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ADDENDUM TO THE CHEMOSTRATIGRAPHY OF THE UPPERMOST CALLOVIAN–MIDDLE OXFORDIAN INTERVAL OF THE TETHYAN FATRICUM DOMAIN (TATRA MTS, KRÍŽNA NAPPE, SOUTHERN POLAND)

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Abstract: New isotope ($\delta^{13}C$, $\delta^{18}O$ of bulk carbonates) and carbonate content data from the uppermost Callovian–middle Oxfordian radiolarites of the Fatricum domain belonging to the north-western segment of the Tethys are added to previously published data. The new data supplement the Długa Valley section, the most nearly complete Bajocian–lower Tithonian section of the Krížna Nappe in the Tatra Mts. The uppermost Callovian and Lower Oxfordian bulk $\delta^{13}C$ values (from 3.1 to 3.3‰) remain nearly constant with highly positive values. Therefore, the positive excursion identified in bulk carbonate $\delta^{13}C$ values is interpreted as a record of the upper Callovian–middle Oxfordian global phenomenon. In this interval, a significant increase of $CaCO_3$ content is recorded, which accompanies facies change from ribbon radiolarites with siliceous shale partings to calcareous radiolarites with rare shale intercalations. The abrupt $CaCO_3$ increase may reflect a turning point in Early Oxfordian carbonate production and recovery of the marine carbonate factory.

Key words: carbon and oxygen isotopes, carbonate content, radiolarites, Western Carpathians, Alpine Tethys.

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INTRODUCTION

The bulk carbon-isotope chemostratigraphy of Jurassic sections is of crucial importance in sequences impoverished in, or completely barren of index fossil remains. Relatively numerous carbon isotope data derive from Tethyan and peri-Tethyan Middle–Upper Jurassic deposits (e.g., Bartolini *et al*., 1999; Jenkyns *et al*., 2002; O'Dogherty *et al*., 2018), with one exception for the Callovian–middle Oxfordian interval, where such data are scarce and display some discrepancy in temporal trend (e.g., Bartolini *et al*., 1999; Cecca *et al*., 2001; Padden *et al*., 2002; Jach *et al*., 2014; Wierzbowski, 2015; Arabas, 2016; O'Dogherty *et al*., 2018).

Bulk carbon-isotope data from Tethyan Jurassic limestones and radiolarites were published by Bartolini *et al*. (1999) and Jach *et al*. (2014). The latter paper deals with the carbon isotope record of pelagic sequences in seven sections of the Fatricum domain. The Długa Valley is the most nearly complete of the studied sections; it has a sufficient biostratigraphical frame and a set of geochemical data. Nonetheless, the section has a tectonic gap covering the uppermost part of the ribbon radiolarites and the lower part of the carbonate radiolarites (see remarks in Jach and Reháková, 2019, p. 5 and fig. 4 therein). The missing interval is of crucial importance because it comprises a turning point in pelagic carbonate production, starting from a worldwide crisis toward a recovery of the carbonate factory. The new field study in an adjacent outcrop has revealed a new supplementary section. This section was sampled and laboratory analyses allowed supplementation of the previously published dataset with carbon isotopic and $CaCO_3$ content data from the Callovian-Oxfordian boundary interval. The composite Długa Valley section is more than 76 m thick on the basis of two exposures. It provides a good example of the Bajocian–Tithonian pelagic geochemical record.

REGIONAL SETTING

The newly examined section is located on the northern slope of the Długa Valley in the Western Tatra Mountains (Central Western Carpathians), in southern Poland (Fig. 1). The section belongs to the Krížna Nappe and represents the

