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Palynostratigraphy and palaeoenvironments around the Albian-Cenomanian boundary interval (OAE1d), North Bulgaria

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Abstract The palynological assemblages from two Albian-Cenomanian boundary sections in North Bulgaria are described. The samples analyzed ^yielded ^a diverse palynological content including dinoflagellate cysts and miospores. Based on dinocyst nutrient and productivity indices ^a ^phase of enhanced nutrient availability and high primary productivity is inferred for the latest Albian interval. The pronounced predominance of peridinioid dinocysts in this interval, namely *O. verrucosum*, *O. scabrosum* and especially *P. infusorioides* is considered to reflect eutrophic conditions. It coincides with the increased phosphorus mass accumulation occurring at the top par^t of the Upper Albian Dekov Formation. OAE 1d is indicated in the Tolovitsa karst spring section, based on palynofacies dominated by high amounts of granular amorphous organic matter (AOM) related to anoxic environmental conditions. These sections serve as evidence suggesting ^a relationship between Cretaceous peridinioid cysts (including *Palaeohystichphora infusorioides*, *Ovoidinium verrucosum*, *O. scabrosum*) and anoxic/suboxic conditions and/or high primary productivity. The pollen spectrum inferred relatively stable vegetation patterns of surrounding continental areas during and after the Albian/Cenomanian boundary interval and the times of OAE 1d formation. The hinterland vegetation integrated mainly ^pteridophyte spores and gymnosperms. The area was par^t of the Southern Laurasian floral province which was characterized by warm temperate to subtropical humid climate. Angiosperms were still minor par^t of this vegetation.

Keywords Albian, Cenomanian, Dinoflagellate cysts, Biostratigraphy, Palaeoenvironments, OAE1d

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1. Introduction

The middle par^t of the Cretaceous is known as ^a typical greenhouse-hothouse climate period characterized by ^a high sea level (Haq, [2014](#page-7-0)) and warm and equa^l ^global climate ([Hay](#page-7-0) and [Floegel,](#page-7-0) 2012). The interval including the Albian-Cenomanian boundary is ^a significant par^t of the mid-Cretaceous geological changes that have been recorded on the ^global scale. Agreement exists on the genera^l absence of permanent ice caps and minor temperature gradients between the equator and poles ([Barron](#page-7-0) et al., 1981; Sloan and [Barron,](#page-8-0) [1990](#page-8-0); [Takashima](#page-8-0) et al., 2006; Masure and [Vrielynck,](#page-7-0) 2009).

^A significant transgression occurred in Albian-Cenomanian times and epicontinental seas covered ^a large par^t of all continental ^plates, increasing the rapid radiation of marine organisms such as dinoflagellates (e.g. Bujak and [Williams,](#page-7-0) [1979](#page-7-0); [Leckie](#page-7-0) et al., 2002). ^A short palaeoclimate-palaeoceanographic event has been identified, characterized by the accumulation of sediments rich in organic matter and ^a short lived positive excursion of the carbon isotopes. This Oceanic Anoxic Event 1d (OAE 1d) developed across the eastern and western Tethys and other localities during the latest Albian (99.5 Ma) ([Leckie](#page-7-0) et al., 2002).

Extensive dinoflagellate cys^t information is available from Albian and Cenomanian sections both in the Tethyan and the Boreal realms (e.g., [Herngreen,](#page-7-0) 1978; [Foucher,](#page-7-0) 1983;

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[Leereveld,](#page-7-0) 1995; [Prossl,](#page-8-0) 1990; [Torricelli,](#page-8-0) 2000; [Pearce](#page-7-0) et al., [2009\)](#page-7-0). These studies demonstrate the high biostratigraphic potential of selected index dinocyst species, but also the value of dinocyst assemblages as palaeoenvironmental proxies for the reconstruction of surface water temperature (SST), productivity and nutrient levels (Masure and [Vrielynck,](#page-7-0) 2009), and sea-level fluctuations (Olde et al., [2015](#page-7-0)).

The purpose of the presen^t work is to presen^t an improved dinocyst framework for the Albian-Cenomanian boundary interval in two studied sections in North Bulgaria which are biostratigraphically well dated, and to use the potential of marine and terrestrial palynomorphs for palaeoecological and palaeoenvironmental interpretations in this par^t of the Tethyan ocean system.

