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ORIGINAL ARTICLE

Redox conditions during sedimentation of the Middle Jurassic (Upper Bajocian–Bathonian) clays of the Polish Jura (south-central Poland)

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Abstract Depositional redox conditions of the uppermost Bajocian–Bathonian (Middle Jurassic) ore-bearing clays of the Gnaszyn/Kawodrza area in the Polish Jura have been determined using an integrated geochemical (Th/U and U/Th ratios, degree of pyritisation (DOP), sulphur stable isotopes, biomarker analysis) and petrographic approach (measurements of pyrite framboid diameters, and microfacies analysis). The Th/U and U/Th ratios indicate that oxic conditions prevailed on the sea-floor during this interval, and ^{34}S isotopes suggest open-system conditions. DOP values, however, are rather scattered, and may reflect oxic, dysoxic, or anoxic conditions. We consider that the DOP values result from reducing conditions within the sediment and the chemistry of the pore-waters, rather than true sea-floor redox conditions. Pyrite framboid populations also indicate that dysoxic conditions prevailed within the sediment, beneath an oxygenated water column. Biomarker data did not provide any evidence of water column stratification or anoxia during sedimentation of the Middle Jurassic clays.

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Introduction

The Middle Jurassic (Bajocian–Bathonian) unconsolidated clay sediments of the Polish Jura (south-central Poland) have been studied by many geologists and palaeontologists over the past decades (e.g., the classic work of Różycki 1953). Extensive research began 10 years ago when the joint Polish-Danish team (see Poulsen et al. 1998) conducted the first interdisciplinary work on these sediments. In particular, the palaeontology of the sediments has received much attention, because of their abundant and well-preserved fossil content. Recently, Zatoń and Marynowski (2004, 2006) analysed the organic matter preserved in uppermost Bajocian clays and concretions. Szczepanik et al. (2007) undertook an analysis of redox conditions of the Middle–lower Upper Bathonian sequence exposed in the Gnaszyn clay-pit (see below). Marynowski et al. (2007a) investigated the composition and source of organic matter from the Middle Jurassic clays of Poland, providing for the first time detailed insight into the depositional environment using biomarkers. Wierzbowski and Joachimski (2007) thoroughly analysed the stable carbon and oxygen isotope ratios obtained from various calcareous fossils, providing data about the temperature regime of the Late Bajocian–Bathonian Polish Basin.

This paper presents an integrated inorganic geochemical, petrographic and microfacies analysis of the uppermost Bajocian–Bathonian clay sequence exposed in the Polish Jura in southern Poland. Combined with organic geochemical data obtained recently by some of the present authors (Marynowski et al. 2007a), our data has allowed, for the first time, the complex reconstruction of the Middle Jurassic redox conditions of the Polish Basin.

Palaeogeography and geological setting

Palaeogeography

During the Middle Jurassic, current-day Poland was situated at a latitude of approximately 40°N (Golonka 2000) (Fig. 1A). The Polish Basin was the eastern extension of the Mid-European epicontinental basin (Fig. 1B). It was bordered by the Fennoscandian land to the north, the Belorussian High and Ukrainian Shield to the east, the Bohemian Massif to the west, and the pre-Carpathian land to the south (see Dadlez 1989; Ziegler 1990; Feldman-Olszewska 1997) (Fig. 1B). It is generally assumed that the Moravian Gate (situated to the west of the Polish Basin, between the Bohemian Massif and pre-Carpathian land) was closed, at least until the Late Bathonian transgression, and the only connection with the Tethys Ocean existed in the southeast via the East-Carpathian Gate (e.g. Dayczak-Calikowska and Moryc 1988; see also Feldman-Olszewska 1997) (Fig. 1B). During the Bathonian, the Polish Basin broadened successively, attaining its greatest extent in the Late Bathonian, when almost the entire area of the Polish Lowlands was submerged (Matyja and Wierzbowski 1998). Sedimentation was dominated by clastics, the depositional systems of which followed transgressive-regressive cycles (Feldman-Olszewska 1997). It has been suggested that Fennoscandia and the Bohemian Massif exerted the primary influence on the transport directions of clastic material, as well as on the infilling character of the Polish Basin (Dadlez 1997; Marynowski et al. 2005; 2007a).

Geology, stratigraphy, and palaeontology

The Polish Jura is a monoclinal structure extending from southeast to northwest of the Cracow-Wieluń Upland in south-central Poland (Fig. 1C). The Upper Bajocian through Bathonian epicratonic sediments, widely known as