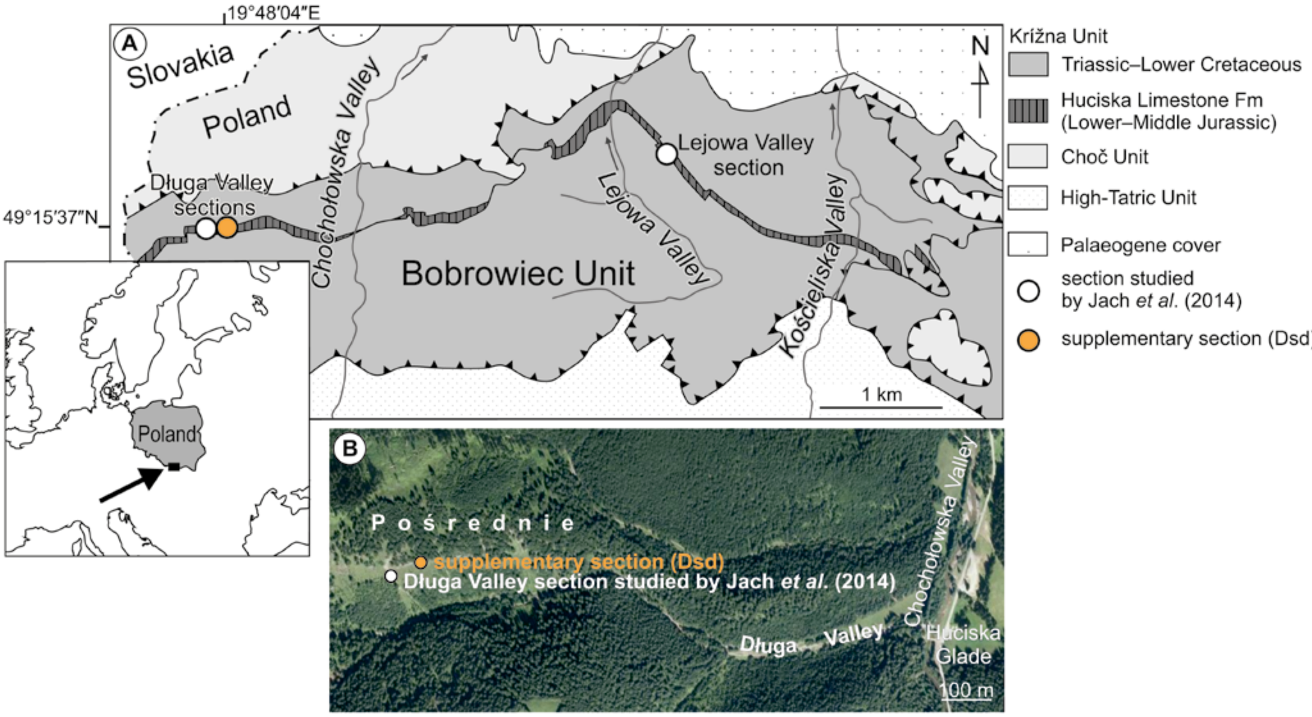

Fig. 1. Location of the studied section. **A.** Tectonic sketch map of the Tatra Mountains (after Bac-Moszaszwili *et al*., 1979) showing locations of the Długa Valley section (Jach *et al*., 2014) and newly studied supplementary section. **B.** Detailed location of previously studied Długa Valley section and newly studied one (Dsd), aerial view (from © 2018 Google Earth, Image © CNES/Airbus).

western Tethys Fatricum Domain, which was one of the domains situated between the Alpine Tethys (also named Alpine Atlantic Ocean) to the north and the Meliata Ocean to the south (Schmid *et al*., 2008). As a consequence, the succession studied shows a strong similarity to the Jurassic of other Tethyan basins.

During the Middle–Late Jurassic, the Fatricum Domain was located between the uplifted Tatricum Domain to the north and the Veporicum Domain to the south. The Jurassic deposits of the Krížna Nappe in the Tatra Mountains represent almost continuous deep-marine successions of Zliechov type (Michalík *et al.*, 2007; Jach and Reháková, 2019). The deposition of carbonate sediments terminated with the onset of uniform radiolarite sedimentation during Late Bathonian (Polák *et al*., 1998; Jach *et al*., 2014). The complete recovery of carbonate sedimentation took place during the Kimmeridgian. In the interval studied, two radiolarite lithotypes are distinguished: ribbon radiolarites of Upper Bathonian–lower Oxfordian, which consist of alternating chert beds and shale partings, and overlying calcareous radiolarites of the middle Oxfordian–lowermost Upper Kimmeridgian (Jach *et al*., 2014).

