). Approximately 1 g of each sample was grinded to powder, powder was dissolved using HCl (5 N) for 24 hours to remove carbonate material, and powder residue was washed with distilled water and dried. $\delta^{15}N$ and $\delta^{13}C_{org}$ measurements were taken via EA-IRMS, Flash EA1112–Thermo V Advantage technique (Garbaras *et al.* 2008) at the Center for Physical Sciences and Technology in Vilnius (Lithuania).

Results

Biostratigraphy

Graptolites are not abundant or poorly preserved in the studied samples (Fig. 2) and a sequence of graptolites biozones was therefore not possible to determine. However, some rare graptolites have provided very important biostratigraphical information.

The lowest samples yielded high diversity graptolite assemblages. Seven species, *Retiolites geinitzianus* Barrande (Fig. 4 B,I), *Barrandeograptus* cf. *pulchellus* (Tullberg) (Fig. 4 $C_{1,2}$), *Monograptus pseudocultellus* Bouček (Fig. 4 F,G), *Monograptus priodon* (Bronn) (Fig. 4 J), *Pristiograptus praedubius* (Bouček), *P. largus* (Perner) (Fig. 4 H), and *Monoclimacis vomerina* (Nicholson) (Fig. 4 A), are recognized there. The stratigraphically long-ranging *R. geinitzianus, M. priodon, Mc. vomerina*, and *P. praedubius* are known from the uppermost Telychian to lowermost Sheinwoodian in peri-Gondwana (e.g. Loydell *et al.* 2009), Bohemia (e.g. Štorch 2023) and Baltica (e.g. Paškevičius 1997) and link the upper Llandovery to the lower Wenlock (*spiralis – murchisoni* biozones). The easily recognizable species *M. pseudocultellus* is known from the *insectus* Biozone (Suyarkova 2012) and the *murchisoni* Biozone (Loydell et al 2017) in Baltica as well as in the *insectus – murchisoni* interval in Bohemia (Štorch 1994). There is relatively high diversity of graptolites in the lowest sampling level but no graptolite species which are informative for high resolution biostratigraphy.

The lowest occurrences of *Cyrtograptus* cf. *centrifugus* Bouček (Fig. 4D) at 0.3 m could mark the *centrifugus* Biozone in Łopianka-2 outcrop (Fig. 2). However, *Cyrt.* cf. *centrifugus* (Fig. 4D) and *Cyrt. bohemicus* Bouček (Fig. 5L) are found together at 0.35 m level and mark the *murchisoni* Biozone of the lowermost Wenlock. *Cyrtograptus centrifugus* can range in the lower part of the *murchisoni* Biozone with *Cyrt. murchisoni* (Carruthers) (Loydell *et al.* 2003) and *Cyrt. bohemicus* (Štorch 2023). The Llandovery-Wenlock boundary is therefore approximately at the level

