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Gamma-ray spectrometry across the Upper Devonian basin succession at Kowala in the Holy Cross Mountains (Poland)

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ABSTRACT:

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The Upper Devonian sequence at Kowala in the Holy Cross Mountains was logged using gamma-ray spectrometry, for investigating the changes of oxygenation level in the Late Devonian basin. The Th/U ratio indicates that oxygen levels were low throughout the Late Frasnian interval, with low peaks during the Kellwasser Events showing anoxic conditions in the basin. The F-F boundary interval was also oxygen deficient, but there may have been a brief reoxygenation at the boundary itself. By the Famennian *crepida* Zone, the basin gradually began to reoxygenate, but in the *trachytera* Zone another anoxic event, the Annulata Event occurred, causing a bloom rather than extinction of specially adapted taxa such as *Guerichia*. Thus the gamma-ray spectrometry data suggests that basinal anoxia prevailed through much of the Late Frasnian. The F-F extinction might have been the result of prolonged stresses imposed on the ecosystem, particularly during the euxinic Upper Kellwasser Event.

Key words: Devonian, Kellwasser Event, Annulata Event, Frasnian-Famennian boundary, Holy Cross Mountains, Gamma-ray spectrometry.

INTRODUCTION

The Frasnian-Famennian event has been identified as one of five mass extinctions during the Phanerozoic (RAUP & SEPKOSKI 1982). It is characterized by sudden drop of ecosystems biodiversity and rapid reduction of marine biomass. It is estimated the crisis affected about 22% of the all marine families and about 57% of all marine genera (SEPKOSKI 1996). The F-F crisis is comparable in magnitude to the celebrated end-Cretaceous event (see HALLAM & WIGNALL 1997), and has become known as the ‘Kellwasser Event’, named after characteristic black shale and limestone horizons which occur in the Upper Frasnian of Germany (HOUSE 1985).

Numerous causal mechanisms have been proposed for the Kellwasser Event. These include catastrophic bolide impact (e.g. GOODFELLOW & *al.* 1989; see also RACKA 1999); oceanic anoxia and related sea-level fluctuations (BUGGISCH 1991, JOACHIMSKI & BUGGISCH 1993, BECKER & HOUSE 1994); cooling oceans (COPPER 1986); warming oceans (BRAND 1989); eutrophication (MURPHY & *al.* 2000); evolutionary steps amongst plants leading to anoxia (ALGEO & *al.* 1995); volcanism and tectonics (FLESSA 1986, see RACKI 1998 for review).

In the Devonian of the Holy Cross Mountains the phenomena related to the F-F mass extinction have previously been investigated by NARKIEWICZ & HOFFMAN (1989), RACKI & BALIŃSKI (1998), RACKI & *al*. (1989, 1999), Baliński & Racki (1999), Filipiak (1999) and Joachimski & *al*. (2001).

A biostratigraphically complete F-F section has been studied in the Kowala Quarry, in the Holy Cross Mountains (Text-fig. 1). On the basis of lithological variations and biostratigraphy both the Lower and Upper Kellwasser Events have been identified (see Joachimski & *al*. 2001). In the course of present study, gamma-ray spectrometry was conducted throughout the sequence and provides additional indicators for oceanic oxygenation levels during this major biotic crisis interval.

FACIES BACKGROUND

During the Devonian the present area of the Holy Cross Mountains formed a part of the belt of tropical to subtropical pericontinental basins along the southern margin of Laurussia (Narkiewicz 1988, Racki 1993).

At the beginning of the Late Devonian, the Early Frasnian transgression terminated the carbonate platform built mostly of coral-stromatoporoid biostromes. Biogenic *in situ* sedimentation survived in isolated areas, of which only the most prominent became the sites of development of major reef complexes such as the Dyminy Reef. The cause of termination of the Middle Devonian platform were low-oxygen conditions along with drowning (Narkiewicz 1988).