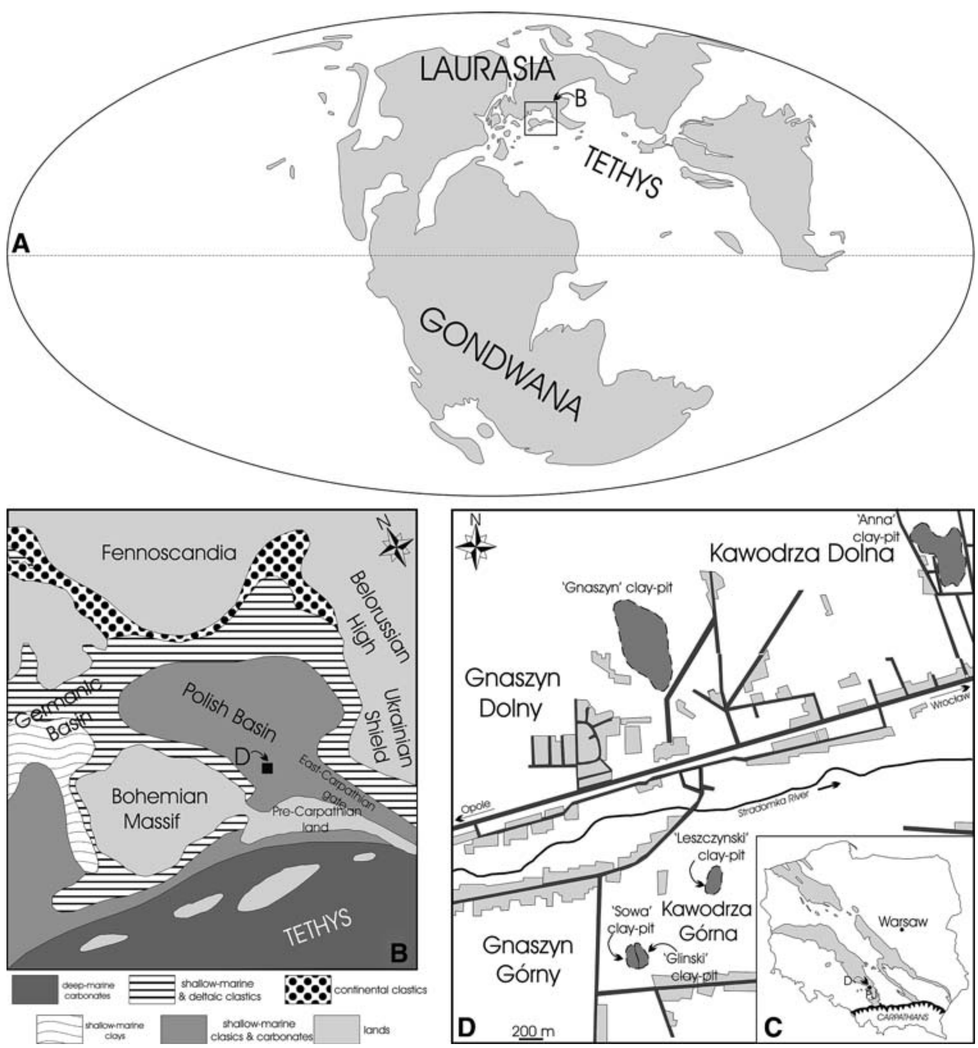

Fig. 1 *A* Polish Basin and adjacent areas on the background map of Laurasia (contour after Golonka 2000, simplified). *B* Polish Basin and adjacent areas during the Bajocian–Bathonian times (after Ziegler 1990, modified). *C* Map of Poland with Jurassic deposits indicated (*shaded*). *D* Gnaszyn/Kawodrza area near Częstochowa, in the Polish Jura, with the location of the studied clay-pits indicated

the ore-bearing Częstochowa Clay Formation (e.g. Dayczak-Calikowska et al. 1997; Kopik 1998; Majewski 2000; Matyja and Wierzbowski 2000; Zatoń and Marynowski 2006; Szczepanik et al. 2007), comprise a lithologically monotonous sequence of dark-grey, unconsolidated clays, with variable coarser fraction content. The clays are intercalated with massive siderite beds, and sideritic/calcitic concretions which occur as single bodies or fairly continuous horizons. The sediments have a gentle dip angle of <2° towards the northeast (Znosko 1960). They are often capped by consolidated Callovian deposits, consisting of limestones, sandstones and sandy-limestones (Dayczak-Calikowska et al. 1997).

The Middle Jurassic ore-bearing clays are widely exposed in several pits in the Polish Jura. The present study focuses on the Gnaszyn/Kawodrza area, near Częstochowa (Fig. 1C, D), where the Upper Bajocian through Bathonian sequence is exposed in a relatively small area. This area is confined to the northern sedimentary region (sensu Różycki 1953), which compared to the southern region, is characterised by a more fully developed and thicker sequence of ore-bearing clays. The clays of this area are rather monotonous with respect to facies, with no obvious variation in the field. The depositional environment of the clays has been interpreted as quiet marine, generally below the storm-wave base (see Matyja et al. 2006a, b, c).

The common ammonite fauna has enabled the biostratigraphical subdivision of the clays. The clays range from the Upper Bajocian Parkinsoni Zone to the Upper Bathonian Orbis Zone (Kopik 1998; Matyja and Wierzbowski 2000; Matyja et al. 2006a, b, c; Zatoń 2007a). The basal Middle Bathonian Progracilis Zone has not been determined by ammonites in the investigated area, although Matyja and Wierzbowski (2000) stated that this zone is present at Faustianka, in the northern part of the Polish Jura. On the basis of dinocysts, Poulsen (1998) assumed that the highest zone of the Upper Bathonian, the Discus Zone, may also be present in the northern Polish Jura. Barski et al. (2004), also on the basis of dinocysts, confirmed the presence of the Discus Zone in the southern part of the area.

General characteristics of the investigated sediments

The sediments investigated in the Gnaszyn/Kawodrza area are exposed in six active clay-pits (Fig. 1D):

Sowa clay-pit. This clay-pit is situated in the western part of Kawodrza Górna (Fig. 1D). The sequence consists of ~8-m dark-grey clays, intercalated with three massive siderite beds. In the lower part of the section, fossil-bearing calcite concretions are extremely common (Zatoń and Marynowski 2004, 2006). Since 2004, the majority of the exposure has been covered by clays from the workings above. The section represents the uppermost Bajocian (Parkinsoni Zone, Bomfordi Subzone)-lowermost Bathonian (Zigzag Zone, Convergens Subzone) (see Matyja and Wierzbowski 2000; Matyja et al. 2006a; Zatoń and Marynowski 2004, 2006).

Gliński clay-pit. This clay-pit is situated adjacent to Sowa (Fig. 1D). The section consists of ~8-m dark-grey clays, with an admixture of sandy material in places, intercalated with a few horizons of massive siderites with limonitised crusts and loosely scattered carbonate concretions. The section represents the Lower Bathonian (Zigzag Zone, Convergens to Macrescens Subzone) (see Matyja and Wierzbowski 2000; Matyja et al. 2006a). However, an ammonite found in the lowermost part of the section (Zatoń 2007a) points to the additional presence of the uppermost Bajocian (Bomfordi Subzone).

Leszczyński clay-pit. This clay-pit is located in the northern part of Kawodrza Górna (Fig. 1D). The section exposed consists of ~12 m dark-grey clays intercalated with two massive siderite horizons, and one horizon of small carbonate concretions in its uppermost part. The exposure represents the Lower Bathonian (Zigzag Zone, Yeovilensis Subzone and Tenuiplicatus Zone) (see Matyja and Wierzbowski 2000, 2001; Matyja et al. 2006b; Zatoń et al. 2006a).

Gnaszyn clay pit. This is the largest and most well-known (see Gedl et al. 2003; Zatoń et al. 2006a, 2007; Matyja et al. 2006c, 2007b; Zatoń 2007a, b) active clay-pit in the whole region, situated at Gnaszyn Dolny (Fig. 1D). It exposes ~20-m dark-grey clays intercalated with several carbonate nodules, some of which are laterally extensive. The sequence represents the Middle Bathonian (Subcontractus Zone)–Upper Bathonian (Hodsoni Zone) (see e.g. Zatoń 2007b, Zatoń et al. 2006a; Matyja et al. 2006c; Szczepanik et al. 2007). It should be noted that the NW European Hodsoni Zone is equivalent to the Middle Jurassic Bremeri Zone in the Sub-Mediterranean biostratigraphic scheme. However, as the ammonite fauna from the Bathonian of the Polish Jura is impoverished in strictly Sub-Mediterranean forms (see Matyja and Wierzbowski 2000), we use the NW European scheme.

