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Maturation patterns in Palaeozoic rocks of north-eastern Bulgaria based on conodont colour alteration index (CAI) data

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Abstract. CAI data have been used to estimate the thermal history of Palaeozoic sedimentary rocks in five boreholes in north-eastern Bulgaria. Generally, the highest CAI values (4.5) have been found in Lower Devonian sediments, and the lowest values in the Lower Carboniferous (1.5), which suggests pre-tectonic coalification. The Palaeozoic rocks attained the observed thermal maturity during Late Carboniferous times. The post-Carboniferous heating should have had no influence on coalification of Palaeozoic sediments. It seems likely that the palaeogeothermal gradient was higher than the present one of ~ 30 °C/km. The economic potential for hydrocarbons in the Devonian sediments of north-eastern Bulgaria is questionable, as most CAI values suggest burial temperatures of about 110–200 °C and 190–300 °C (CAI 3 and 4, respectively) resulting in the supramature state of the organic matter. The occurrence of hydrocarbons might be possible in some of the Carboniferous sediments, based on their estimated palaeotemperatures between 50 °C and 90 °C.

Key words: conodont maturation, Palaeozoic, Middle Devonian, Upper Devonian, stratigraphy

Introduction

Studies of conodont colour and the textural alteration of conodont elements can provide important information on the thermal maturation of the sedimentary rocks in which they occur. Thermal maturation is an important property for hydrocarbon and mineral exploration. Colour alteration of conodonts was first noted by Ellison (1944), but the colour differences (CAIs) were placed in a systematic context by Epstein et al. (1977) and Rejebian et al. (1987). The change of colour is the result of trace amounts of organic matter sealed within conodont elements. The colour changes are progressive and irreversible, and can be assigned to different temperature regimes during metamorphism. Epstein et al. (1977) and Rejebian et al. (1987) used an Arrhenius plot to extrapolate their experimental data to geologic time scales.

During recent decades, many regional CAI studies in Europe have been used to assess the organic maturation and hydrocarbon potential in diagenetic to very low grade metamorphic regimes (e.g., Helsen and Königshof 1994, Garcia-Lopez et al. 1997, Sarmiento et al. 1999, Boncheva et al. 2002, Wiederer et al. 2002). Palaeotemperature data provided by CAI studies are useful in elucidating the thermal history of sedimentary basins, and in determining thermal aureoles related to igneous intrusions (e.g., Kovacs and Arkai 1987, Burnett 1988, Königshof 1991). Textural alteration of conodonts can provide information regarding the hydrothermal fluids and tectonic stresses that the conodonts have encountered (e.g., Königshof 2003 cum lit.). This study presents maturity data from Palaeozoic subsurface sediments of north-eastern Bulgaria, using conodont colour alteration indices to assess the possibility that these sediments may have acted as source rocks for the hydrocarbons now mainly found in Mesozoic sedimentary rocks. Furthermore, studies of the textural alteration of conodonts have also been undertaken to elucidate the nature of the metamorphism that affected the Palaeozoic sedimentary rocks.

Materials and methods

More than 200 samples have been collected from five subsurface localities (drill cores) in north-eastern Bulgaria (Boncheva and Königshof 2004). These localities are, from west to east, the Gornyak, Ograzhden, Kardam, Belgun, and Vaklino boreholes (Fig. 1). Most of the samples are derived from drill cores, while others are from drill cuttings. Most conodont samples investigated in this study were obtained from limestones, though some are from dolostones and calcareous siltstones of Late Devonian to Early Carboniferous age. The samples usually yield sparse conodont faunas (some up to 50 specimens/kg), but enough to determine their grade of metamorphism and age. Additional fossil groups have been considered with respect to stratigraphy. Samples were always processed in the same manner, using 7.5 % formic acid during a maximum of approximately 40 hours of maceration. Conodont elements were hand-picked directly from the insoluble residue, and the CAI was determined by comparison with a CAI standard set produced in the laboratory and from naturally altered samples. Textures and surfaces of conodonts were examined by optical microscopy and, in gold-coated conodonts and/or conodont fragments, with scanning electron microscopy (SEM) at the Forschungsinstitut Senckenberg in Frankfurt a. Main. Conodont material described here is housed at the collections of the Bulgarian Academy of Sciences, Sofia.