In the Late Frasnian a strong transgressive pulse resulted in the drowning of the remaining reef complexes which has been developing on the carbonate platform since the Middle Devonian. Only limited shallow-water carbonate sedimentation continued in the central areas of the former Dyminy Reef. The Late Frasnian event marks also the onset of a development of carbonate deposits, including pelagic as well as deeper-water benthic fauna, traditionally called Manticoceras Limestones (Szulczewski 1995b).

Such sediments were generally deposited on submarine elevations built of drowned organic build-ups, as well as within shelf-basinal facies. The latter represented the most widespread depositional system during the Late Frasnian to Famennian. However, over several topographic highs, a submarine nondeposition prevailed, locally remaining during the Late Frasnian, a considerable part of the Famennian or even into the Tournaisian (Szulczewski 1995b).

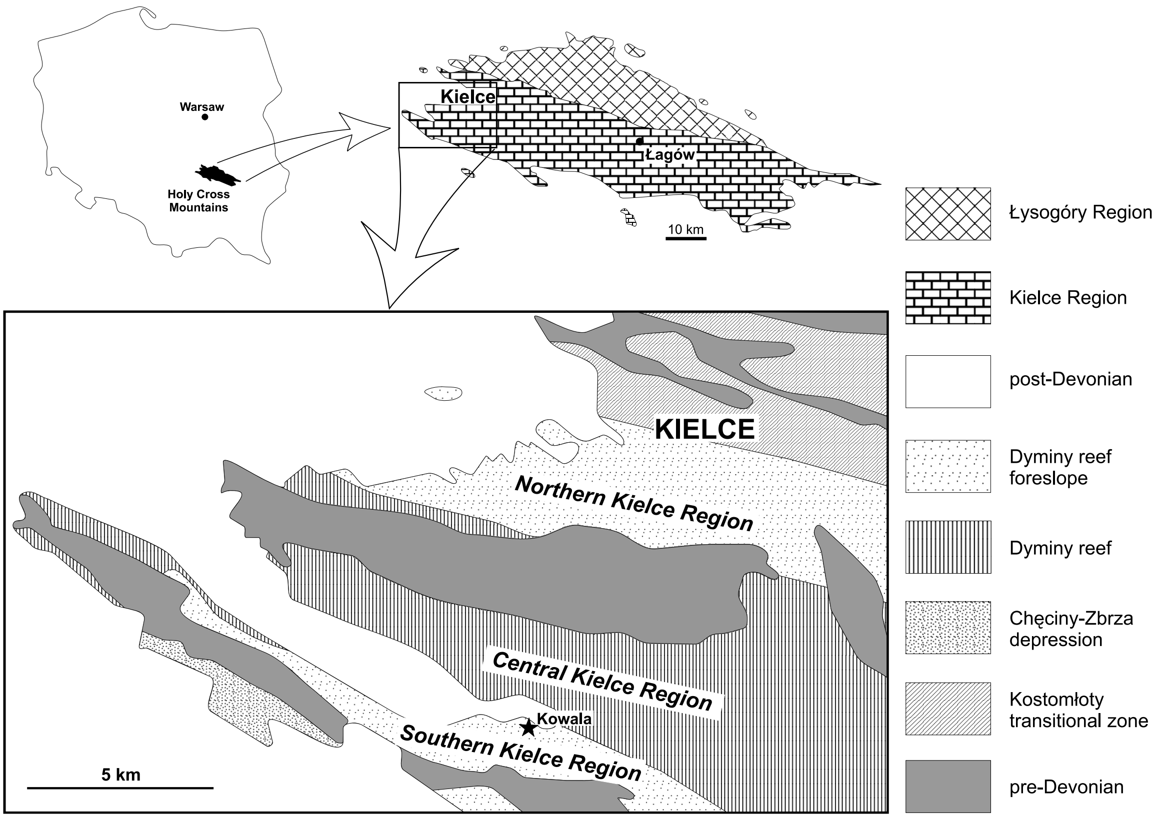

Fig. 1. Paleogeographic map of western part of the Kielce Region in the Holy Cross Mountains (after Racki, 1993, fig. 2)