Anna clay-pit. This clay-pit is situated at Kawodrza Dolna (Fig. 1D). Approximately 15 m of clays are intercalated with three horizons of carbonate nodules in the lower part of the section, and a massive sandy siderite bed in its uppermost part (see Matyja and Wierzbowski 2003; Zatoń 2007a). The section represents the Upper Bathonian (Hodsoni–Orbis Zone).

The above sections have been documented in several papers cited above, and this data will not be reproduced again. Instead, a schematic lithostratigraphical composite section is presented, along with an indication of the biostratigraphic range of each section investigated (Fig. 2).

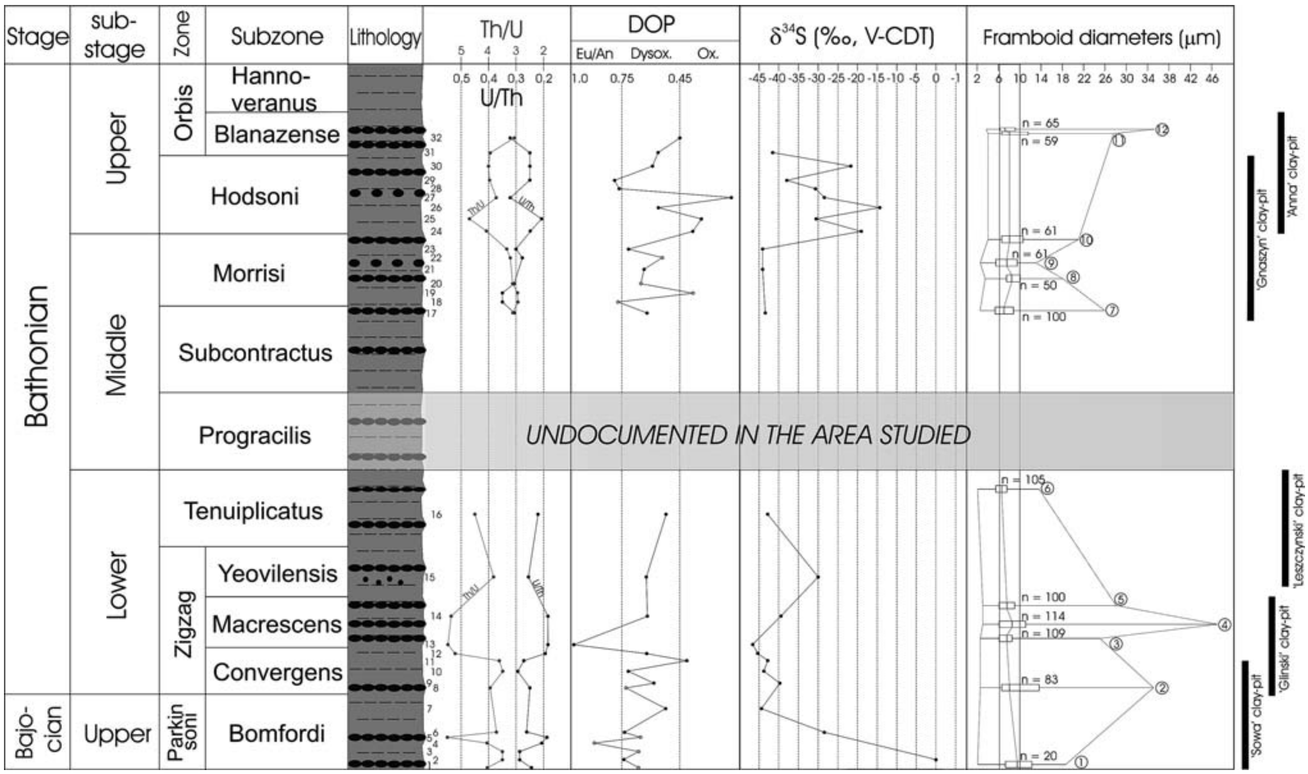

Fig. 2 Schematic lithological section of the uppermost Bajocian–Bathonian ore-bearing clays of the Polish Jura, with the redox indices used in the present study: Thorium/Uranium (Th/U) ratio, Uranium/Thorium (U/Th) ratio, degree of pyritisation (DOP), stable isotopes of sulphur ^{34}S, and framboid pyrite diameters (shown here as *box-and-whisker plots*, the *grey bar* indicates the range of mean size diameter), *Eu/An* euxinic/anoxic, *Dysox.* dysoxic, *Ox.* oxic, *n* number of measurements, the *numbers in circles* correspond to the numbers of samples used for microfacies study. *Black bars* on the *right* show the stratigraphic ranges of the clay sections investigated, *1–32* numbers of clay samples analysed

Palaeontological content

The Bajocian–Bathonian ore-bearing clays are known for their rich and diverse fossils, many of which have not yet been fully described. The clays and co-occurring carbonate concretions contain diverse ammonites (e.g. Kopik 1998, 2006; Matyja and Wierzbowski 2000; Matyja et al. 2006a, b, c; Zatoń and Marynowski 2006; Zatoń et al. 2007), brachiopods (Wiśniewska-Żelichowska 1978), bivalves (e.g. Pugaczewska 1986), gastropods (Kaim 2004), echinoderms (see Gedl et al. 2003; Boczarowski 2004; Szczepanik and Sawłowicz 2005; Salamon and Zatoń 2007), crabs (Krobicki et al. 2005; Krobicki and Zatoń 2008), various encrusting biota (see Zatoń et al. 2006b), diverse fossil conifer wood some of which possess uniquely preserved original biomolecules (Marynowski et al. 2007b), caytoniacean leaves (Zatoń et al. 2006a), as well as various microfossils such as spores and pollen, dinocysts, ostracods, foraminifers (e.g. Błaszyk 1967; Poulsen 1998; Gedl et al. 2003), cephalopod arm hooks, pterobranchs (Kulicki and Szaniawski 1972) and polychaete jaws (Szaniawski 1974). Vertebrate regurgitates containing rich invertebrate remains can also be found in large numbers (Zatoń et al. 2007; Zatoń and Salamon 2008).