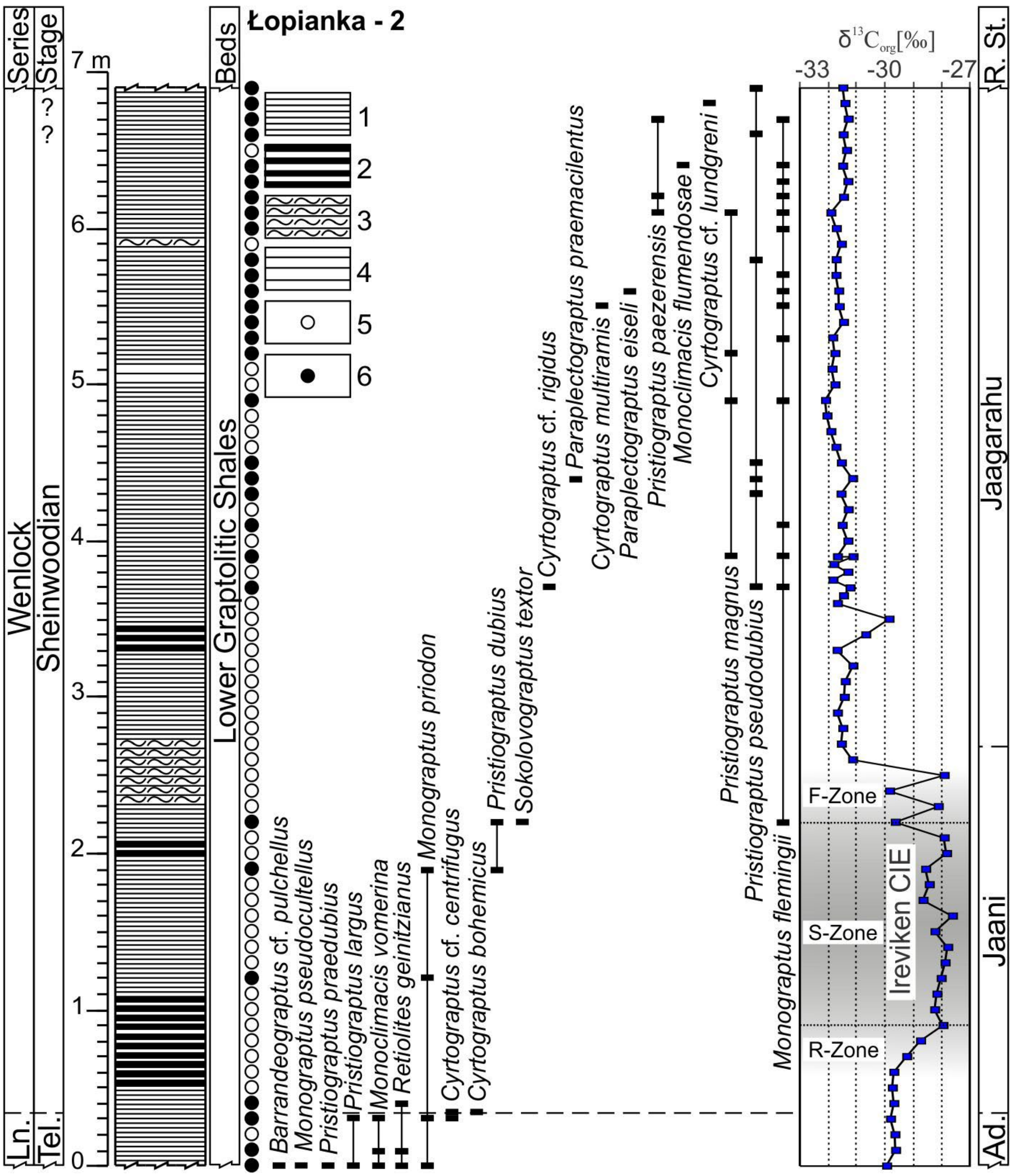

Fig. 2. Stratigraphical framework, sampling levels, distribution of graptolites and carbon isotope ($\delta^{13}C_{org}$) stratigraphy of the Łopianka–2 outcrop and correlation with Regional stages (after Paškevičius *et al.* 1994) of the Baltic Silurian Basin. Abbreviations: Ad. – Adavere; Ln. – Llandovery; R. St. – Regional Stage; Tel. – Telychian. Legend: 1 – black shales; 2 – lydites; 3 – greyish shales; 4 – greenish shales; 5 – samples without graptolites; 6 – samples with graptolites.