In the Famennian, almost the entire area comprises dark, clayey-calcareous bituminous deposits with abundant early-diagenetic pyrite. The depositional environment is interpreted as poorly oxygenated (BERKOWSKI 1991, SZULCZEWSKI 1996), well below a photic zone and storm wave-base (NARKIEWICZ 1988, MARYNOWSKI & *al.* 2000), with progressing transformation from anaerobic to well oxygenated, accompanied by shallowing up (SZULCZEWSKI 1992, 1995b).

The Kowala Quarry lies in the northern part of the Southern (Kielce) region (Text-fig. 1) and exposes a Frasnian-Famennian sequence of typical off-reef hemipelagic deposits (see NARKIEWICZ 1988, RACKI & *al.* 2002), consisting of micrites interbedded with marls and shales.

METHODS

A sequence in excess of 200 m was logged in detail in 3 trenches in the quarry floor, and above the northern quarry wall, from the *hassi-jamiae* conodont Zone to the *trachytera*-Lower *expansa* Zone (according to SZULCZEWSKI 1995a, 1996). The Kowala section was analysed at regular intervals using a field portable gamma-ray spectrometer Envispec GR 320, which assays concentrations of potassium (K), thorium (Th) and uranium (U). This provides a measure of redox conditions because of the enrichment of U in anoxic conditions (WIGNALL & MYERS 1988). The device samples a dish-shaped area approximately 1.5 m in diameter, so readings were taken at approximately 1m intervals. Each readout of U, Th and K took three minutes.

The Th/U ratio can be used to distinguish between anoxic, dysaerobic, and oxic depositional environments. In marine sediments, K and Th are closely correlated because both are present mainly in the detrital clay fraction. U also occurs in the detrital clay fraction; however, unlike Th, it is also carried partly in solution as uranyl carbonate complexes (WIGNALL & MYERS 1988). Under reducing conditions, U^{6+} is reduced to U^{5+} or U^{4+}. These reduced forms are not stable in solution and precipitate in the sediment as authigenic uranium associated with organic matter. Th is not reduced and is entirely detrital.

The U is fixed at the sediment-water interface under reducing conditions in the presence of a sorbent, which is usually organic matter or phosphate. Therefore, sediments enriched in authigenic U tend to be deposited under anoxic conditions that allow both large amounts of organic matter to accumulate and U to be fixed (WIGNALL & MYERS 1988). Thus under anoxic conditions, there is elevated U and the Th/U ratio decreases.

The values for the Th/U ratio which indicate anoxia or dysaerobia are variable, depending on factors such as the clastic content, and sedimentation rate. Detrital sediments generally have higher Th contents than carbonates with the result that the Th/U ratio of anoxic shales is less than 3 (MYERS & WIGNALL 1987) but for carbonates the ratio is typically less than 1. Gamma ray spectrometry is not considered a definitive indicator of palaeoxygen levels and it is necessary to back up any interpretation of the data with another technique, such as pyrite framboid analysis. Nevertheless, gamma ray spectrometry provides a useful preliminary indicator of trends in oxygen levels.

The sequence has already been studied for trace elements, kerogen composition and carbon isotopes (JOACHIMSKI & *al.* 2001, see also RACKI & *al.* 2002), as well as for biomarkers (MARYNOWSKI & *al.* 2000), but has not been studied using gamma ray spectrometry until now.