Material and methods

Clay and carbonate concretion samples were gathered from the uppermost Bajocian–Bathonian in active clay-pits in the Gnaszyn/Kawodrza area (Fig. 1D). Only the basal Middle Bathonian Progracilis Zone and the top Upper Bathonian Discus Zone have not been sampled because of their uncertain and/or undocumented occurrence in the area. Several geochemical and microscopic methods have been employed in order to determine the environmental conditions prevailing during sedimentation of the ore-bearing clays. Geochemical parameters are discussed below, and apart from the biomarker data, are documented in Table 1.

U/Th and Th/U ratios

These ratios can be used to infer redox conditions during deposition of the sediment, and the U/Th ratio is considered to be a more reliable indicator of redox than other trace metals, such as V/Cr or Ni/Co ratios (see Jones and Manning 1994). Generally, low U/Th ratios (<0.75) indicate well-oxygenated conditions, whilst high values (>1.25) indicate anoxic conditions, and those between 0.75 and 1.25 indicate dysoxic conditions (see Jones and Manning

Table 1 Geochemical parameters used in the present study of clay samples

Sample number	Th (ppm)	U (ppm)	Th/U	U/Th	DOP	δ^{34}S (V-CDT)
1	9.8	2.4	4.08	0.24	0.66	nd
2	10.2	2.9	3.52	0.28	0.74	0.0
3	11.0	3.1	3.55	0.28	0.65	nd
4	11.3	2.4	4.1	0.21	0.89	nd
5	12.1	2.2	5.5	0.18	0.63	nd
6	12.3	3.3	3.73	0.27	0.73	−28.5
7	nd	nd	nd	nd	0.54	−44.4
8	13.3	3.4	3.91	0.25	0.71	nd
9	nd	nd	nd	nd	0.58	−39.8
10	12.8	3.7	3.46	0.29	0.72	−43.8
11	12.8	3.5	3.66	0.27	0.42	−42.9
12	15.6	3.0	5.2	0.19	0.64	−45.4
13	17.4	3.2	5.44	0.18	0.9	−46.7
14	16.0	3.0	5.33	0.18	0.63	−39.4
15	9.9	2.6	3.81	0.26	0.65	−30.0
16	9.5	2.1	4.52	0.22	0.53	−42.9
17	10.2	3.2	3.18	0.31	0.64	−43.4
18	11.0	3.8	2.89	0.34	0.77	nd
19	13.1	4.5	2.91	0.34	0.35	nd
20	11.5	3.6	3.19	0.31	0.67	nd
21	nd	nd	nd	nd	0.56	−44.1
22	8.4	2.6	3.23	0.31	0.67	nd
23	11.9	3.6	3.3	0.30	0.72	−44.1
24	12.0	2.9	4.14	0.24	0.38	−19.0
25	14.6	3.1	4.71	0.21	0.34	−30.5
26	nd	nd	nd	nd	0.56	−14.3
27	9.9	3.2	3.1	0.32	0.18	−28.5
28	nd	nd	nd	nd	0.77	−30.7
29	9.9	2.5	3.96	0.25	0.78	−38.0
30	11.2	2.8	4.0	0.25	0.61	−21.7
31	9.0	2.3	3.91	0.25	0.57	−41.6
32	9.0	2.9	3.1	0.32	0.45	nd

nd Not determined

1994). Myers and Wignall (1987; see also Wignall and Myers 1988; Wignall 1994) used the Th/U ratio for distinguishing redox conditions. Detrital sediments, such as mudstones and clays, have higher Th contents than carbonates, and the Th/U ratio in anoxic mudstones is generally less than 2. In mudstones deposited in oxygenated conditions, the Th/U ratio typically ranges from 3 to 5 (e.g. Wignall 1994).

Twenty-seven clay samples have been analysed for U and Th concentrations using INAA and ICP-AES (ACTL-ABS, Canada). U and Th contents are given in ppm. Samples from the Subcontractus–Hodsoni Zones were collected in Gnaszyn Dolny (Gnaszyn clay-pit) originally for the study of Szczepanik et al. (2007). We have used here only the samples that correlate closely to those horizons from which one of us (MZ) obtained samples for DOP and sulphur stable isotope analysis. The same procedure was applied to some of the samples collected by one of us (PS) from the uppermost Bajocian–lowermost Bathonian section of the Sowa clay-pit at Kawodrza Górna.

Degree of pyritization (DOP)

Raiswell et al. (1988) showed that the DOP index allowed the distinction between oxic, dysoxic, and euxinic environments. The concept of DOP as a palaeoenvironmental tool is based upon the microbiological processes related to sulphate reduction (see Raiswell and Berner 1985a, b; Raiswell et al. 1988, 2001; Wignall 1994; Strauss 1997). Under euxinic (that is permanently anoxic) conditions, practically all of the available reactive iron reacts to form pyrite. Sediments formed under such conditions record DOP values >0.75. Dysoxic conditions result in DOP values from 0.45 to 0.75. Under oxic conditions, only small amounts of pyrite form and therefore the values are low, generally <0.45 (Raiswell et al. 1988, 2001; see also Wignall 1994). In well-oxygenated, normal-marine conditions, the DOP values are generally around 0.4 (Hatch and Leventhal 1992).

Two sets of clay samples have been used: The first (23 samples, black points on Fig. 2) was collected by the senior author (MZ) and analysed in the laboratory of the School of Earth and Environment, University of Leeds, UK. The second set (nine samples, grey points on Fig. 2) was collected by PS and analysed in the Institute of Geological Sciences, Jagiellonian University, Cracow, Poland. DOP was calculated according to the formula of Raiswell and Berner (1985a, b): DOP = $Fe_{py}/Fe_{py} + Fe_{HCl}$, where Fe_{py} is the percent of pyrite iron (calculated by multiplying total S[%] × 0.871) and Fe_{HCl} is the percent of reactive iron, dissolved in hot and concentrated HCl. Fe_{HCl} was extracted from 100 mg of powdered sample by 1 N HCl during 24 h at room temperature and measured by ICP-OES (at Leeds) and F-AAS (at Cracow). The whole analysis follows that described by Raiswell et al. (1988).

Pyrite framboid analysis

The application of pyrite framboids as redox indicators was first presented from modern settings by Wilkin et al. (1996) and subsequently applied to ancient (Jurassic) environments by Wignall and Newton (1998). Since then, framboid analysis has been utilised in various sedimentary rocks of different ages (e.g. Wignall and Twitchett 2002; Wignall et al. 2005; Racki et al. 2004; Bond and Wignall 2005; Marynowski et al. 2007c; Shen et al. 2007). Wignall and Newton (1998) presented a thorough discussion of framboid diameters as palaeoenvironmental indicators.