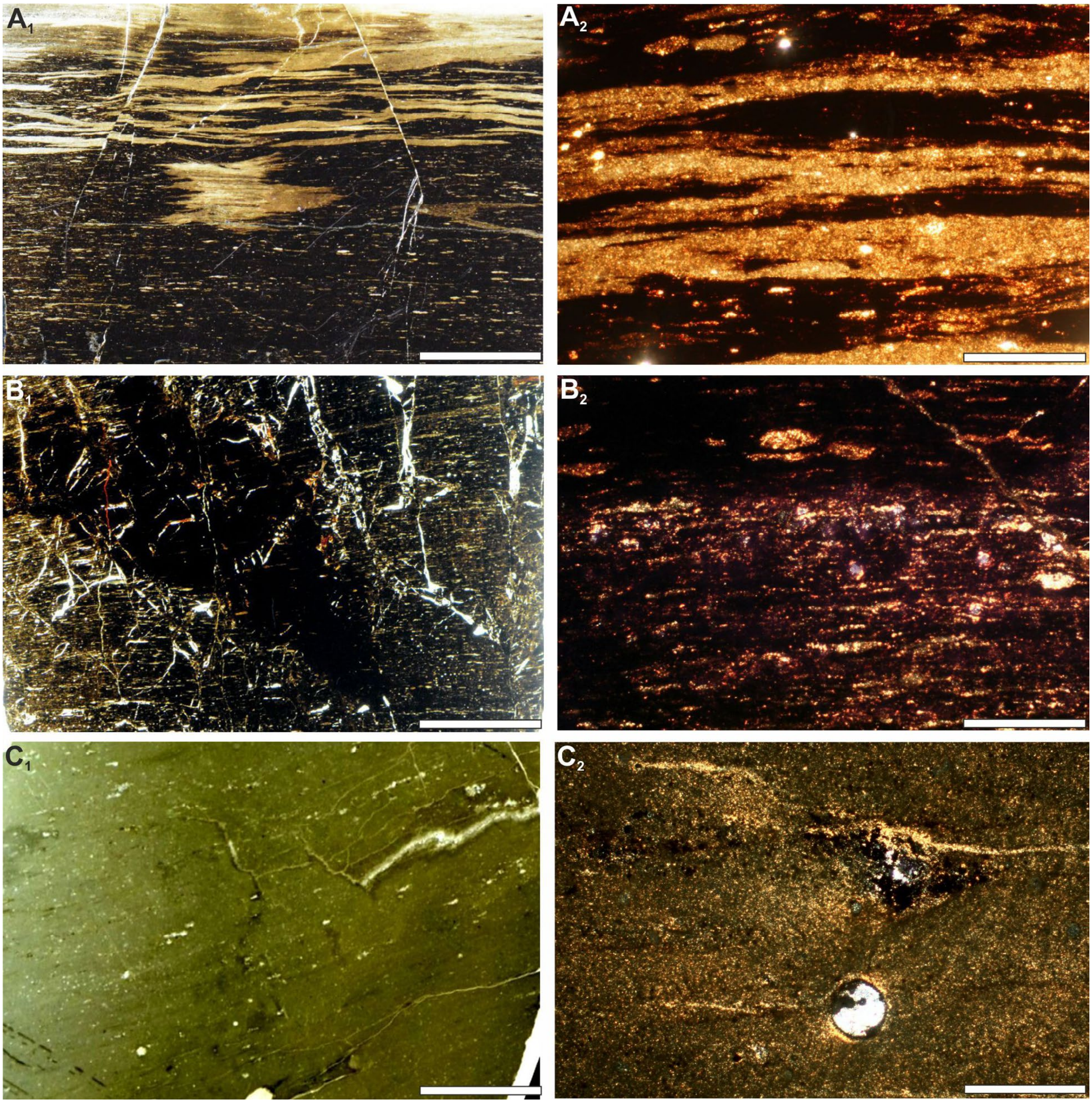

Fig. 3. Thin sections of various shales from the Łopianka–2 outcrop. A, the contact zone between green and black shale (A_1), laminated black shale (A_2), depth 5.9 m. B, finely laminated black shale (B_1), rich in organic matter with quartz veins (B_2), depth 6.9 m. C, indistinctly laminated green shale (C_1) with radiolarians or spherical structure filled with quartz (C_2), depth 2.3 m. Scale bar: A_1, B_1, C_1 – 1 cm; A_2, B_2, C_2 – 0.5 mm.

with these both *Cyrtograptus* species in Łopianka–2 outcrop.

The next level with age-diagnostic graptolites for biostratigraphy lies at 3.7 m (Fig. 2). Here *Cyrt.* cf. *rigidus* Tullberg (Fig. 5B) indicates the *rigidus* biozone. The graptolite assemblage *Cyrt. multiramis* Törnquist (Fig. 5H) (Štorch 2023) and *Pristiograptus magnus* (Fig. 5E) (Loydell et al 2010; Urbanek et al. 2012) species indicate the uppermost Sheinwoodian (*perneri* Biozone) or lowermost Homerian (*lundgreni* Biozone) in the Łopianka-2 outcrop. Long range retiolitid species *Paraplectograptus eiseli* (Manck) are found in this interval (Fig. 5C). This species range from the *riccartonensis* Biozone up to the *lundgreni* Biozone