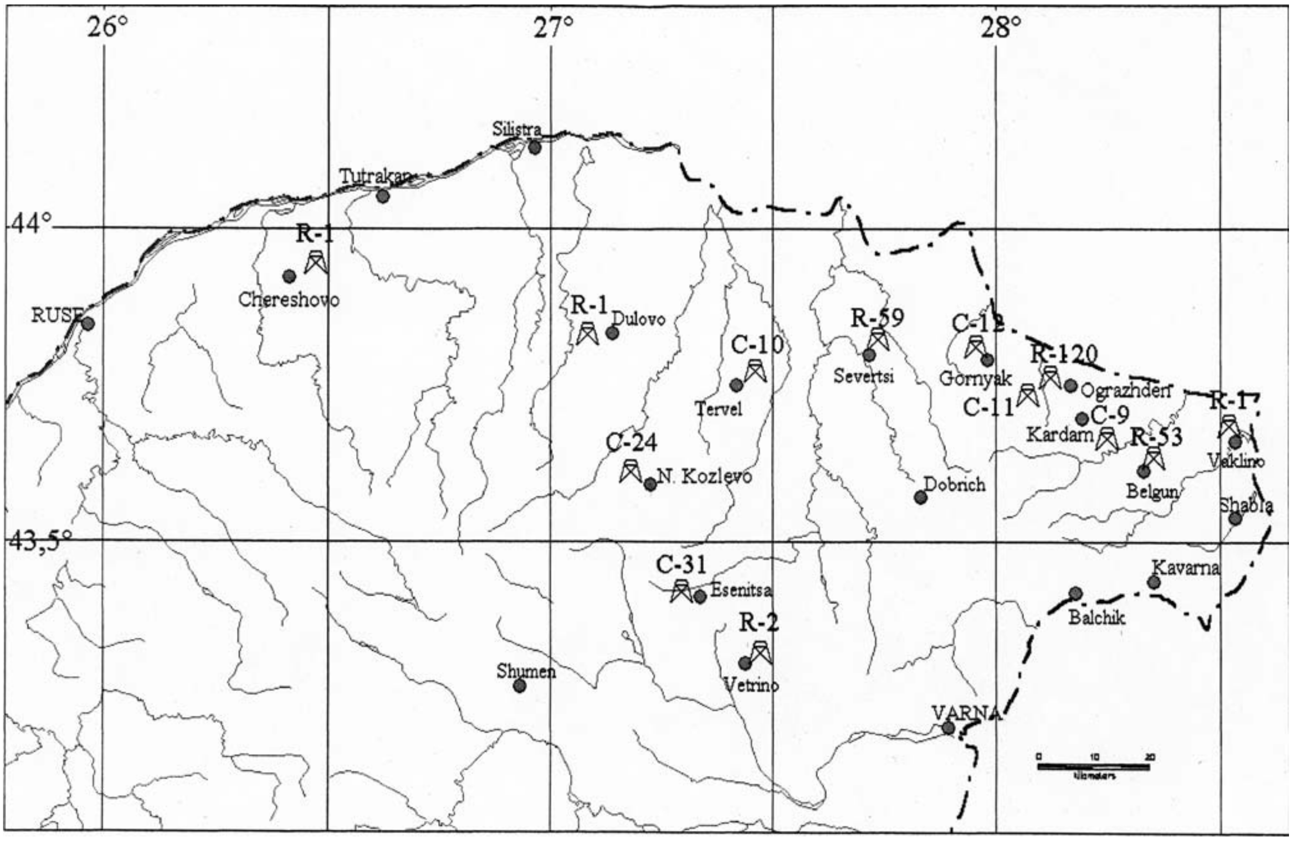

Figure 1. Location of the investigated boreholes Gornyak, Ograzhden, Kardam, Belgun, and Vaklino in the eastern part of the Moesian Terrane.

Geological setting

The main Palaeozoic sequences occur in the south-eastern and south-western part of Bulgaria, while no Palaeozoic sequences exist at the surface in the north-eastern part. Therefore, the results of this report are based on drill cores and drill cuttings. The investigated area in which the boreholes are located lies between Romania to the north, the Black Sea to the east, and the east-west striking mountain chain of Stara Planina (or Balkans) to the south.

The following structural units can be distinguished from north to south (Fig. 2): the Moesian Platform, the Balkan Zone, the Srednogorie Zone, and the Morova-Rhodopian Zone (Bonchev 1979, Vuchev et al. 1994, Georgiev et al. 2001).

Northern Bulgaria is considered to be a continental microplate (Moesian microplate, after Tzankov 1993) that was separated from the northern passive margin of Perigondwana in the Early Palaeozoic (Yanev and Tzankov 1997). The Protomoesian microcontinent (Fig. 3) is considered to be comprised of the Moesian Terrane and the Balkan Terrane in the south, which are joined by the South European Variscan Suture (Haydoutov and Yanev 1997, Yanev 2000, Georgiev et al. 2001). The collision of these terrains took place during Late Carboniferous and Permian time. The distinction of these two terrains is based on differences in thickness, lithology, and facies, especially in Devonian and Carboniferous sequences. For example, no evidence of Stephanian sediments exists in the Moesian Terrane in contrast to the Balkan Terrane to the south (Haydoutov and Yanev 1997). Although the Moesian Terrain and the Balkan Terrain were both part of Gondwana, their different lithologies indicate origins in different parts of Gondwana and Perigondwana as described in detail by Yanev (2000).

The Moesian Platform is considered as a morphotectonic subdivision (Bonchev 1982) with a Palaeozoic basement covered by folded Mesozoic sediments. The investigated area belongs to the eastern part of the Moesian Platform (Fig. 1), in which are exposed 1 to 2 km thick layers of rocks of Cretaceous, Palaeogene, and Neogene age, while subsurface rocks are of Silurian, Devonian, Carboniferous, Permian, Triassic, and Jurassic age (Tari et al. 1997). Pre-Carboniferous Palaeozoic successions are anchi- to epimetamorphic reversed during the Early Carboniferous (Yanev 1996). However, rocks younger than Devonian show no sign of metamorphism and are relatively undeformed. According to Sengor (1984), the main Permian-Lower Cretaceous structures are connected with the Cimmerian Orogeny.

The Moesian Platform (Fig. 2), forming the foreland of the Alpine thrust belt, is composed of up to 4–5 km thick, relatively undeformed, dominantly shallow marine Mesozoic sediments resting on a gently folded Palaeozoic basement, which is traditionally subdivided into Wallachian and Dobrudjan blocks, and the Intra-Moesian fault (Visarion et al. 1988). The western side of this fault zone is made up of epi- and mesometamorphic Precambrian rocks,

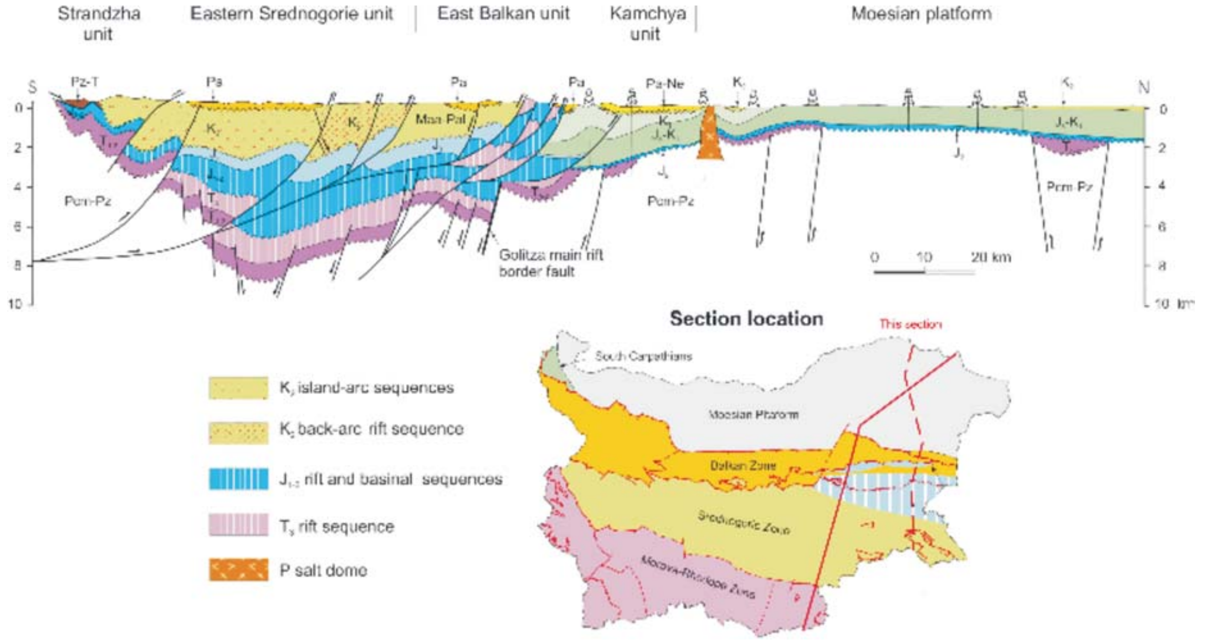

Figure 2. Morphotectonic and palaeotectonic subdivision of Bulgaria. Interpretation based on seismic profiles, well and surface data (slightly modified after Georgiev et al. 2001).

whereas the eastern unit is characterized by weakly metamorphosed, Upper Vendian-Lower Cambrian flyschoid deposits (greenschists), unconformably overlying a middle Proterozoic metamorphic succession. The sediments of the Moesian Platform that overlie the crystalline basement can be subdivided into the following lithologic units ("megasequences"):