SITE DESCRIPTION

The supplementary section studied (GPS coordinates 49°15′37″N, 19°48′04″E) is located 60 m to the north-east of the previously studied Długa Valley section (Jach *et al*., 2014). It crops out at the foot of a steep rock cliff, located in the Długa Valley along the southern slope of the Pośrednie ridge. The 4-m-long section fills the gap in the previously published Długa Valley section (Jach *et al*., 2014; Figs 2–4) and comprises ribbon radiolarites (2 m), covered by calcareous radiolarites (2 m; Figs 3, 4).

MATERIAL AND METHODS

The new section was sampled bed by bed. The rock samples are stored at the Institute of Geological Sciences, Jagiellonian University in Kraków.

Oxygen and carbon isotope compositions of 11 bulk sediment samples was measured. The section was sampled at approximately 0.36 m intervals. Only homogenous micritic samples were analysed. Carbon and oxygen stable isotope compositions ($\delta^{13}C$ and $\delta^{18}O$) were analysed in the Stable Isotope Laboratory of the Institute of Geological Sciences, Polish Academy of Sciences, in Warsaw. Powdered samples were produced using a diamond-tipped drill. The powder was reacted with 100% phosphoric acid (density 1.94 g/cm^3) at 70°C, using the Kiel IV online carbonate preparation device, connected to the Thermo-Finnigan Delta+ mass spectrometer. The quality of analysis was controlled by measurements of the international standard NBS-19 with each sample series (3–5 NBS-19 measurements per sample series). All values are reported as per mil relative to V-PDB. Reproducibility was checked on the basis of long-term repeatability of NBS19 analysis and occurred better ±0.05 for $\delta^{13}C$ and ±0.09‰ for $\delta^{18}O$, 1σ.

A plot of the $\delta^{18}O$ and $\delta^{13}C$ values and Pearson correlation coefficient was made and calculated to check the