Measurements of pyrite framboid diameters were carried out on a CAMSCAN scanning electron microscope (SEM) housed at the School of Earth and Environment, University of Leeds, using 12 carbon-coated polished blocks prepared from samples of the carbonate concretions. The carbonate concretions were chosen for the framboid study because, being well lithified, they were easily prepared as polished blocks. It is important to note that the size-frequency distribution of pyrite framboids from the carbonate concretions does not differ significantly from those enclosed in the host clays (MZ & PS, pers. observ.), and thus the framboid distribution of the concretions likely reflects the true depositional conditions. Framboid measurements were undertaken in back-scattered electron (BSE) mode. Diameters were measured directly from the screen at magnification ×2,460 (see Wignall and Newton 1998 for detailed procedures). Such a procedure may underestimate true framboid diameters; however, calculations show that standard deviations from their real diameter do not exceed 10% (Wilkin et al. 1996; see also Wignall and Newton 1998; Wignall et al. 2005). In each sample, at least 50 (ideally >100) framboids were measured, depending on the overall abundance of framboids in each sample. In one case, only 20 measurements were possible due to the extremely low number of framboids in that sample.

Sulphur stable isotopes

Sulphur (^{34}S) isotope analyses extend the scope of methods described above used in the reconstruction of depositional environments, and provide a control on these results. Furthermore, ^{34}S allows the determination of either open or closed system conditions. In the first case, the diffusion of sulphates dissolved in a water column into the underlying sediment is practically unlimited (see Strauss 1997).

Sulphur (^{34}S) isotope analyses were performed on 22 clay samples using a Micromass Isoprime continuous flow mass spectrometer coupled to a Eurovector Elemental Analyser at the School of Earth and Environment, University of Leeds, UK. Metal sulphides, in this case CuS, were weighed (0.050–0.090 mg) in tin cups and converted to SO_2 by flash combustion at 1,020°C in the presence of oxygen. Excess oxygen is removed by reaction with hot copper at about 650°C and the SO_2 is separated from other impurities using a chromatographic column and a helium carrier gas. $^{34}S/^{32}S$ is derived from the integrated mass 66 and 64 signals from the pulse of sample SO_2, compared to those in an independently introduced pulse of reference gas. These ratios are then calibrated using international standards to the Vienna-Canyon Diablo Troilite (V-CDT) scale in per mill notation (‰). The precision obtained for repeat analysis of standard materials is generally better than ±0.3‰ (1 standard deviation).

Microfacies analysis

Petrographic investigation was carried out on the same 12 samples that were analysed for the framboid diameter study. Thin sections were produced in the polishing laboratory at the School of Earth and Environment, University of Leeds. Again, these samples were chosen because unlike the host soft clays, they are lithified and thus much better suited for collecting as oriented samples and for thin section preparation. The bioclast composition of the carbonate concretions does not differ from that of the host clays, as revealed by washing and sieving of clay samples by one of us (MZ). Therefore, it may be assumed that the carbonate concretion thin-section data reflects the conditions prevailing during deposition of the host clays.

The microscopic observations were conducted using an Olympus polarization microscope coupled with Leica DFC 320 camera. In the each oriented sample, >200 to >400 bio- and lithoclasts were counted, moving through several vertical surfaces. However, only common quartz and, characteristic for some samples, ooids were counted. Each clast detected was counted and determined, where possible. The method follows that of Flügel (1982).

Results

U/Th and Th/U ratios

The U/Th values for the uppermost Bajocian–Bathonian sediments range from 0.18 to 0.34 (Fig. 2). From the uppermost Bajocian to the Tenuiplicatus Zone of the Lower Bathonian, the values are confined to narrow interval between 0.18 and 0.29. Around the Subcontractus/Morrisi Zone boundary of the Middle Bathonian, the U/Th ratio increases to 0.34, but above this it returns to the values similar to the lower part of the sequence. The Th/U values range from 2.89 to 5.5 (Fig. 2).

U/Th and Th/U ratios were recently studied from the Middle Bathonian–lowermost Upper Bathonian of the Gnaszyn clay-pit by Szczepanik et al. (2007). However, this study is the first to present such data for the entire exposed uppermost Bajocian–Bathonian sequence of the Gnaszyn/Kawodrza area.

DOP

DOP values for the investigated sections are widely scattered, ranging from 0.18 to 0.9. Most values, however, are confined to the interval between 0.45 and 0.75 (see Fig. 2). DOP results for the entire exposed uppermost Bajocian–Bathonian clays of the area are presented here for the first time.

Pyrite framboids

In the all samples studied, framboid diameters vary widely from 2 to 47 µm. Their mean values, however, occupy a quite narrow range, between ~6 and 10 µm (Fig. 2). Numerous large framboids (13–47 µm) also occur. Although pyrite framboids are scattered in the sediment matrix, many of them are associated with fossils (e.g. they fill foraminifer skeletons, ammonite septa, empty spaces of echinoderm stereom or wood cells). Those occurring in fossils have not been measured. Beside framboids, other forms of pyrite, such as euhedral crystals, also occur. Apart from the Bajocian/Bathonian transition (Zatoń et al. 2008), the pyrite framboid distributions are presented here for the first time.

Sulphur stable isotopes

The investigated sequence records negative δ^{34}S values, ranging from −14.3 to −46.7‰ (Fig. 2). Only one strongly positive value of δ^{34}S (0‰), has been detected, occurring in the lowermost part of the uppermost Bajocian (Bomfordi Subzone). Generally, there is a clear trend towards negative values in the lower part of the sampled uppermost Bajocian–Lower Bathonian section. An increase in δ^{34}S values occurs at the beginning of the Upper Bathonian, but these values are still negative. This is the first time that sulphur stable isotope analyses have been presented for this interval of the Polish Jura.