1. Upper Cambrian to Lower Devonian sediments representing the "lower siliciclastic sequence". The basal part contains arkose sandstones and quartzitic sandstones with intercalations of siltstones and shales. This sequence is unconformably overlain by Silurian graptolite shales with an average thickness of 800 m. The Lower Devonian is represented by quartzitic sandstones and siltstones, some rare limestones and conglomerates: the average thickness of these rocks is about 370 m.
2. Middle Devonian and Lower Carboniferous sediments are predominantly composed of massive limestones and dolomites with intercalated bituminous limestones and evaporites, reaching a thickness of up to 3500 m in the north-eastern part of the Moesian Platform (Balkan Terrane; Haydoutov and Yanev 1997, Yanev 2000).
3. The Upper Carboniferous part is represented by coal-bearing sediments, overlain by siltstones, marls, and sandstones. This succession shows a thickness of 700–800 m which is well documented in several wells (Vinogradov 1988). The base of the Permian and Triassic sequences are characterized by two unconformities,

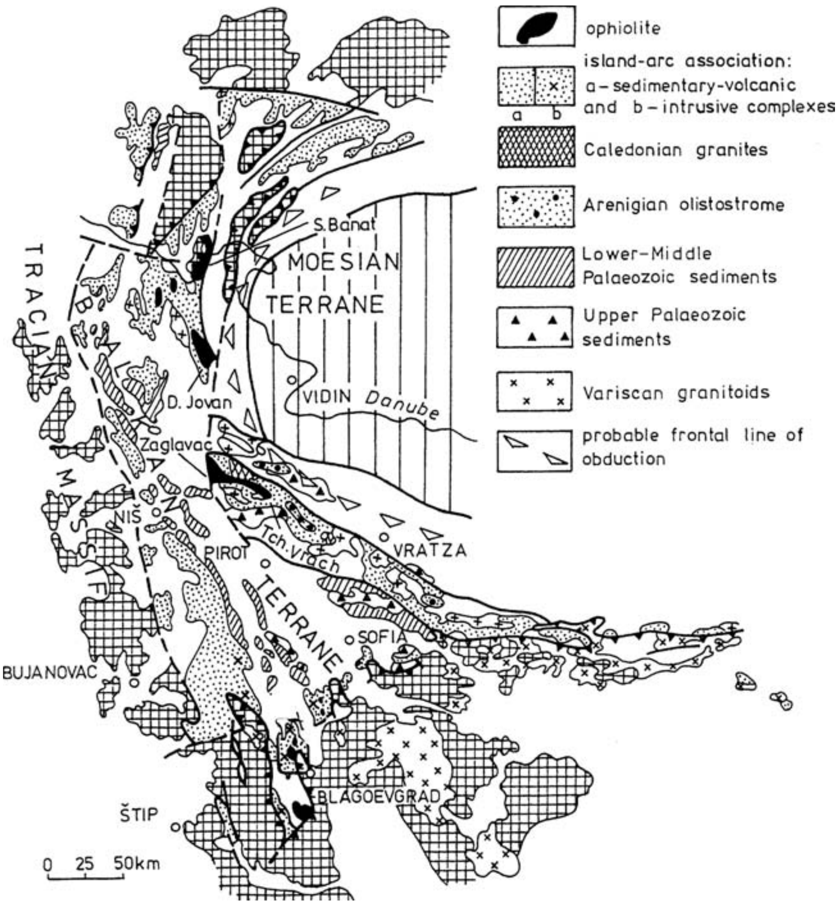

Figure 3. Geological sketch of the Protomoesian microcontinent, western part (slightly modified after Haydoutov and Yanev 1997).

but there is little evidence for significant deformation or deep erosion. Permian sediments are present in some depressions, and are composed of red continental sediments in the lower part and evaporite- and halite-bearing deposits in the upper part (Yanev 1993).

According to Yanev (2000) the Moesian and the Balkan Terrane underwent a northward migration from the southern, moderately humid zone during the Ordovician, to a northern arid zone in the Permian. The collision of the Moesian and the Balkan Terrain took place in the Early Carboniferous, and continued during the Late Carboniferous and Early Permian (e.g., Janev 1988). Due to these tectonic events the sedimentological record is different from area to area (Tables 1–5), indicating uplift and erosion as well as faulting and thrusting.

Major unconformities in the Mesozoic and Cenozoic sediments occur at the base of the Triassic, Jurassic, Upper Cretaceous, and Eocene, and are related to major compressional tectonics due to the Alpine orogeny. Recent plate tectonic concepts suggest that the Alpine orogen of Bulgaria includes a mosaic of lithospheric fragments of local, proximal, and exotic origin that accreted onto Laurasia during the closure of the Tethys (Robertson and Dixon 1984, Sengor et al. 1984, 1988, Ziegler 1990, Dercourt et al. 1993, Ricou 1994, Stampfli et al. 1998, 2001), which has been interpreted as the second accretion after that between the Perigondwanan fragments (e.g., Moesian Terrane) and Paleo-Europe, which took place in the Late Devonian–Early Carboniferous (Yanev 1996, 2000).

Hydrocarbon exploration of the Moesian Platform

The Fore Carpathian-Balkan oil and gas basin corresponds to a wide depression confined between the mountain fold belts of the East and South Carpathians in the north, Banat in the west, Stara Planina in the south, and the Dobrogea massif in the east. This basin covers an area of approximately 90 thousand km^2. North of the Danube it is situated within Romania and also in northern Bulgaria, while its western part extends as a narrow strip into Yugoslavia. The hydrocarbon fields of the Moesian Platform mainly occur in the western and northern part belonging to Romania. Commercial hydrocarbon accumulations were encountered in Cenozoic, Mesozoic, and Palaeozoic sediments of various lithologies, and the distribution of oil and gas field are very irregular (Monahov et al. 1981, Vuchev et al. 1994, Georgiev and Dabovski 1997). Hydrocarbon accumulations in Palaeozoic rocks have been found in the northern part of the Moesian Platform (the Devonian Bibeshti-Bulbuceni field in Romania), whereas the most productive fields are located in the central part of the Moesian platform west of Bucharest, within Mesozoic sediments. Reservoirs have been found also in eastern parts of the Moesian Platform (Georgiev and Atanasov 1993). Most of the fields are oil-bearing. More than two-thirds of the oil and gas/condensate pools have been found in Mesozoic reservoirs. The hydrocarbon potential of the Palaeozoic sediments is much debated. To date, only a few accumulations have been discovered in Palaeozoic sedimentary rocks, and the clayey marls and shales of Silurian and Devonian age may be considered as oil-source rocks (Yanev and Tzankov 1997). According to Paraschiv (1984), hydrocarbons of the Moesian Platform have been formed in several stages since the Palaeozoic. These authors maintain that the hydrocarbons were frequently remobilized and redistributed into younger sediments. However, it is not unlikely that Palaeozoic rocks could have produced significant amounts of hydrocarbon in areas characterized by strong Mesozoic subsidence, e.g. in the Cracow-Silesia region (Belka 1993). On the other hand, only one oil pool has been encountered in the northern part of the Moesian Platform (Bibeshti-Bulbuceni field, Monahov et al. 1981). Based on CAI data, the economic potential of hydrocarbons, especially oil, in Devonian sediments in north-eastern Bulgaria, is questionable. Most CAI values suggest burial temperatures of about 110–200 °C and 190–300 °C (CAI 3 and 4, respectively), resulting in the supramature state for organic matter (Rejebian et al. 1987). Furthermore, most of the productive oil and gas fields on the Bulgarian part of the Moesian Platform are in Mesozoic sediments (Monahov et al. 1981). Hydrocarbons might occur in some Carboniferous sediments, based on the estimated palaeotemperatures of only 50–90 °C (CAI 1.5). Further investigation of the deformation structure and source rock potential is necessary in north-eastern Bulgaria for obtaining detailed information on the potential for hydrocarbon resources in these Palaeozoic sediments.