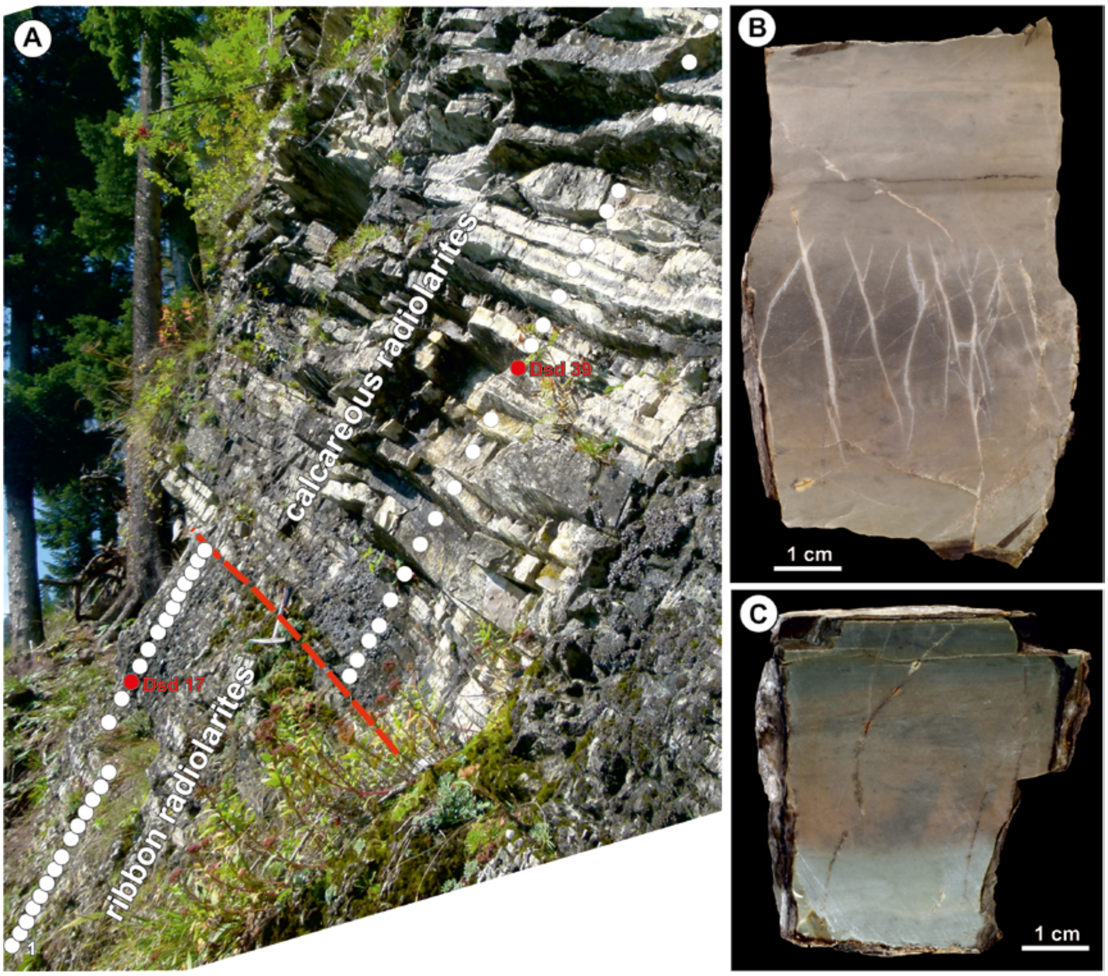

Fig. 2. Supplementary Długa Valley section (Dsd) in the Western Tatra Mountains. **A.** General view; white dots indicate studied rock samples; red dots indicate samples presented on the photographs (B and C), red dashed line divides two described facies. **B.** Calcareous radiolarites. Sample Dsd 39, polished surface. **C.** Ribbon radiolarites. Sample Dsd 17, polished surface.

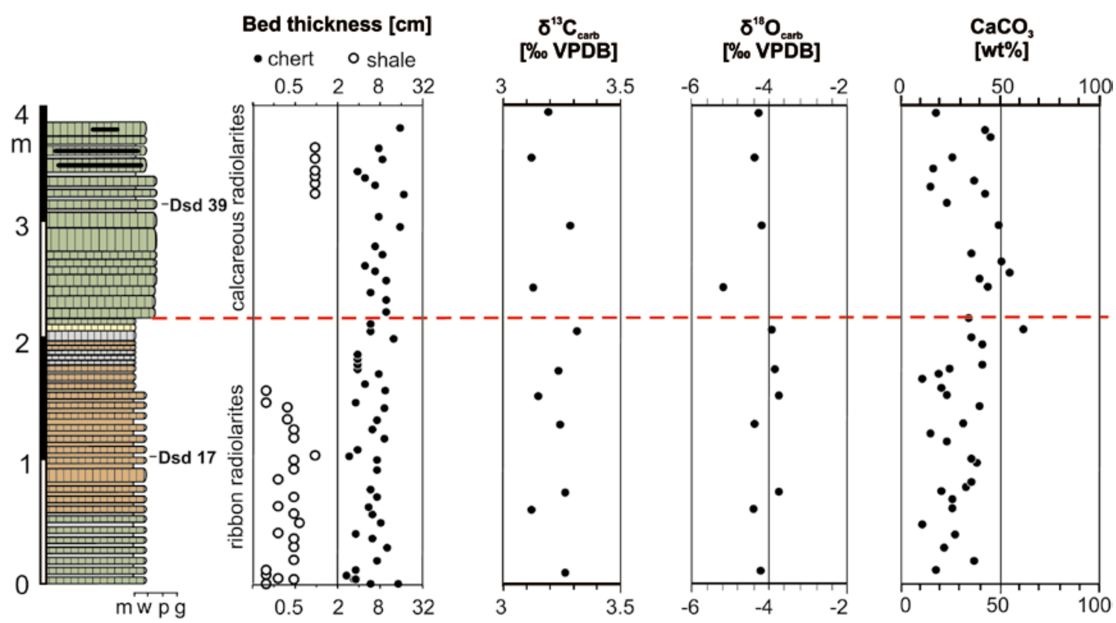

Fig. 3. Geochemical record of the supplementary Długa Valley section (Dsd; bed thickness, carbon and oxygen isotope values and $CaCO_3$ content of the rocks). See Figure 4 for legend.