Microfacies analysis

The quantitative and qualitative composition of each thin-section studied is shown in Fig. 3. Quartz grains clearly dominate. Ooids are present only in two samples (samples 8, 9, see Fig. 3). Amongst the bioclasts, benthic foraminifers (uni- and multiserial), echinoderms (crinoid ossicles, echinoid spines, holothurian sclerites and other indeterminate ones) and filaments (shell fragments) are numerous, but their relative proportions vary between samples. Less numerous ostracods, bryozoans, gastropods, some problematic taxa and wood fragments also occur in the samples studied; however, they are not figured because of their single occurrence in only a few samples. The shelly fauna is disarticulated. In some samples, distinct pits are visible (e.g. sample 5), probably made by boring algae or fungi. Ostracods occur either as articulated and disarticulated carapaces, but the latter dominates. Echinoderm ossicles exhibit a well-preserved stereom structure, in the interspaces of which pyrite and siderite crystals occur. The bioclasts are scattered and densely packed in the matrix, without any signs of sorting visible. In some places, however, the litho- and bioclasts are distinctly aligned by bioturbation. Only in samples 8–10 is there a lack of bioturbation.

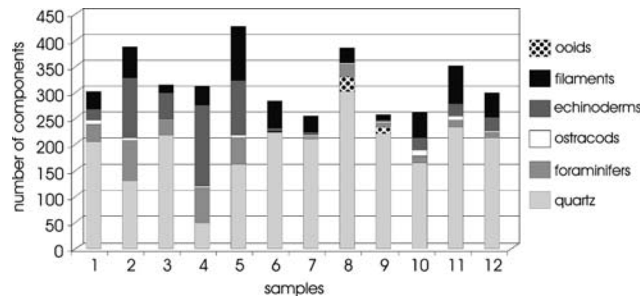

Fig. 3 Ratio of particular bio and lithoclastic components in the concretion samples investigated. Here, the sample numbers correspond directly to the sample numbers in *circles* shown in Fig. 2

The matrix in samples 2–6 is sparitic and composed of sideritic spherules. These samples come from massive siderite beds of Lower Bathonian (Zigzag to Tenuiplicatus Zones) of Kawodrza Górna. The remaining samples come from carbonate nodules and are characterised by a micritic matrix. The quartz grains are sharp-edged and do not show any signs of abrasion. Besides quartz, the samples also contain single crystals of ferrous oxides (e.g. sample 1), chlorite and barite (samples 2, 5), sphalerite crystals with inclusions of organic mater (sample 6).

Taking into account the most numerous components, samples 1–7 and 10–12 represent a general filament-echinoderm-foraminifer microfacies. However, according to the various contribution of particular components in the samples, sub-microfacies can also be distinguished, e.g. an echinoderm-foraminifer-filamentous sub-microfacies (samples 2–4) or a filamentous-echinoderm-foraminifer sub-microfacies (samples 5–7, 10–12). Other samples, from the Middle Bathonian (Morrisi Zone) of Gnaszyn Dolny, represent an ooid-filamentous-foraminifer and an ooid-foraminifer-filamentous sub-microfacies (samples 8, 9) (Fig. 4).

Organic matter analysis

All organic geochemical data are based on previously published materials and is presented here as a comparison for the redox analyses. Total organic carbon (TOC) contents from the Middle Jurassic clays ranged from 0.7 to 5.5% with the average and the most common content between 1 and 2%. One of the major factors controlling the TOC concentration is the carbonate content of the clays (Bojesen-Koefoed 1996; Marynowski et al. 2007a). Kerogen from the clays was characterised by Bojesen-Koefoed (1996) as terrestrial type III (according to Espitalié et al. 1985). Taking into account the molecular composition of the extracts from the clays, terrestrial biomarkers such as high molecular weight *n*-alkanes with an odd carbon number predominance (especially n-C_{25}, n-C_{29}, n-C_{31}), fernenes (see Zatoń et al. 2006a, Marynowski et al. 2007a), perylene, cadalene,