Results

CAIs and microstructures of conodont surfaces

The structure of conodont surfaces has been studied to evaluate the relationship between CAI data, tectonic deformation, and probable hydrothermal processes. Most of the studied conodont specimens are excellently preserved (see Plate I), and tectonic deformation and/or hydrothermal alteration are rare. Furthermore, most conodont samples of all grades of metamorphism show no signs of weathering. Microscopically visible brittle deformation and fracturing of the conodonts are also very scarce, and occur mostly in conodonts with CAI > 4.0 in samples derived from dolostones. This feature, and a sugary surface texture of the conodont elements, is present in dolostones and partly dolomitized limestones in samples from the Ograzhden (Plate I, fig. 1), Kardam, Belgun, and Vaklino boreholes, and has also been described from other regions (Königshof 2003). In some samples collected from the Gornyak borehole, conodonts show partly recrystallized grey surfaces that are quite uncommon throughout the sample suite. This feature may be attributed to diagenetic alteration as was pointed out by Königshof (1992, 2003). The influence of hydrothermal activity due to ascending solutions has not been observed in the samples.

CAI data derived from borehole samples

CAI determinations were carried out from more than 200 samples from five cored boreholes (Fig. 1). When possible, CAI values have been compared with other indices of inorganic metamorphism, such as calcite thermometry and scarce illite crystallinity data which have been published recently for an area in the north-eastern part of Bulgaria (Yanev and Stefanov 2002). Maximum temperature ranges based on observed CAIs are given in Tables 1–5. Additionally, these tables show the estimated overburdens required to produce the observed CAI values, assuming an average palaeo-geothermal gradient of approximately 3.5 °C/100 m. The columns of approximated maximum overburden are based on data from the literature. Colour alteration indices of Palaeozoic conodonts in the drill samples range in value from 1.5. to 4.5. Palaeozoic conodonts, mainly from Devonian strata, show CAI values from 3 to 4, whereas younger conodonts show lower values (CAI 1.5 for Lower Carboniferous conodont samples).

Gornyak borehole (Table 1)

The Gornyak borehole is situated in the northwest of the investigated area (Fig. 1). Conodont samples were derived from grey micritic limestones and oolitic limestones of Late Devonian age. In contrast to the investigated boreholes of the eastern part, the thickness of the Upper Devonian sediments in the Gornyak borehole reaches only a few meters, which is attributed to tectonic processes and erosion. The section between 854.5 m and 868 m is characterized by a core loss. Based on borehole surveying (seismic boring records), it can be assumed that Devonian sediments are overlain by Jurassic sediments with an angular unconformity. Alteration data show a CAI value of 3 for Famennian conodont elements. In comparison to the other borehole samples, the high CAI value in the Upper Devonian is unusual with respect to the estimated low sedimentation rate. This can be explained only by the assumption of thrusting and faulting during orogenesis, followed by strong erosion. Evidence of these processes is given by the surface structure of the conodont elements: in some samples, especially those from below the thrust fault, conodonts show sugary surfaces which are not related to contact metamorphism and/or hydrothermal activity, but may be due to alteration by percolating fluids and/or differential pressures connected with folding and thrusting.

Table 1. Gornyak borehole: CAI data, stratigraphy, and lithology

Depth (m)	Age	Conodont fauna and/or other fauna	CAI and estimated temperatures	Calculated maximum overburden (m)	Approximate maximum overburden (m) to produce observed CAI values (3.5 °C/100 m)	Textural alteration of conodonts	Lithology/tectonics
0–10	Quaternary						sands and clay
10–75	Neogene						sands and clay
75–132	Upper Cretaceous	ammonoides and echinoderms					limestones
132							erosional surface
132–846	Middle Jurassic to Early Cretaceous	ammonoides					clayey limestones, massive limestones
846–854.5	Early Jurassic						sandstones
854.5–868	Core loss						? at 855 m, Jurassic/Devonian boundary (erosional surface)
868	Devonian						dark grey micritic limestones
871.9–872	Early Famennian	*Hibbardella* sp., *Roundya* sp., *Bryanthodus* cf. *macrodentus*, *Apatognathis varians klapperi*, *Prioniodina prona*	CAI 3 (110 °C–200 °C)	~ 1900 m (~ 1000 m Lower Carboniferous)	3100–5700 m	conodonts show recrystallized surfaces	grey micritic limestones
872–873.7	Early Famennian	*Icriodus curvatus*, ?*Polygnathus* sp.	CAI 3 (110 °C–200 °C)	~ 1900 m	3100–5700 m	conodonts show recrystallized surfaces	
873.7–877 (boring depth at 877 m)							cuttings, no core

Ograzhden borehole (Table 2)
The Devonian sedimentary sequence of this borehole is represented by an alternation of shales and limestones in the lower part, and limestones with breccia and dolomites in the upper part. Middle Devonian strata are about 180 m thick, whereas the Upper Devonian (Frasnian) rocks have a thickness of 950 m. Famennian and Tournaisian sediments are absent due to erosion. The stratigraphy of the Devonian sediments is based on conodonts, whereas the Carboniferous sediments have been identified using the miospore assemblages (Boncheva et al. 1994). The Carboniferous starts with dark shales including distinct coal layers in the lower part, and sandstones and coal layers with root horizons in the upper part. This sequence is of Late Visean age and is about 510 m thick. Mesozoic and Cenozoic sediments with a thickness of 845 m overlie an erosional surface on the Carboniferous.

Conodonts from the lower Givetian to upper Eifelian show CAI values of 4.0. Lower CAI grades have been observed in Givetian and Frasnian conodont elements, which show a CAI of 3. Some of the Frasnian conodonts have a sugary surface (Plate I, fig. 1) due to diagentic alteration. In the Lower Carboniferous, no conodont data are available due to lithology (sandstones, coal layers). The calculated maximum overburden and the approximate maximum overburden to produce an assumed palaeogeothermal heat flow of 3.5 °C/100 m are in general accordance.