RESULTS

Lithological description

The section in Kowala Quarry have been previously studied by SZULCZEWSKI (1968, 1971, 1995a, 1996), BERKOWSKI (1991, 1993, 2002), RACKI (1990), RACKI & BIERNAT (1986), RACKI & *al.* (1999, 2002) and JOACHIMSKI & *al.* (2001). SZULCZEWSKI (1971) and BERKOWSKI (1990) (*vide* SZULCZEWSKI 1995a, 1996) divided the sequence into units A to L. Later RACKI (1996) divided the unit H into more precise units H-1 to H-4 (see also RACKI & BALIŃSKI 1998). During the present study the sequence was logged and divided into 14 units on the basis of lithology, to provide a more detailed sub-division.

The studied section begins with Unit 1, corresponding in part to unit H-1 of RACKI (1993), in the *hassi-jamiae* Zone (Text-fig. 2) which comprises interbedded massive micrites, laminated shales, and calcarenites, with occasional marls. The colour is generally light to medium grey. Above this Unit 2 (Lower to Upper *rhenana* Zone) is similar, although the proportion of micrites increases, and many of the micrites are finely laminated.

Unit 3 sees a distinct facies change to nodular and bioturbated grey micrites interbedded with laminated marls (Text-fig. 2). It also contains a distinctive 14 cm thick finely laminated dark shale, which is interpreted as the Lower Kellwasser Event. Ostracods are common throughout units 1-3 but decline in importance in unit 4. *Amphipora* is also common at this level. The bulk of the remaining fauna is comprised of tentaculitoids (occasionally in abundance), rugosan corals, crinoids, *Manticoceras* ammonoids, *Lingula* and other brachiopods and bivalves. Units 2 and 3 together correspond to unit H-2 (sensu RACKI 1996, see also RACKI & BALIŃSKI 1998).

Unit 4 contains the F-F boundary and comprises mostly finely laminated medium to dark grey micrites with some thin shales, calcarenites and marls. Certain beds below the F-F boundary contain abundant tentaculitoids. Approximately 2.5 m beneath the boundary there is a dark, finely laminated shale, which is interpreted as the Upper Kellwasser Event. The boundary itself is characterised by two thin cherts, with a crinoidal hash immediately above. Across the boundary the lithology is much the same, but the fauna becomes impoverished, and dominated by an *Orbiculoidea* assemblage including *Lingula, Manticoceras,* and extremely well preserved algal straps. Tentaculitoids and *Amphipora* are not observed above the boundary. Unit 4 is roughly equivalent to unit H-3 (sensu RACKI 1996, see also RACKI & BALIŃSKI 1998, RACKI & *al*. 2002).

The Lower Famennian rocks of Unit 5 are similar to those of Unit 4, comprising interbedded finely laminated shales and medium to dark grey micrites, although the proportion of shales increases slightly (Text-fig. 2). Bioturbation is still absent. The fauna remains dominated by *Orbiculoidea* and contains numerous *Pterochaenia*.

Unit 6 and 7 comprise interbedded micrites and shales (Text-fig. 2). Although some of the micrites remain laminated, the majority are bioturbated, indicating a return of benthos. Several micrites are pyritic. The fauna remains poor, but there are occurrences of ostracods in these units.

Units 8 and 9 are similar, being made up of decimetric alterations of laminated shales and medium grey, pyritic, bioturbated micrites and also some sparites. Units 5-9 are an equivalent of unit H-4 (sensu RACKI 1996).

Above this, Unit 10 sees a distinct facies change to a nodular shales with interbeds of micrite. Above this are a few metres of interbedded shales and laminated micrites (Unit 11). These units correspond to unit I of SZULCZEWSKI (1995a, 1996).

The sequence continues with approximately 50 m of bioturbated, pale grey micaceous mudstones with micrite nodules (Unit 12 = unit J sensu SZULCZEWSKI 1995a, 1996). The fauna is still limited, but a new species of bivalve becomes common.