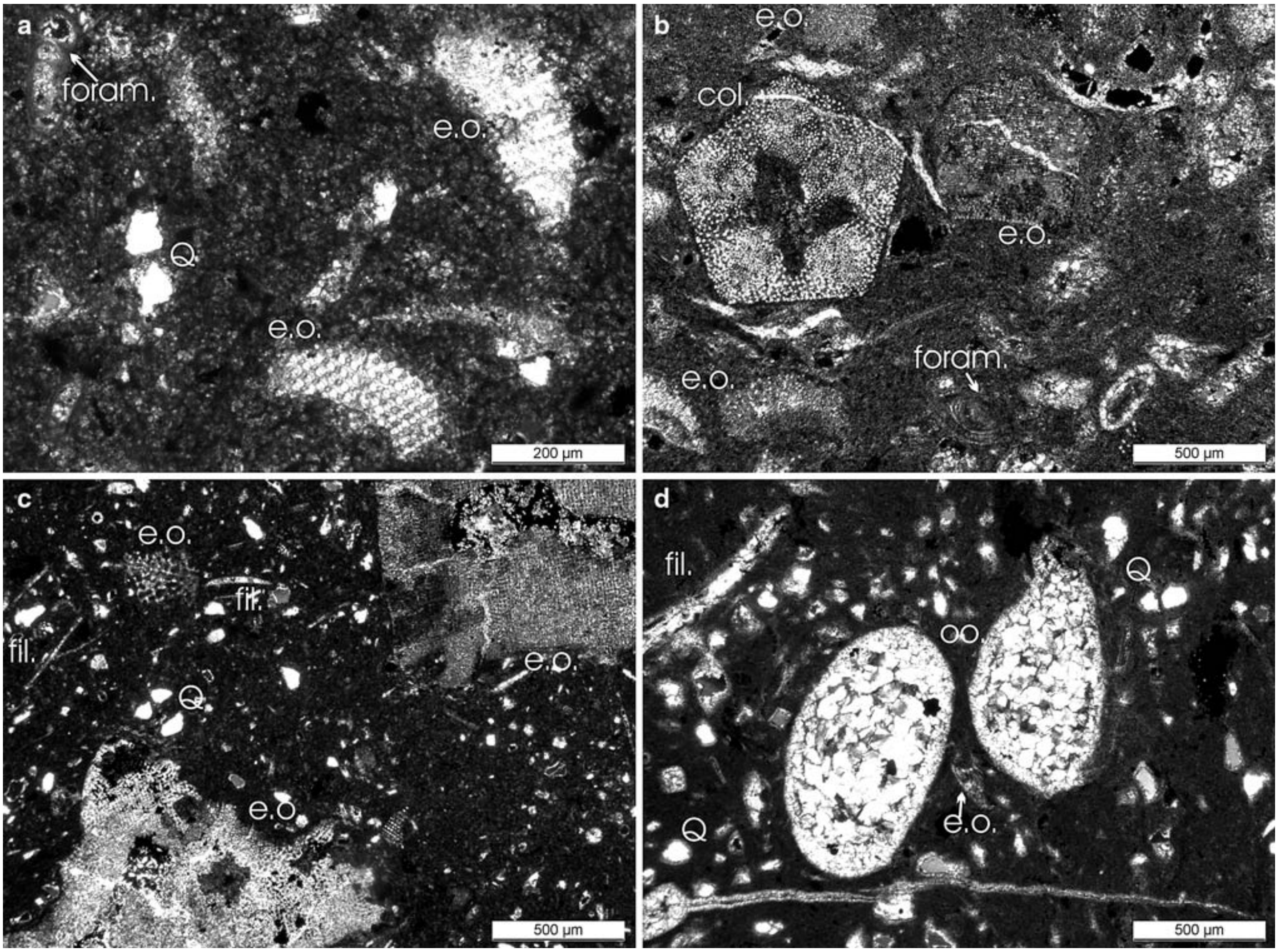

Fig. 4 **a**, **b** Echinoderm-foraminifer-filamentous sub-microfacies (samples 2 and 3 in Fig. 2, respectively). **c** Filamentous-echinoderm-foraminifer sub-microfacies (sample 10 in Fig. 2). **d** Ooid-foraminifer-filamentous sub-microfacies (sample 9 in Fig. 2), *col.* crinoid columnal, *e.o.* echinoderm ossicles, *fil.* filaments, *foram.* foraminifer, *oo.* ooids, *Q* quartz

simonellite, retene, calamenene, cadina-1(10),6,8-triene and dehydroabietane (Marynowski et al. 2007a) usually dominate in the aliphatic and aromatic fractions confirming the preponderance of terrestrial organic matter (OM) in the clays.

The OM from the Middle Jurassic Polish Jura is immature. Values of T_{max} obtained from Rock Eval analysis are between 415 and 425°C (Bojesen-Koefoed 1996). Immature OM character is emphasised by vitrinite reflectance values between 0.25 and 0.30% (Marynowski et al. 2007a) and the occurrence of relatively unstable polar biomolecules in the fossil wood fragments (Marynowski et al. 2007b, 2008a, b).

Discussion

The Upper Bajocian–Bathonian sequence of the ore-bearing clays of the Gnaszyn/Kawodrza area is lithologically very monotonous, consisting of unconsolidated, dark-grey clays without distinct facies changes. The only exception is the occurrence of loose or continuous horizons of carbonate nodules. In some intervals, encrusted and bored hiatus concretions (sensu Voigt 1968) occur, indicating periods of non-deposition and/or erosion of the host sediment (Zatoń et al. 2006b). The inorganic geochemical indices studied here do not show significant variations, suggesting stable redox conditions prevailed in the Polish Basin during Late Bajocian–Bathonian time-interval. Szczepanik et al. (2007) reached a similar conclusion based on different geochemical analyses of the Middle/Upper Bathonian sequence exposed in the Gnaszyn clay-pit.

The U/Th and Th/U ratios indicate that the redox conditions were oxic at the sea-floor. All the U/Th ratios are well below 0.75, and almost all Th/U ratios fall between 3 and 5 (only two Th/U ratios, 2.89 and 2.91, are lower, but still very close to 3). The highly negative stable δ^{34}S isotope values, although highly variable throughout the entire section (see Fig. 2), point to open-system conditions. The highest value (0‰) at the base of the sequence studied points to more closed conditions. However, such a value may be attributed to localised reducing conditions, which may have occurred when organic matter, for example in the form of wood fragments which are very common in the whole sequence, decayed (see Zatoń and Marynowski, 2004; Marynowski et al. 2007a). The analysis of organic matter from the same horizons by Zatoń and Marynowski (2004, 2006), further supports a well-oxygenated environment. The benthic faunal assemblages, studied by Matyja et al. (2006a) further point to optimal living conditions in a well-oxygenated environment, with sufficient food supply.

As shown in Fig. 2, DOP values vary widely throughout the section, and are not indicative of any one redox regime, rather out of accordance with the previously discussed indices. Szczepanik et al. (2007) obtained similar results and suggested that the DOP values reflect diagenetic processes within the sediment rather than redox conditions in bottom waters. Roychoudhury et al. (2003) stated that the DOP values reflect the conditions within the sediment and chemistry of its pore waters. Therefore, it seems that the scattered DOP values throughout the uppermost Bajocian–Bathonian section reflect the chemistry of the sediment. Similar dysoxic/anoxic conditions were also reflected in the V/V + Ni values (Szczepanik et al. 2007). The authors of the present paper also obtained similar 'false' results of V/V + Ni for this sequence, but they are not shown in Fig. 2. Marynowski et al. (2007a) stated that more reducing conditions prevailed below the sediment-water interface, where the decay processes of organic matter formed specific anoxic microenvironments. Such a microenvironment was favourable for the formation of carbonate concretions and even the massive carbonate (siderite) beds. This conclusion is supported by the trace fossil assemblage, which is poorly diversified in the uppermost Bajocian–Lower Bathonian sediments of the Sowa and Gliński clay-pits (Matyja et al. 2006b).

Pyrite framboid distributions exhibit a wide range of diameters throughout the sequence (Fig. 2). Similar size-frequency distributions were observed in the Middle/Upper Bathonian clay samples from the Gnaszyn clay-pit by Szczepanik et al. (2007). The presence of large framboids in each sample studied, together with their high mean values (6–10 μm), is suggestive of their formation within a dysoxic sediment, beneath an oxygenated water column (see Wignall and Newton 1998). Small framboids, below 5 μm, are also present in the samples, but only in very small numbers. Additionally, the presence of many euhedral pyrite crystals reflect their diagenetic origin within the sediments (see also Szczepanik et al. 2007). Thus, the framboids present are considered to form diagenetically, within the sediment in open-system conditions, when large amount of dissolved sulphate was delivered from the overlying water column. This may have been promoted by bioturbators penetrating the surficial sediments.

The TOC content is relatively high in many clay samples, but the molecular character of extracts from clays suggests that sedimentary conditions during OM deposition were oxic to suboxic. This is indicated by the relatively low concentrations of C_{33}–C_{35} homohopanes, moderate to high Pr/Ph values and no compounds characteristic of anoxia and water column stratification (Zatoń and Marynowski 2004, 2006; Marynowski et al. 2007a). Raised TOC values are related to the enhanced transport of terrestrial OM into the Polish part of the Mid-European Epicontinental Basin, which is reflected in the presence of type III kerogen and the high relative concentration of terrestrial biomarkers (see Marynowski et al. 2007a).