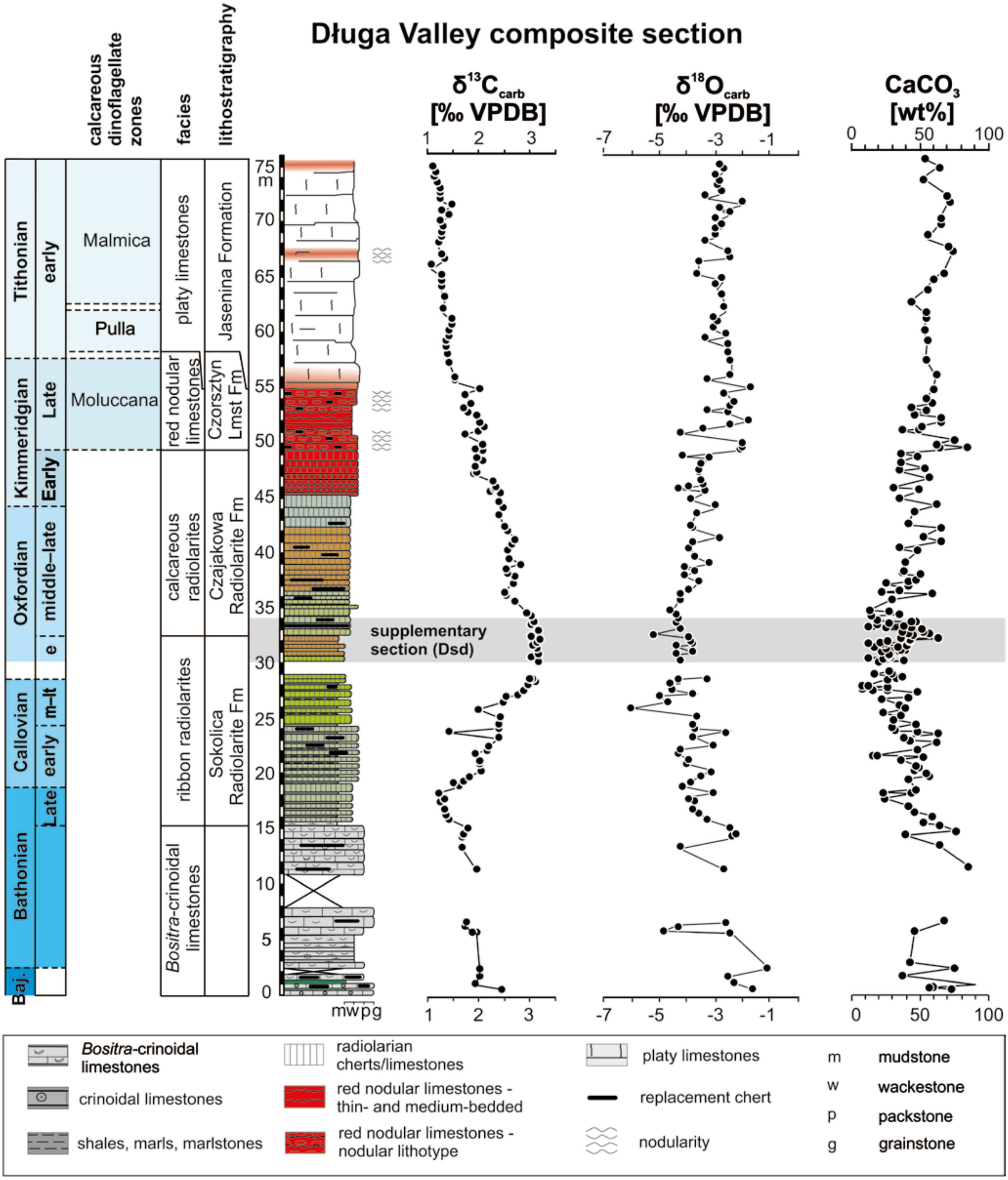

Fig. 4. Composite section (Jach *et al.*, 2014, supplemented) of the Długa Valley section in the Western Tatra Mountains. Lithology, stratigraphy, carbon and oxygen isotope ratios, and $CaCO_3$ content of the rocks (after Jach *et al.*, 2014), modified and supplemented. Log colour reflects the natural rock hues.

possible effect of diagenesis on the stable isotope composition. Newly obtained and previously published data from the Długa Valley section were used in calculations (Jach *et al.*, 2014).