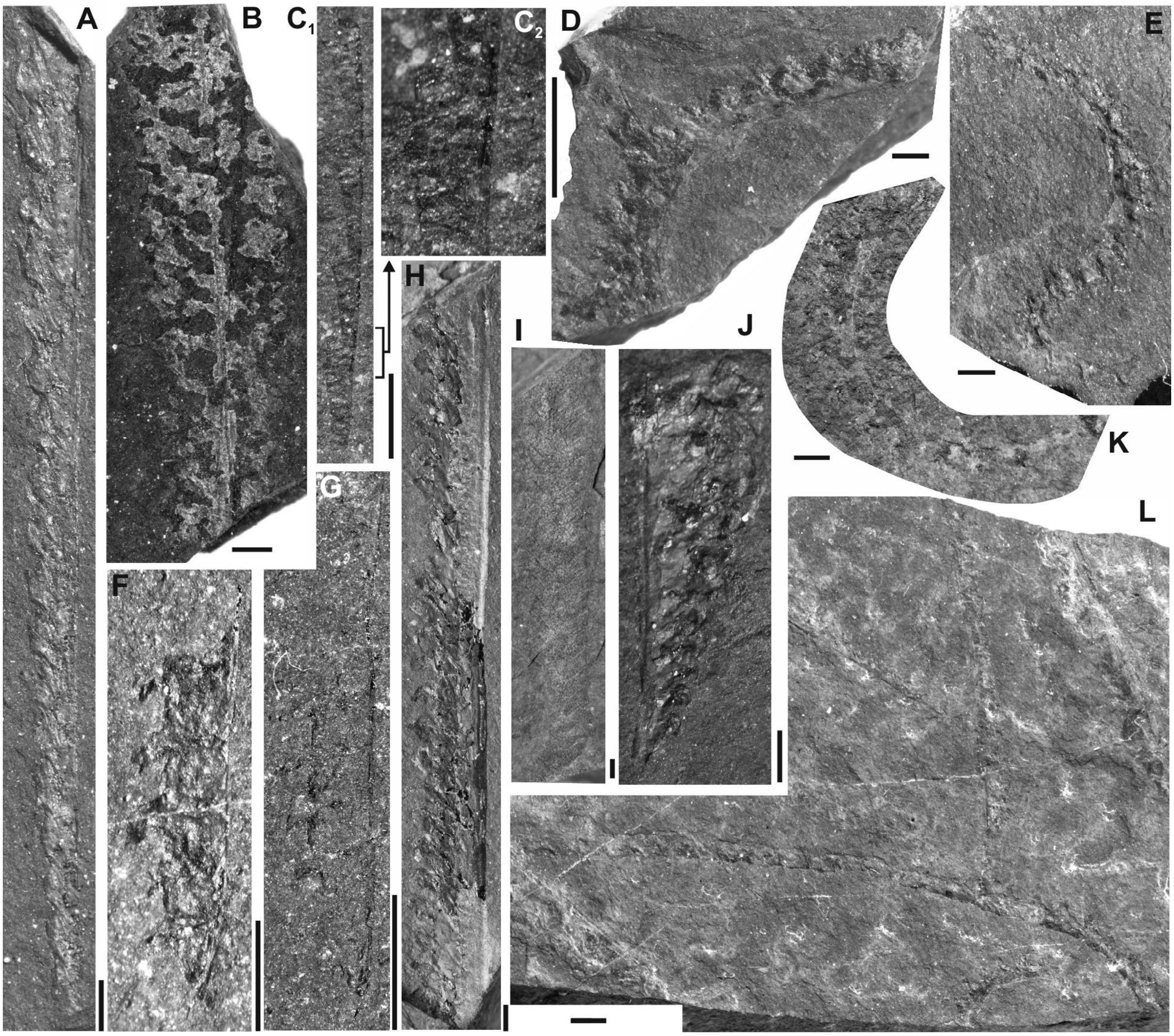

Fig. 4. Graptolites from Łopianka–2 outcrop. A, *Monoclimacis vomerina* (Nicholson), no. LOP-2-189, (level 0 m). B, I, *Retiolites geinitzianus* Barrande. B, no. LOP-2-186, (level 0 m). I, no. LOP-2-175 (level 0.1). $C_{1,2}$, *Barrandeograptus* cf. *pulchellus* (Tullberg), no. LOP-2-194a, (level 0 m). D, E, *Cyrtograptus* cf. *centrifugus* Bouček, no. LOP-2-167, (level 0.3 m). E, no. LOP-2-196 (level 0.35 m). F, G, *Monograptus pseudocultellus* Bouček, (level 0 m). F, no. LOP-2-190, G – no. LOP-2-191. H, *Pristiograptus largus* (Perner), no. LOP-2-194, (level 0 m). J, *Monograptus priodon* (Bron), no. LOP-2-168, (level 0.3 m). K, L, *Cyrtograptus bohemicus* Bouček. K, no. LOP-2-203 (level 0.35 m). L, no. LOP-2-200, (level 0.35 m). Scale bar 1 mm.