Kardam borehole (Table 3)
The Middle Devonian sediments are composed of calcareous shales, quartzites, and sandstones at the base, followed by massive and reef limestones with intercalated anhydrite layers and dolomites in the Eifelian and Givetian. The thickness of the Middle Devonian sequence reaches about 1780 m.

Table 2. Ograzhden borehole: CAI data, stratigraphy, and lithology

Depth (m)	Age	Conodont fauna and/or other fauna	CAI and estimated temperatures	Calculated maximum overburden (m)	Approximate maximum overburden (m) to produce observed CAI values (3.5 °C/100 m)	Lithology/tectonics
0–92	Cenozoic and Mesozoic sediments					sandstones, limestones, and marls
92						erosional surface
92–836	Early Cretaceous					clayey limestones, marls
836						erosional surface
836–845	Early Jurassic					micritic limestones, sandy limestones
845						erosional surface
845–853	Carboniferous					black shales and siltstones
853						erosional surface
853–984	Late Visean	Late Visean miospore assemblages (Boncheva et al. 1994)	Calcite data < 100 °C–< 300 °C			alternation of dark shales, sandstones and coal layers
984–1368	Late Visean	Visean miospore assemblages (984 m–1198 m), Boncheva et al. 1994				dark shales, at the base of this interval alternation of sandstones and coal layers
1368 m						erosional surface
1368–1415	Frasnian	*Polygnathus webbi*	CAI 3 (110 °C–200 °C)	~ 2800 m (~ 900 m Lower Carboniferous sediments and ~ 450 m Upper Devonian)	3100–5700 m	limestones, partly dolomitized
1415–1518						without core
1518–1621	Frasnian	*Polygnathus decorosus*, *Po. dubius*, *Po. xylus xylus*, ?*Po. webbi, Ozarkodina congesta*	Calcite data < 100 °C–< 300 °C Quartz data (150 °C–300 °C) CAI 3 (110 °C–200 °C)	~ 2800 m	3100–5700 m	dark limestones, partly dolomitized
1621–1673						without core

Table 2, continued

1673–1681	Frasnian					dark limestones
1681–1739						without core
1739–1742	Frasnian					dark limestones
1742–1800						without core
1800–1804						limestones with breccia layer and conglomerates
1804–2095		*Icriodus deformatus asymmetricus*	CAI 3 (110 °C–200 °C)			(section with much core loss) limestones and dolomites
2095–2317	Frasnian					dolomites and shales
2317–2326						
2319–2321	Early Frasnian	Trilobites, crinoids *Icriodus alternatus helmsi*	CAI 3 (110 °C–200 °C)	~ 3700 m	3100–5700 m	grey massive limestones
2321–2340						without core
2340–2391						black shales
2319–2394	Middle Devonian Givetian	*Icriodus subterminus*	CAI 3 (110 °C–200 °C)			alternation of shales with clayey limestones
2394–2398	Middle Devonian Early Givetian–Late Eifelian	*Icriodus norfordi, I. orri, I. obliquimarginatus, I. aff. amabilis, I. werneri*	CAI 3 (110 °C–200 °C) CAI 4 (190 °C–300 °C)	~ 3700 m ~ 3700 m	3100–5700 m 5400–8500 m	alternation of shales with clayey limestones
2398–2407						without core
2407–2417	Middle Devonian Early Givetian–Late Eifelian	*Icriodus amabilis, I. werneri, Polygnathus xylus xylus, Pand. expansa* (at 2412 m)	CAI 4 (190 °C–300 °C)	~ 3700 m	5400–8500 m	alternation of shales with clayey limestones
2417–2493						shales and clayey limestones and some intervals without core
2493–2500 (boring depth at 2500 m)						black shales

Within the Middle Devonian two major erosional surfaces occur. Above the Middle Devonian, from 924 to 945 m, fine-grained dolomites occur which probably belong to the Upper Devonian based on sedimentological and lithological criteria. No other Famennian and Carboniferous strata exist in this borehole section due to erosion. The Devonian is covered unconformably by Jurassic sediments. Conodont elements of the Eifelian at the base of the sequence show CAI values of 3, which agrees well with the calculated overburden and maximum overburden for producing the observed CAI values. The only exception concerns the interval between 1583 and 1585 m, but this may be explained by uplift processes in the Givetian. Evidence for this uplift is given by both sedimentological criteria and the surface textures of conodonts. The lowest CAI values occur at the top of the Middle Devonian, showing CAI values of 2. The sugary surface and fracturing of some conodont elements sampled in this interval may be due to diagenetic alteration. This feature has been described also in dolostones in the Eifel area, Rheinisches Schiefergebirge (Königshof 1992, 2003).

Belgun Borehole (Table 4)

This borehole reaches a depth of 3001 m, and the sediments show a dip between 15–20° (Spassov 1976). The succession between 2869–3001 m consists of limestones, dolomitic limestones, and interbedded siltstones and shales. Conodont samples from this interval indicate a Famennian age. There is core-loss between 2860–2869 m. The next younger level between 1186–2861 m belongs to the Upper Visean, which is represented by shales, siltstones, and rare intercalated limestones at the base of this sequence in which conodonts have been found. The interval between 2590–2860 m is composed of very similar sediments in comparison to the interval 1509–1747 m of the Vaklino borehole in the northeast. An erosional surface marks the boundary between Carboniferous sediments and the overlying Jurassic sediments. The CAI values of the conodont elements range from 3 in the lower part to 2 in the upper part of the Devonian sequence, whereas those of the Upper Visean show a CAI of 1.5. This last value is the lowest that has been found in this sequence, reflecting temperatures of

Table 3. Kardam borehole: CAI data, stratigraphy, and lithology

Depth (m)	Age	Conodont fauna and/or other fauna	CAI and estimated temperatures	Calculated maximum overburden (m)	Approximate maximum overburden (m) to produce observed CAI values (3.5 °C/100 m)	Textural alteration of conodonts	Lithology/tectonics
0–835	Jurassic and younger sediments						
924–945	?Frasnian						fine-grained dolomites, in some parts slightly calcareous
945–949	Middle Devonian (Givetian)	*Polygnathus xylus xylus, Pol. pseudofoliatus*	CAI 2 (60 °C–140 °C) Calcite data < 100°C–< 300°C	~ 1950 m (~ 1000 m for Upper Devonian and Lower Carboniferous)	1700–4000 m		
949.80–1037	Early Givetian	*Polygnathus pseudofoliatus, Pol. xylus ensensis,* *Pol. xylus xylus*	CAI 2 (2.5) (60 °C–140 °C)	~ 2000 m	1700–4000 m		detrital, massive limestones
1037–1043 1053–1423	Early Givetian	*Diplododella aurita, Pandorinellina insita, Synprioniodina gracilis*	CAI 2 (2.5) (60 °C–140 °C)	~ 2400 m	1700–4000 m		dolomite breccia and conglomerates with clayey-carbonate cements dark-grey massive, fine-grained dolomites in alternation with limestones and slightly dolomitized limestones
1423							erosional surface
1423–1430							clastic limestones and dolomites, partly cavernous (karstification?)
1583–1585							
1583.20	Middle Devonian (Givetian)	*Icriodus subterminus*	CAI (2.5) 3 (110 °C–200 °C) Calcite data < 100°C–< 300°C Quartz data ca 150 °C–300 °C	~ 2500 m	3100–5700 m	sugary surface	limestones and less dolomites in the lower part dark-grey micritic limestones with intraclastics and some thin anhydrite beds
1588–1937							dark-grey massive limestones with interbedded anhydrite layers; in the upper part organic rich limestones, massive dolomites/dolomitized limestones in the lower part brecciated limestones and dark-grey limestones (rich in organic material)
1937							erosional surface – discordans
1937–2323							carbonates, anhydrites and dolomites, less limestones
2323							erosional surface