The calcium carbonate content of 41 rock samples was analysed by using a calcimetre by Eijkelkamp, according to the method of Scheibler. The analysis of $CaCO_3$ content was conducted at the Institute of Geological Sciences of the Jagiellonian University in Kraków.

RESULTS

A correlation between the supplementary section and the Długa Valley section, previously studied by Jach *et al.* (2014), was established. On the basis of thickness, colouration, absence of shale partings, occurrence of secondary chert and similar $CaCO_3$ content, the uppermost layers of supplementary section with layers, found above the tectonic gap in the previously published Długa Valley section, were correlated. The position of the bottom of the new section is more problematic. It is estimated that about 1 m of the section is still missing on the basis of detailed comparison of the Długa Valley sections with the Lejowa Valley section, located a distance of ca 3 km away (see Jach and Reháková, 2019).

Facies

The interval studied comprises the transition from the ribbon to the calcareous radiolarites. The former (lower 2 m of the section; Fig. 3) are assigned to the Sokolica Radiolarite Formation (Lefeld *et al.*, 1985) and consist of thin-bedded (3 to 10 cm thick) greenish, grey and reddish-grey radiolarian cherts, alternating with shale partings (shales 0.5 cm thick) of similar colouration. Radiolarian mudstone and wackestone are the dominant microfacies. The matrix is recrystallized and locally silicified. In the section, the $CaCO_3$ content of the chert beds ranges from 12 to 39 wt% and averages 27 wt%.

The calcareous radiolarites (upper 2 m of the section; Fig. 3) formally belong to the Czajakowa Radiolarite Formation (Lefeld *et al.*, 1985). They comprise thin- to medium-bedded (6 to 18 cm thick) greenish and grey radiolarian cherts with scarce shale partings, up to 1 cm thick. The partings occur exclusively in the interval between 3.2 m to 3.6 m from the base of the studied section. Radiolarian wackestone constitutes the predominant microfacies. Beside radiolarian tests, bioclasts such as rare fragments of filaments (*Bositra*-like thin-shelled bivalve) and echinoderms occur. In the upper part of the section, the $CaCO_3$ content of chert beds ranges from 16 to 62 wt% and averages 40 wt%.

The facies change from ribbon to calcareous radiolarites is abrupt and manifested by an increase in carbonate content, a thickening-upward trend, with the disappearance of shale partings and a colour change from green, grey-reddish to light green and variegated (Figs 2 and 3).

Values of $\delta^{13}C$ and $\delta^{18}O$

In the section studied, the values of $\delta^{13}C$ range from 3.1‰ to 3.3‰ and remain constantly high. The values of the $\delta^{18}O$ range from –5.2‰ to –3.8‰ and show a discernible, negative, temporal trend.

The oxygen and carbon isotope data are moderately correlated ($R^2 = 0.25$), indicating a limited diagenetic influence on the isotopic records (see Marshall, 1992). Therefore, the $\delta^{13}C$ values seem to record the primary signal and can be used for chemostratigraphic purposes. Composite bulk sediment records of $\delta^{13}C$ and $\delta^{18}O$ for the Bajocian–lower Tithonian were constructed, using previously published data from the Długa Valley section (Jach *et al.*, 2014) and the new isotope data (Figs 3, 4).

DISCUSSION

The pelagic sequence investigated rarely contains fossils suitable for isotope analyses. Therefore, the isotope composition of the bulk fine-grained carbonate samples was studied. The measured values of the $\delta^{13}C$ in bulk sediment samples are similar to published Jurassic carbon isotope data (see Katz *et al.*, 2005). It is worth noting that the Late Jurassic was a period of important evolutionary changes in oceanic carbonate productivity. This was due to the Kuenen Event (Roth, 1989), associated with a shift in carbonate source from the benthic environment to the pelagic water column, with calcareous plankton production (Roth, 1989; Suchéras-Marx *et al.*, 2019). Since the deposits studied indicate pelagic conditions, with a limited influence of shallow environments, the variation in $\delta^{13}C$ values should reflect mainly the impact of the biological carbon pump on the isotope composition of carbonates deposited (Coimbra *et al.*, 2014).