(Maletz 2024). Stratigraphically non-diagnostic taxa range in the topmost part of the investigated interval and demonstrate an upper Sheinwoodian or lower Homerian age (*lundgreni* Biozone) (Fig. 2). There is also a graptolite identified as *Cyrt.* cf. *lundgreni* Tullberg (Fig. 5I) at the 6.8 m level which may refer to the *lundgreni* Biozone (lower Homerian).

Organic matter carbon isotopes

According to the $\delta^{13}C_{org}$ data, the investigated interval of the łopianka–2 outcrop could be subdivided into three intervals. The first or lower interval comprising 8 samples (0–0.7 m) is marked by $\delta^{13}C_{org}$ values that vary with small fluctuations from −29.24 to −29.95 ‰ (Fig. 2). The $\delta^{13}C_{org}$ values gradually increase to −27.96 ‰ and vary by around −28 ‰ in the 0.9–2.5 m interval; it is this interval which has the highest $\delta^{13}C_{org}$ values. The $\delta^{13}C_{org}$ again falls to −31.15 ‰ at 2.6 m of the measured section and, with some fluctuations, varies between −32–−31 ‰ in the 2.6–6.9 m interval. This is the studied interval with the lowest $\delta^{13}C_{org}$ values, punctuated by one small positive $\delta^{13}C_{org}$ excursion at a depth of 3.5 m. There, the $\delta^{13}C_{org}$ rises to −29.86 ‰ (Fig. 2). Accordingly,