Unit 13 comprises massive to finely laminated shales interbedded with massive or nodular micrite interbeds and concretion bands. There are several black shales, including a 60 cm thick, very finely laminated shale covered in an opportunistic bloom of *Guerichia* and clymeniids. This is tentatively interpreted as an equivalent to the Annulata Event which left similar beds during the *trachytera* Zone elsewhere in the world (WALLISER 1996). The fauna in Unit 13 is dominated throughout by *Guerichia*, which dominate in Famennian macrofaunal assemblage – especialy in *trachytera* and *expansa* Zones (ŻAKOWA & RADLICZ 1990), although brachiopods (BIERNAT & RACKI 1986), blind trilobites (BERKOWSKI 1991), ostracods, gastropods, cephalopods and rugosan corals are common in the micrites (see BERKOWSKI 1993). Above this, interbedded shales and micrites continue through Unit 14, but these are mostly obscured by a road (Text-fig. 2). Units 13 and 14 correspond to Unit K of BERKOWSKI (1991, see also SZULCZEWSKI 1995a, 1996).

Gamma ray spectrometry

The sequence has been studied in detail with gamma-ray spectrometry, and the results are presented graphically in Text-fig. 2. The ratio is low (generally <1) for most of the Upper Frasnian. The two Kellwasser Events exhibit a negative shift in the ratio, to 0.90 from a previous value of 1.47 (LKE), and 0.82 from 1.57 (UKE). The ratio around the F-F boundary remains low, although there is a brief positive pulse at the boundary itself. The ratio is low during the Lower Famennian (e.g. 0.28 in unit 4, a shale 2m above the boundary) (Text-fig. 2). Throughout units 5 to 8 the Th/U ratio rises gradually to values generally between 2 and 4.

The ratio in Unit 13 drops somewhat, but levels remain in general around 2, except for the postulated Annulata Event, which records a dramatic negative shift, from a value of 1.66 immediately below to just 0.30. Above this shale the ratio rises rapidly once again to >2.

The overall trend is clear (Text-fig. 2): the ratio is low in the Upper Frasnian and around the boundary, with distinct low points during each of the Kellwasser Events. The ratio remained low during the Lower Famennian before gradually rising through the Upper *triangularis* and *crepida* Zones. A further dramatic negative shift occurs during the Annulata Event, above which the ratio rises rapidly.

Interpretation

The trend in the Th/U ratio shows the oxygenation history for the F-F section at Kowala. The ratio indicates that oxygen levels were low throughout the Late Frasnian interval, with minimums during the Kellwasser Events, which were true anoxic events. This is supported by the presence of abundant, tiny pyrite framboids

(< 4 μm mean diameter) (Text-fig. 2) which suggest that conditions became euxinic during each event (WIGNALL & NEWTON 1998). Whereas the majority of results from the boundary interval suggest sustained anoxia, several beds show an increase in Th/U ratio to more normal values at the F-F boundary. This hints at a temporary reoxygenation at the boundary (RACKI & *al*. 2002). However, it may be the result of intense bleaching of the boundary interval by oxidising groundwaters, causing discrepancies in trace metal contents. The lithology remains finely laminated, and unoxidised patches of the boundary beds have been found to contain pyrite framboids. Thus it is possible that anoxia prevailed throughout this interval. Facies analysis of the Late Frasnian beds supports low oxygen levels, being mostly comprised of medium to dark grey micrites and laminated shales, and almost entirely devoid of bioturbation.

The Th/U ratio increases gradually through the Upper *triangularis* and *crepida* Zones, indicating a slow, progressive reoxygenation of the basin. The presence of tiny pyrite framboids indicates, that certain periods during the Upper *triangularis* Zone remained intensely anoxic, and these levels again correlate to low points in the Th/U ratio. Thus it appears that while conditions were gradually improving, there were still periods of intense anoxia in the Early Famennian.