Table 3, continued

2323–2524							detrital limestones and black shales, clayey limestones with calcareous shales
2324–2800							alternation of anhydrites and dolomites, less limestones
2800–3065 2951–2944 3059–3064	Middle Devonian (Eifelian)	*Icriodus corniger,* *I. subterminus,* *I. amabilis*	CAI 3 (110 °C–200 °C)	~ 3800–4000 m	3100–5700 m		fine-grained and coarse-grained biodetrital limestones, partly reef limestones, dark-grey to black limestones, at the lowermost part quartzites occur with alternations of clayey limestones, marls and sandstones
3065							erosional surface
3066–3713 (boring depth at 3713 m)	Middle Devonian (Eifelian)	*Icriodus expansus,* *I. subterminus*	CAI 3 (110 °C–200 °C)	~ 4000–4700 m	3100–5700 m		calcareous argillites at the base shales, quartzites and sandstones

50–90 °C. The calculated maximum overburden supports the estimated maximum thickness required for producing the observed CAI values, based on an assumed palaeothermal heat flow of 3.5 °C/100 m.

Vaklino borehole (Table 5)

The Vaklino borehole reaches a depth of 3480 m. The interval between 2660–3480 m is characterized by clayey limestones, bioclastic limestones, dolomites, and anhydrites in the upper part of this succession, all of which belong to the Middle Devonian. Condont elements show CAI 4, indicating temperatures of at least 190 °C. The top of this lower part is marked by an erosional surface. Middle Devonian and Upper Devonian sediments (1836–2260 m) overlie this erosional surface, interrupted by two major fault zones at 2647 m and 2319 m. The interval between 2291–3480 m has been dated based on conodont assemblages. The Middle Devonian carbonate-sequence is overlain by micritic limestones that probably belong to the Upper Devonian (based on their lithological similarity to other Upper Devonian sequences). Also, the transition to the Carboniferous, which has been placed below the dolerites (1747 m), is based on changing lithology only. According to miospores and conodonts, the youngest Palaeozoic sediments in this borehole belong to the Late Visean. CAI values of 1.5 suggest a palaeotemperature of at least 50 °C. This interval between 1045–1618 m is mainly composed of shales, sandstones, and rare micritic limestones with intercalations of a few cherts. Jurassic and younger sediments overlie a prominent erosional surface of Late Visean age. The succession is characterized by some erosional surfaces (Table 5) in Devonian sediments due to uplift and denudation. This may explain the relatively low maximum overburden, which is not in accordance with the estimated maximum overburden for producing the observed CAI values. However, based on the CAI data for the Lower Carboniferous and the calculated thicknesses, a palaeothermal heat flow of 3.5 °C/100 m is assumed at least for the Carboniferous.

Discussion

Temperature is the most sensitive parameter in hydrocarbon generation, though no measurable parameter can be directly converted to palaeotemperature (Tissot et al. 1987). However, maturation indices such as CAI offer an indirect approach. One important advantage of using CAI data for thermal maturity studies, even in core material, is that each sample with a sufficient number of determinable conodonts can be dated. The limitation regarding other thermal maturity estimation methods, such as vitrinite reflectance data, is due to the lower sensitivity of temperature ranges. In many cases, palaeotemperatures are different from present temperatures due to depth changes resulting from subsidence, erosion, and changes in geothermal gradient. It seems likely that the palaeogeothermal gradient was higher than the present one of ~ 3.0 °C/100 m, which has recently been determined in north-eastern Bulgaria (Bojadgieva and Gasharov 2001). The assumed palaeogeothermal gradient of ~ 3.5 °C/100 m in north-eastern Bulgaria is based on the observed CAI data and the calculated overburden (Tables 1–5). The latter parameter is also in accordance with the estimated thicknesses given by Haydoutov and Yanev (1997), and Yanev (2000). The estimated temperature ranges based on the CAI method are mainly the result of sedimentary burial. With the exception of the Gorniak borehole, there is no evidence for significant cover by thrusting and/or textural alteration of conodonts due to pressure solution effects. Any significant influence from magmatic activity is also lacking. Although heat from post-Carboniferous basalts influenced coalification in the southern Dobrudzha region, this can be excluded for the in-

Table 4. Belgun borehole: CAI data, stratigraphy and lithology

Depth (m)	Age	Conodont fauna and/or other fauna	CAI and estimated temperatures	Calculated maximum overburden (m)	Approximate maximum overburden (m) to produce observed CAI values (3.5 °C/100 m)	Textural alteration	Lithology/tectonics
0–1186 m	Jurassic and younger sediments						dark grey and black siltstones (?) and argillites with coal layers and sandstones and to a minor degree pyroclastic layers
1186 m	Jurassic/Visean discordance						erosional surface
1186–2860	Late Visean						alternation of shales and siltstones
1671–2092		*Gnathodus symmetricus*	CAI 1.5 (50–90 °C)	~ 2000–2500 m (~ 500 m Tournaisian)	1400–2500 m		alternation of argillites and siltstones with intercalated micritic limestones and marls
2860–2869							core loss
2869.4–2869.5	Famennian	*Ozarcodina regularis*, *Angulodus bidentatus*, *Acodina curvata*, *Polygnathus dubius*, *Po. webbi*	CAI 2 (60–140 °C)	~ 3300 m	1700–4000 m		fine-grained crinoidal limestones
2869.5–2872		*Palmatolepis glabra lepta*, *Pal. perlobata schindewolfi*, *Pal. gracilis gracilis*, *Polygnathus perplexus,* *Falcodus variabilis*	CAI 2 (60–140 °C)	~ 3300 m	1700–4000 m		dark-grey limestones with intercalated shales
2872–2872.8		*Palmatolepis glabra lepta*, *Pal. perlobata*, *Angulodus walrati*, *Polygnathus dubius*, *Spathognathodus strigosus*, *Falcodus variabilis*, *Hindeodella subtilis*, *Lonchodina* sp.	CAI 2, 2.5 (60–140 °C)	~ 3300 m	1700–4000 m		fine-grained dolomitic limestones
2872.8–2874		*Palmatolepis glabra lepta*, *Pal. perlobata schindewolfi*, *Pal. gracilis gracilis*	CAI 3, and CAI 4 (110–200 °C)	~ 3300 m	3100–5700 m	conodont surfaces partly recrystallized	
2874–2995 (boring depth at 3001 m)		*Palmatolepis glabra lepta*	CAI 3, and CAI 4 (110–200 °C)	~ 3300–3500 m	3100–5700 m		dark-grey limestones with high organic content and black siltstones and argillites with siderite concretions