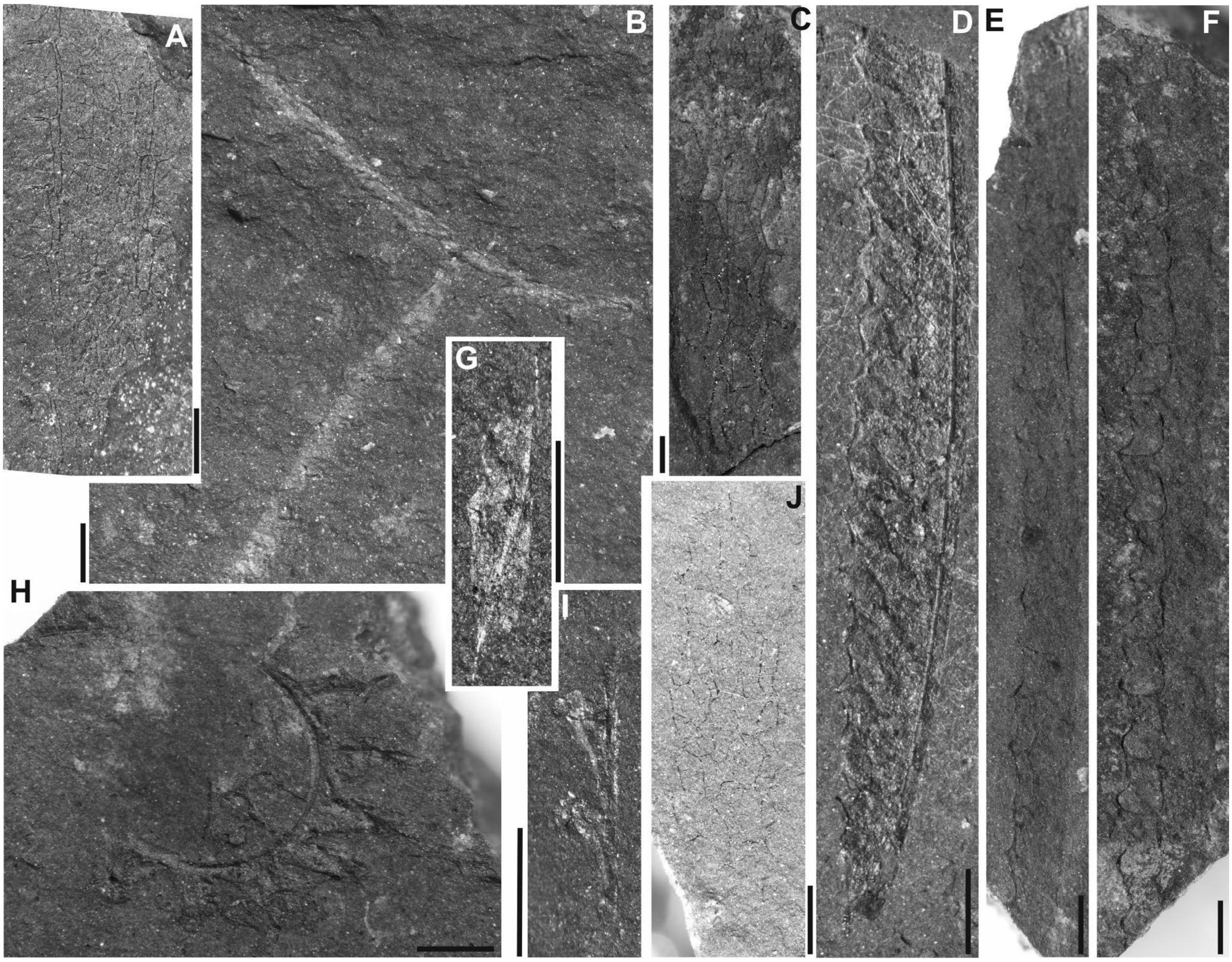

Fig. 5. Graptolites from Łopianka–2 outcrop. A, *Sokolovograptus textor* (Bouček & Münch), no. LOP-2-29, (level 2.2 m). B, *Cyrtograptus* cf. *rigidus* Tullberg, no. LOP-2-145, (level 3.7 m). C, *Paraplectograptus eiseli* (Manck), no. LOP-2-77, (level 5.6 m). D, F, G, *Pristiograptus pseudodubius* (Bouček). D, no. LOP-2-157, (level 4.3 m). F, LOP-2-68, (level 5.4 m). G, no. LOP-2-156, (level 4.3 m). E, *Pristiograptus magnus* Urbanek, Radzevičius, Teller, Kozłowska, no. LOP-2-115, (level 5.2 m). H, *Cyrtograptus multiramis* Törnquist, no. LOP-2-64, (level 5.5 m). I, *Cyrtograptus* cf. *lundgreni* Tullberg, no. LOP-2-51, (level 6.8 m). J, *Paraplectograptus praemacilentus* (Bouček & Münch), no. LOP-2-134, (level 4.4 m). Scale bar 1 mm.

$\delta^{13}C_{org}$ values vary between −32.11 and −27.62 ‰ in the studied interval (approximately 4.5 ‰).

Discussion

Integrated evidence from graptolites and stable carbon isotopes indicate the presence of the early Sheinwoodian or Ireviken $\delta^{13}C_{org}$ excursion in the Łopianka–2 outcrop. Three distinct chemostratigraphical zones can be established in the section (Fig. 2). The rising zone (R-Zone) of the Ireviken positive $\delta^{13}C_{org}$ isotope excursion is rather thin, starts 0.3 m above the first occurrence of *Cyrt. bohemicus* and does not coincide with the base of the *murchisoni* Biozone. The discrepancy of the beginning of $\delta^{13}C_{org}$ excursion and the base of the *murchisoni* Biozone is also documented in Gotland (Hartke et al 2021) and in the Banwy River section, Wales (Loydell and Frýda 2007). However, the lowermost interval, below R-Zone can be correlated with the upper part of Adavere Regional Stage in the East Baltic (Fig. 2). The subsequent zone of stable isotope values (S-Zone) is defined as a long-lasting steady interval (about 1.3 m) of high $\delta^{13}C_{org}$ values (Fig. 2) with only rare graptolite occurrences. The long stratigraphical ranges of the graptolites identified in the Łopianka–2 section do not allow for distinguishing biozones and, in particular, biozonal boundaries. The falling zone (F-Zone) is defined by a rapid decrease of $\delta^{13}C_{org}$ values. The thickness of the F-Zone is about 0.4 m. There are no graptolite occurrences in the F-Zone interval. All intervals of the positive $\delta^{13}C_{org}$ excursion could be correlated with the Jaani Regional Stage (Fig.2) of Lithuania (Želvys et al 2022). Above the F-Zone $\delta^{13}C_{org}$ values are relatively stable and are lower than those in the interval of the isotope excursion (Fig. 2). This interval could be correlated with the Jaagarahu Regional Stage in the East Baltic.

Conclusions

The integrated stratigraphical analysis of the Łopianka–2 outcrop section revealed that the studied interval corresponds to the uppermost Telychian and Sheinwoodian. Graptolites are rare but, according to $\delta^{13}C_{org}$ data, the Ireviken positive excursion is well recorded in the studied section. In the Łopianka – 2 outcrop the Ireviken positive $\delta^{13}C_{org}$ isotope excursion starts 0.3 m above the first occurrence of *Cyrt. bohemicus* and does not coincide with the base of the *murchisoni* biozone. The last but peculiar characteristics of the $\delta^{13}C_{org}$ record is that it displays higher values before the Ireviken positive carbon isotope excursion then after it.

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