Unit 6 sees a return of bioturbation and the Th/U ratio stabilises at values of 2 to 3, suggesting that by the *crepida* Zone benthic oxygen levels had substantially improved. This trend continues for some time with grey nodular beds being common up to the *trachytera* Zone (Unit 13). The presence of nodules and pale-medium grey colour further indicates increased oxygen levels. The post-Kellwasser Event fauna remains impoverished throughout this period, with dysoxia tolerant forms dominating. It appears to have taken a considerable time for the recovery of fauna, even once oxygen levels improved.

Oxygen levels appear to have fluctuated rapidly during *trachytera* Zone (Unit 13), as evidenced by interbedded laminated dark shales and grey micrites and the frequent occurrence of *Guerichia* in the shales. BERKOWSKI (1993) stated that absence of true benthos and presence of organic matter in the black shales confirm anoxic conditions on the basin floor. Fauna from the micrites is typical of dysaerobic environments. Gamma ray spectrometry also testifies to intense anoxia during the Annulata Event. Above this level the oxygen levels increased rapidly back to pre-Annulata levels, and the fauna seems unaffected by this anoxic event.

The fauna is poor throughout the entire sequence, due to the low oxygen levels throughout the Late Frasnian. In the Frasnian, tentaculitoids and allochtonous *Amphipora* are common, but they do not occur above the F-F boundary. Entomozoacean ostracods are also common in the Late Frasnian, and these are thought to be well suited to oxygen poor settings (CASIER & LETHIERS 1998). However these are not observed above the boundary until the top of the Upper *triangularis* Zone, suggesting that oxygen levels became too low even for these. The relatively varied Frasnian fauna is replaced by an impoverished *Orbiculoidea* assemblage in the Famennian comprising dysoxia tolerant forms. The fauna recovers only very slowly during the Famennian, as the basin gradually reoxygenated. The faunal crisis appears to be related to prevailing oxygen poor conditions during the Late Frasnian and two anoxic pulses during the Kellwasser Events.

CONCLUSION

Gamma ray spectrometry together with facies analysis of the complete F-F section at Kowala Quarry suggests, that oceanic oxygen levels were very low during the Late Frasnian. The Lower and Upper Kellwasser Events have been identified, and these correlate to low points in the Th/U ratio. These were true anoxic events. MARYNOWSKI & *al*. (2000) found in deposits from Kowala aryl isoprenoids as the product of green-sulphur bacteria (*Chlorobiaceae*), point to the presence of anoxic conditions even in the photic zone. This has been confirmed by JOACHIMSKI & *al*. (2001), who found in these deposits biomarkers diagnostic for such a kind of bacteria.

The boundary interval was also oxygen deficient, but there is conflicting evidence for a possible brief reoxygenation at the boundary itself. Oxygen levels remained low into the Famennian but the basin gradually began to reoxygenate by the *crepida* Zone. Following recovery in oxygen levels a further brief anoxic event, the Annulata Event is marked by a dramatic negative shift in the Th/U ratio in the *trachytera* Zone, but it did not cause extraordinary extinctions or originations, but rather a blooming of apparently specially adapted taxa (e.g. *Guerichia*, clymeniids, entomozoan ostracods). It is considered the event was caused by a short-term flooding of epeiric seas with oxygen depleted waters due to a rise of the oceanic anoxic layer (WALLISER 1996).

A variable Frasnian fauna is reduced to an impoverished *Orbiculoidea* assemblage in the Famennian. Tentaculitoids and *Amphipora* become extinct. This faunal crisis might be linked to the spread of persistent basinal dysoxia during the Late Frasnian and in particular the euxinic pulses during the two Kellwasser Events. This lends support to a model for the cause of the F-F extinction incorporating anoxia as the proximate kill mechanism.