Table 5. Vaklino borehole: CAI data, stratigraphy, and lithology

Depth (m)	Age	Conodont fauna and/or other fauna	CAI and estimated temperatures	Calculated maximum overburden (m)	Approximate maximum overburden (m) to produce observed CAI values (3.5 °C/100 m)	Textural alteration	Lithology/Tectonics
0–1044	Jurassic and younger rocks						
1045	Jurassic/ Visean boundary						erosional surface
1045–1532	Late Visean	Miospores from the interval at 1375 m: *Lycospora pusilla*					
1532–1618	Late Visean	*Gnathodus symmetricus*	CAI 1.5 (50 °C–90 °C)	~ 2000–2100 m (~ 500 m Tournaisian)	1400–2500 m		argillites, sandstones, micritic limestones and a few cherts
1618–1697							dolerites
1697–1698							micritic limestones
1698–1747							dolerites
1747	C/D boundary						the boundary is placed here based on the lithological change
1748–1863	?Late Devonian						pelletal-micritic limestone
1835							erosional surface
1836–1970	?Late Devonian						banded limestones
1971–2051							dolomites with rare micritic limestones
2051							erosional surface
2051–2299	Middle Devonian (Givetian)	*Polygnathus varcus, Icriodus* sp. *I. brevis*	CAI 3 (110 °C–200 °C)	~ 2800 m	3100–5700 m		“carbonate-sulfate formation” containing anhydrites, dolomites and less limestones
2299–2301		*Polygnathus ansatus, P. linguiformis weddigei, P. parawebbi, P. xylus xylus, P. varcus, Icriodus difficilis, I. eslaensis, I. laticarnatus, Ozarkodina ballai, O. lata*	CAI 3 (110 °C–200 °C)	~ 2800 m	3100–5700 m		“carbonate-sulfate formation” containing anhydrites, dolomites and less limestones
2301–2319 2301.70	Middle Devonian (Eifelian)	*Polygnathus linguiformis alveolus, P. linguiformis gamma morph.*	CAI 4 (190 °C–300 °C)	~ 2800 m	5400–8500 m		limestones and dolomites
2319							fault zone
2319–2647 2319–2321.60	Late Devonian (Famennian)	*Hibbardella* cf. *ortha, Palmatolepis glabra lepta, Pa. gracilis gracilis, Pa. marginifera marginifera, Pa. glabra distorta*	CAI 4 (190 °C–300 °C)	~ 2800 m	5400–8500 m		micritic limestones
2647							fault zone

Table 5, continued

2647.90–2651	Middle Devonian (Givetian)		CAI 4 (190 °C–300 °C)	~ 3150 m	5400–8500 m	sugary surface	limestones, partly dolomitized
2660							erosional surface
2660–3179	Middle Devonian (Givetian)						anhydrites, dolomites and some limestones
3179–3355	Middle Devonian (Eifelian)	*Icriodus* sp.	CAI 4 (190 °C–300 °C)	~ 3700–3850 m	5400–8500 m		conglomerates main ly composed of clayey limestones, bioclastic limestones and dolomites
3355–3480 (boring depth at 3480 m)	?Early Devonian						siliciclastics and carbonates

vestigated area based on the absence of basalts and/or igneous intrusions.

The data indicate variable sedimentation rates in the Devonian and Carboniferous. High sedimentation rates can be excluded for the Middle and Late Devonian based on the observed sedimentological features. During that time, dolomites, gypsum, and anhydrites formed along the eastern margin of the Moesian Terrane (Haydoutov and Yanev 1997, Yanev 2000, Seghedi et al. 2004). Furthermore, the sedimentological data show that during the Mid and Late Devonian, and during the Late Visean, tectonic activity in the Moesian Terrane caused subaerial exposure and consequent depositional breaks. The thicknesses of the sedimentary sequences seem to diminish to the east, and therefore increasing numbers of stratigraphic breaks occur. On the other hand, rapid subsidence followed by uplift and high erosion rates can be assumed within the Carboniferous (Boncheva 2004). This might have caused the high CAI values of 4 in the Vaklino borehole in the Middle and Upper Devonian. Post Carboniferous sediments can reach a thickness of several thousand meters. There is no carbonate basement in the areas with very thick Upper Carboniferous or Permian sediments in north-eastern Bulgaria (Yanev 2004, pers. comm.). Based on the observed Carboniferous CAI values, which are very low (CAI 1.5), the Late Carboniferous heating should have had no influence on the coalification of the Devonian sediments, since temperatures remained below the maximum reached during the Early Carboniferous. The post-Carboniferous sediments should likewise have had no influence on CAIs, because most of the Permian and Mesozoic cover has been subsequently affected by erosion. Finally, regarding hydrocarbons, it should be mentioned that lithological data support the assumption that some sediments, especially Devonian dolomites, may serve as potential reservoir rocks in north-eastern Bulgaria, although this seems unlikely based on high CAI data of Middle Devonian and Upper Devonian sediments. On the other hand, these rocks might have served as reservoir rocks due to the lateral migration of younger hydrocarbons. It is possible that bituminous sequences in the Lower Carboniferous may have acted as source rocks for hydrocarbons which have been found in the Mesozoic (Yanev 1997). Based on observed CAI temperature ranges of 50–90 °C it is possible that hydrocarbons occur in Carboniferous sediments, which is in accordance with illite crystallinity data from northern Bulgaria (Yanev and Stefanov 2002) and the vitrinite reflectance data of the Dobrudgea coal basin (Nikolov and Stoyanov 1988) representing the diagentic zone. Clearly, more data on the complex structural geology of the working area are necessary.

Conclusions

This regional study of thermal maturation patterns is mainly based on CAI data. The conclusions most relevant to the thermal and burial history of Palaeozoic sequences in north-eastern Bulgaria are:

- The stratigraphic position had a notable effect on the diagenesis/metamorphism of the sediments. Conodont alteration values range from CAI 1.5 to 4.5, and generally increase with increasing stratigraphic age. Diagenetic conditions were a dominant factor in Lower Carboniferous and Upper Devonian sediments, whereas the CAI data of Middle Devonian sediments indicate the transition to anchizonal conditions (CAI 4). The CAI data confirm a pre-tectonic maturation in all investigated boreholes. Tectonic faulting and thrusting generally has no influence on CAI values, except for the data from the Gorniak borehole.
- The maturation levels of Devonian and Early Carboniferous rocks are not the result of strong subsidence and/or overburden during post-Carboniferous. The time of maximum burial is placed in the Early Carboniferous. For shorter heating times, the calculated temperatures would be even higher.
- There is no indication that carbonate Palaeozoic sequences in the investigated area have been affected by igneous activity. Coal layers have been affected by igneous activity in the Dobrodgea coal basin by sills and dykes.
- A palaeo-geothermal gradient of 3.5 °C/100 m in the

Early Carboniferous is considered to be the most realistic value for north-eastern Bulgaria.