Oxic conditions are further supported by the fossil content. It is well-known that during dysoxia, the diversity of benthic organisms may be very low and during anoxia it may drop to zero (see Wignall and Myers 1988; Wignall 1993, 1994). Here, the common benthic organisms (either epi- and endofauna) detected either macroscopically or in thin section, and the presence of bioturbation traces, points to well-oxygenated bottom waters. Taphonomy supports this interpretation. The common ammonite fauna does not show any evidence for 'half-ammonite preservation' (e.g. Seilacher et al. 1976; Tanabe et al. 1984; Maeda and Seilacher 1996). This type of ammonite preservation results in the upper part of horizontally lying ammonite shells being lost due to dissolution of its aragonitic material. Tanabe et al. (1984) stated that such preservation is characteristic for anoxic conditions prevailing on the sea-floor.

On the basis of foraminifers, Smoleń (2006) noted that during the sedimentation of the Lower Bathonian (Yeovilensis Zone) sediments of Kawodrza Górna (Leszczyński clay-pit), conditions at the sea-floor were either favourable (oxic) and stressful (dysoxic). Matyja et al. (2006b) concluded that on the basis of the benthic assemblages from the same clay-pit, living conditions at the sea-floor were worse than during sedimentation of the older uppermost Bajocian and Lower Bathonian deposits outcropping in the Sowa and Gliński clay-pits. Similar reducing conditions were interpreted for some intervals of the Middle-Upper Bathonian clays of the Gnaszyn clay-pit, on the basis of the benthic assemblages (Matyja et al. 2006c). Such a change in facies may have been related to the sea-level fluctuations, leading to the increased input of terrestrial matter and weakening of the offshore currents (due to sea-level rise), which resulted in the occurrence of dysoxic conditions. Sporadic, short storm episodes may have periodically improved oxygenation conditions at the sea-floor (Matyja et al. 2006c). It seems that short-lasting dysoxia may have occurred episodically and, very probably, only across a limited area of a sea-floor. The decay of an increased amount of organic matter may have produced local reducing conditions (Marynowski et al. 2007a).

The palaeoenvironmental conditions during the sedimentation of the Upper Bajocian–Bathonian clays of the Polish Jura area differ markedly from other Jurassic basins characterised by deposition of black shales. Whilst the redox conditions in the epicratonic Polish Basin during the Late Bajocian–Bathonian were favourable for the benthic biota, with only sporadic and possibly very localised dysoxia, other Jurassic basins witnessed periodic or prevailing anoxia. An example of such a basin from Poland is recorded in the Middle Jurassic deep-water sediments of the Pieniny Klippen Belt, situated in the southern Carpathian segment of Poland, which originally formed part of the northern Tethys Ocean. These sediments comprise dark-grey shaley marlstones (Harcygrund Shale Formation, Lower Bajocian) and black shales with spherosiderites (Skrzypny Shale Formation, Aalenian–Lower Bajocian), which were deposited under dysoxic to anoxic conditions in a partly restricted basin with a partly stratified water-column (see Tyszka 1994).

Anoxic conditions prevailed during the deposition of the Posidonia Shale (Lower Toarcian) in SW Germany, punctuated by short periods (weeks to years) of oxygenated bottom waters. Such conditions are thought to be controlled by sea-level changes and monsoon-influenced climate (Röhl et al. 2001). Intermittent water-column anoxia was also inferred for the fossiliferous Oxford Clay Formation (England) of Callovian age by Kenig et al. (2004). The authors based their assumption on the presence of isorenieratane, the biomarker produced by green sulphur bacteria (Chlorobiaceae). Such compounds are completely lacking in the Upper Bajocian–Bathonian clays of the Polish Jura (Zatoń and Marynowski 2004; Marynowski et al. 2007a). Fluctuating anoxic-dysoxic conditions with brief oxic episodes have also been inferred for the Late Jurassic Kimmeridge Clay (Dorset, England) by Wignall and Myers (1988). Later, on the basis of the pyrite framboid distributions, Wignall and Newton (1998) inferred euxinic conditions in the water-column during the deposition of the organic-rich Kimmeridgian shales.

Similar redox conditions to those recorded in the Polish Jura, were recently detected in the Callovian clays of Łuków (eastern Poland) and northern Lithuania. The Łuków clays are allochthonous for the region and were transported from the north (Baltic Sea area, see Olempska and Błaszyk 2001) during the Pleistocene glaciation. Using biomarker and pyrite framboid analyses of the Łuków carbonate concretions, Marynowski et al. (2008a, b) obtained similar results to those we present here for the Upper Bajocian–Bathonian clays of the Polish Jura. However, until detailed geochemical and microfacial investigations of other basins with Bajocian–Bathonian sediments are performed, strict comparisons of redox conditions prevailing in a time-equivalent environment will not be possible.

Conclusions

Geochemical (Th/U and U/Th ratios, ^{34}S, biomarkers) and petrographic (pyrite framboid distributions and microfacies analyses) investigations indicate that the depositional conditions during Middle Jurassic (latest Bajocian–Bathonian) in the Polish Basin were oxic and thus favourable for the organisms inhabiting the sea-floor. Localised anoxic microenvironments may have developed as a result of the decomposition of organic matter. Such conditions, however, mainly prevailed below the sediment–water interface.

This study suggests that the Th/U and U/Th ratios are the most reliable indices of redox conditions. The results of these two indices are supported by the sulphur stable isotopes, which point to open system conditions. DOP (degree of pyritisation), however, is characterised by widely scattered values, pointing to a variable redox regime. The majority of DOP values suggest dysoxic conditions, and it is suggested that the DOP values reflect the conditions within the sediment and chemistry of its pore waters, rather than the true redox conditions on the sea-floor. This is supported by the pyrite framboid distribution—their diameters are suggestive of their formation within a dysoxic sediment, beneath an oxygenated water-column. Biomarker analyses show that there are no compounds characteristic of anoxia and water-column stratification during sedimentation in the Middle Jurassic Polish Basin.

The integrated geochemical and microfacial investigation presented here, combined with the thorough organic matter analysis of Marynowski et al. (2007a), should serve as a reference point for further detailed palaeoecological investigations in such monotonous clay sequences as that of the Polish Jura.

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