The analysed $\delta^{13}C$ values are constantly higher than ca. 3.1‰ and fit well into the pronounced positive excursion, visible in the composite $\delta^{13}C$ curve (Fig. 4; Jach *et al.*, 2014). The recorded, long-lasting, positive excursion in the upper Callovian–middle Oxfordian from the supplementary Długa Valley section correlates well with general trends published (Bartolini *et al.*, 1999; Katz *et al.*, 2005). It is worth mentioning that the same interval in the Ždiarska Vidla section (Płaczliwa Skała in Polish) in the Belianske Tatra Mts shows a similar pattern, but with small positive excursions (Jach *et al.*, 2014).

Generally, a few published $\delta^{13}C$ curves for the upper Callovian–middle Oxfordian interval differ in trend. The $\delta^{13}C$ curves obtained for the Boreal/Subboreal province or from terrestrial organic matter show a long-lasting, positive excursion (Pearce *et al.*, 2005; Nunn *et al.*, 2009; Wierzbowski *et al.*, 2013), whereas those for the Tethyan and peri-Tetyan areas in bulk-rock or belemnite data mostly show a prominent, positive excursion with one or two positive shifts (Bartolini *et al.*, 1999; Cecca *et al.*, 2001; Katz *et al.*, 2005; Pearce *et al.*, 2005; Lavastre *et al.*, 2011; Wierzbowski, 2015; Arabas, 2016; Al-Mojel *et al.*, 2018, Carmeille *et al.*, 2018). Some discrepancies in the $\delta^{13}C$ patterns may result from a too low sampling resolution, the presence of sampling gaps, stratigraphic hiatuses (e.g., Carmeille *et al.*, 2018) or the influence of local factors, such as upwelling (Wierzbowski, 2015). The more positive late Callovian–early Oxfordian $\delta^{13}C$ values may reflect increasingly eutrophic conditions and enhanced burial of organic matter during a global sea-level rise (e.g., Louis-Schmid *et al.*, 2007; Wierzbowski *et al.*, 2009). Nutrient-rich conditions brought a carbonate production crisis in the entire Tethyan region and the widespread occurrence of radiolarites (Baumgartner, 2013).

The uppermost Callovian–middle Oxfordian interval includes the early Oxfordian turning point of carbonate productivity of the oceans, for which a recovery of the marine carbonate factory is observed. The section studied records changes from thin-bedded ribbon radiolarites with shale partings to thin- to medium-bedded micritic carbonate enriched radiolarites, which seems to reflect a turning point in the recovery of the marine carbonate factory (Louis-Schmid *et al*., 2007). The thickening-upward trend most probably indicates a change in accumulation rate, due to increased carbonate production and a higher contribution of the total $CaCO_3$ content in the sediment.

The newly studied supplementary section contains a clearly visible boundary between ribbon radiolarites and overlying calcareous radiolarites and documents the existence of two different radiolarite units. They were formally defined by Lefeld *et al*. (1985) as the Sokolica Radiolarite Formation and the Czajakowa Radiolarite Formation, respectively. However, Polák *et al*. (1998) suggested a redefinition of the previously introduced lithostratigraphic units and proposed the Ždiar Formation as a new unit, comprising the entire radiolarite succession. The section studied demonstrates that if the view of Polák *et al*. (1998) were to be accepted, then two units with the rank of member should be defined.

CONCLUSIONS

The composite section of the Długa Valley provides an example of the Bajocian–lower Tithonian pelagic record in the Tethyan Fatricum domain (Krížna Nappe in the Tatra Mts). New $\delta^{13}C$ and $\delta^{18}O$ of bulk carbonate samples and $CaCO_3$ content analyses were derived from the previously unrecognized, uppermost Callovian and lower Oxfordian interval.

The bulk $\delta^{13}C$ values in the uppermost Callovian and middle Oxfordian remain nearly constant, with highly positive values. Such a pattern corresponds well with the global $\delta^{13}C$ pattern. A trend of increasing $CaCO_3$ content is observed in the uppermost Callovian–middle Oxfordian interval, which is accompanied with the facies change from ribbon radiolarites with shale partings to calcareous radiolarites with rare shale intercalations. It may reflect the end of the early Oxfordian carbonate production crisis and most probably records a recovery of the marine carbonate factory.

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