- The CAI data presented here cannot resolve the debate on the potential occurrence of hydrocarbons in Palaeozoic sediments in north-eastern Bulgaria, as pointed out by Yanev (1997). Generally, CAI values of 3 and 4 (in some cases 4.5) in Devonian sediments suggest burial temperatures of about 110–200 °C and 190–300 °C, respectively, resulting in the supramature state for organic matter. CAI data for Carboniferous sediments show values of 1.5, representing palaeotemperatures of at least 50 °C.
- CAI data provide important information on the burial temperatures of Palaeozoic rocks, though the question concerning the existence of Palaeozoic reservoir rocks requires further detailed investigation of the complex geological structure in north-eastern Bulgaria.

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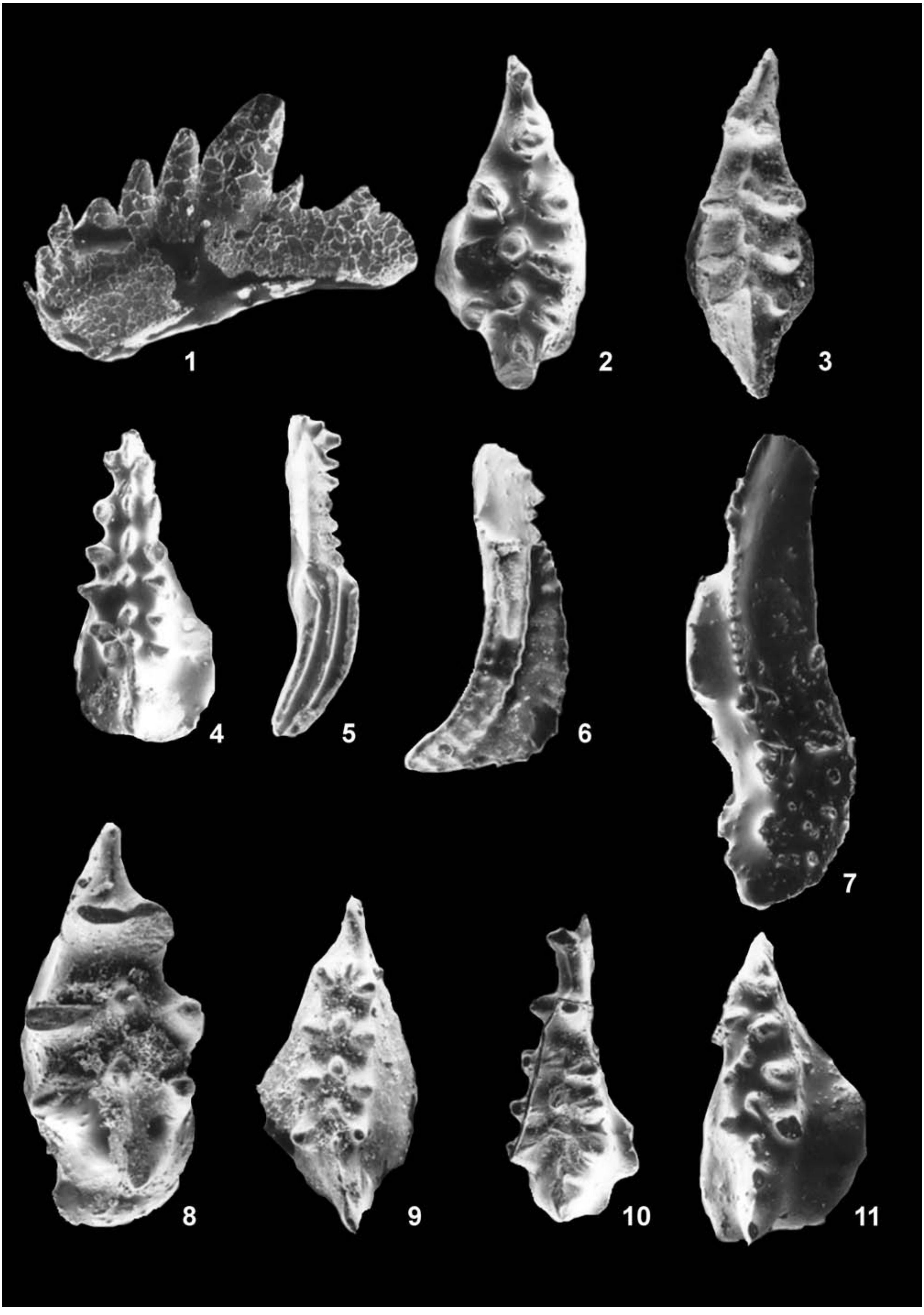
Plate I

1 – *Ozarkodina congesta* Stauffer; ×100; Ograzhden, 1518 m; Frasnian (CAI 3).
2 – *Icriodus werneri* Weddige; ×100; Ograzhden, 2396.7 m; Middle Devonian–Late Eifelian–Early Givetian (CAI 3).
3 – *Icriodus amabilis* Bultynck and Hollard; ×100; Ograzhden, 2396.7 m; Middle Devonian–Late Eifelian–Early Givetian (CAI 3).
4 – *Icriodus brevis* Stauffer; ×75; Vaklino, 2299.1 m; Middle Devonian–Givetian (CAI 3).
5 – *Polygnathus xylus xylus* Stauffer; ×75; Vaklino, 2299.3 m; Middle Devonian–Givetian (CAI 3).
6 – *Polygnathus parawebbi* Chatterton; ×75; Vaklino, 2299.2 m; Middle Devonian–Givetian (CAI 3).
7 – *Palmatolepis glabra distorta* Branson and Mehl; ×75; Vaklino, 2319.6–2321.6 m; Late Devonian–Famennian–*marginifera* Zone (CAI 4).
8 – *Icriodus aff. subterminus* Youngquist; ×100; Ograzhden, 2394 m; Middle Devonian–Late Eifelian–Early Givetian (CAI 3).
9 – *Icriodus aff. amabilis* Bultynck and Hollard; ×75; Ograzhden, 2395–2395.2 m; Middle Devonian–Late Eifelian–Early Givetian (CAI 3–4).
10 – *Icriodus orri* Klapper and Barrick; ×75; Ograzhden, 2395.1 m; Middle Devonian–Late Eifelian–Early Givetian (CAI 3–4).
11 – *Icriodus deformatus asymmetricus* Ji; ×75; Ograzhden, 2095–2096 m; Late Devonian (CAI 3).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