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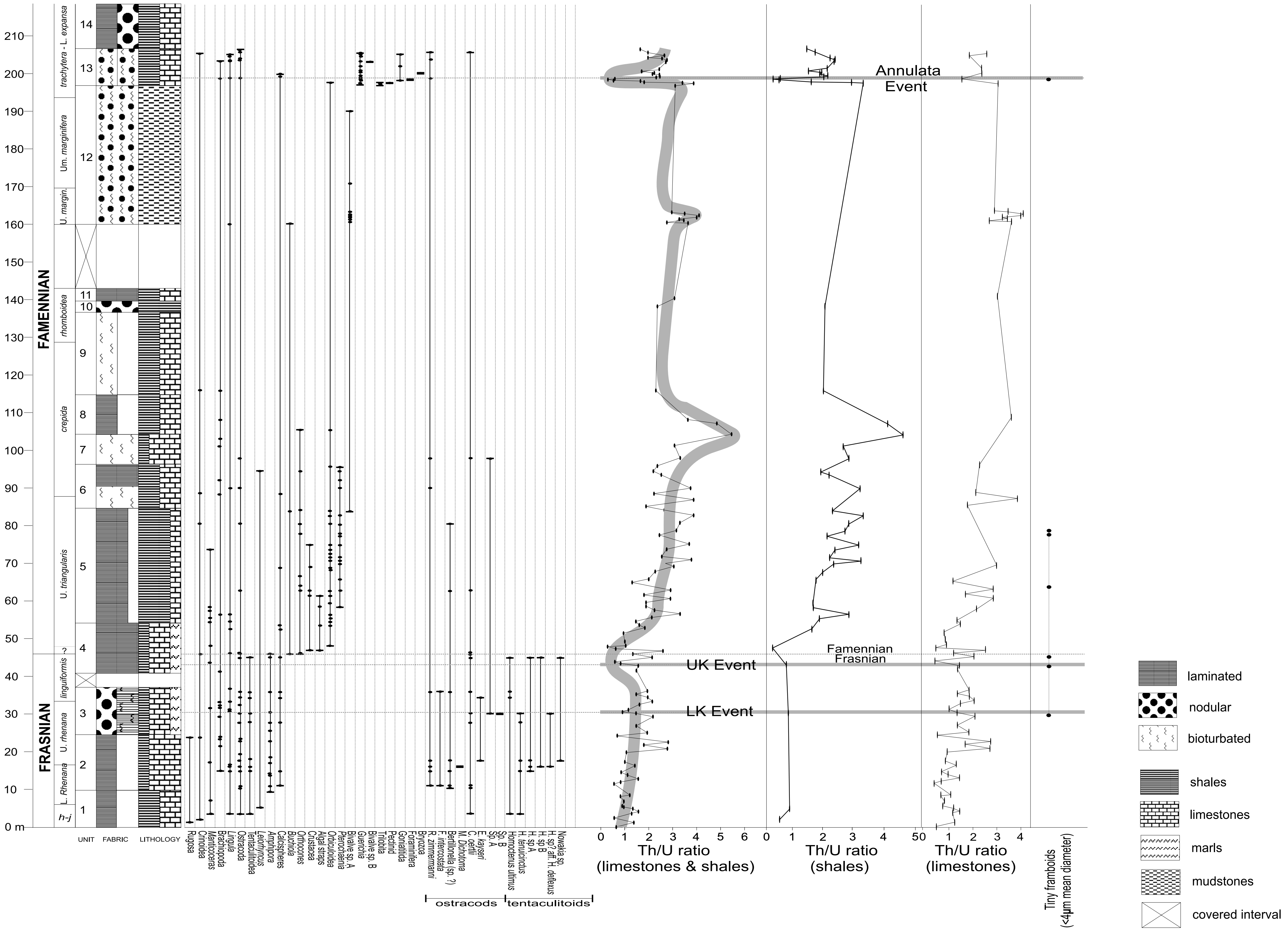

Fig. 2. Summary log of Kowala Quarry section, including conodont zonation based on SZULCZEWSKI (1995a, 1996) and RACKI & BALIŃSKI (1998), faunal ranges of most common fossils, the Th/U ratio and pyrite framboids distribution