2. Geological setting

Cretaceous strata in Bulgaria developed along the northern margin of the Tethyan ocean (Peri-Tethys, e.g., [Nikolov](#page-7-0) et al., [2009](#page-7-0)) on the adjoining European fragment of the Eurasian ^plate. From palaeogeographic point of view this Cretaceous basin was par^t of the Mediterranean Province of the Tethys. Today, the territory of North Bulgaria covers parts of two major tectonic units-the Moesian ^platform and the Alpine orogenic belt. Within the Alpine orogen we distinguish the Balkan and the Carpathian system ([Dabovski](#page-7-0) and [Zagorchev,](#page-7-0) 2009). The Moesian ^platform forms the foreland of the Alpine orogen. Its southern boundary is ^a complex system of flexures along which the Fore-Balkan and the Balkan zone overrides the ^platform. The sedimentary cover of the ^platform consists of subhorizontal Mesozoic and Cenozoic successions. Cretaceous strata are widespread in these different units, showing specific features in each of them. Now Albian and Cenomanian sediments are composed of different facies ranging from shallow water carbonates to deep-water flysch-type siliciclastics.

Studies of [Minkovska](#page-7-0) et al. (2002) and [Nikolov](#page-7-0) et al. [\(2009\)](#page-7-0) summarized published data for the Albian stage and inferred first Austrian tectonic compression and the genesis of ^a restricted anoxic foreland basin, characterized by deposition of anoxic black marls, locally interbedded with ^glauconitic sandstones. This basin was reduced considerably in time, but was still open towards its northern par^t (North Bulgaria and Romania) during the mid-Cretaceous. [Minkovska](#page-7-0) et al. (2002) and [Nikolov](#page-7-0) et al. (2009) explained this considerable reduction and especially the appearance of anoxic conditions as evidenced by black shale deposition by the first tectonic inversions of the Austrian tectonic ^phase. The early Cenomanian basin inherited in genera^l the boundaries of the late Albian. Sediment accumulation continued from late Albian to early Cenomanian without ^a significant break both in the Carpathian basin and in the western and central parts of the Moesian ^platform (Moesian epicontinental basin), where our sections were studied.

3. Studied sections

The Albian-Cenomanian boundary interval is palynologically studied in two representative outcrop sections, situated in northern tectonic units of Bulgaria [\(Figure](#page-2-0) 1). The sedimentary successions in these sections have been traditionally assigned to two different facies types-North-European and Carpathian (Jolkichev, 1989; [Dabovski](#page-7-0) et al., 2009). They originated in different depositional environments corresponding to the two types of interrelated Late Cretaceous basins in North Bulgaria-the North-European-Moesian shallow epicontinental basin and the Carpathian-type basin ([Dabovski](#page-7-0) et al., 2009). The Albian-Cenomanian interval of the North-European-facies type is restricted to the western par^t of the Moesian ^platform and is represented by shallow marine sediments such as ^glauconitic sandstones (Dekov Formation) and marls with thin intercalations of limestones (the Sanadinovo Formation). The Carpathian facies type comprises ^a deeper marine succession of clay-carbonate rocks including gray to grey-greenish marls with packets of argillaceous limestones (the Rabisha Formation).

The Sanadinovo section [\(Figure](#page-4-0) 2) comprises the most representative upper Albian-Cenomanian outcrop succession in the epicontinental setting. It comprises coarse grained ^glauconitic sandstones, belonging to the upper par^t of the Dekov Formation (Ivanov and [Stoykova,](#page-7-0) 1990) of Late Albian age (S. dispar ammonite zone, see [Ivanov](#page-7-0) et al., 1982) overlain by marls, intercalated with thin layered marly limestones recognized as Sanadinovo Formation, representing the lower-middle Cenomanian interval. The biostratigraphic framework of this section is based mainly on ammonites with information from foraminifers and calcareous nannofossils [\(Ivanov](#page-7-0) et al., [1982](#page-7-0); Jolkichev, 1989).

The section near Tolovitsa karst spring ([Figure](#page-5-0) 3) is situated northwest of Rabisha mound, Vidin district. Its succession is originally referred to as Carpathian-type Cretaceous and includes about ³⁸ ^m grey and grey-green thin-bedded marls with red spots in the upper par^t belonging to the Rabisha Formation. Originally, these strata were determined to be Albian-Cenomanian without further subdivision [\(Tzankov,](#page-8-0) [1960\)](#page-8-0). Recent nannofossil investigations of Sinnyovsky ([Sinnyovski](#page-8-0) and Pavlishina, 2014) subdivided this succession in two nannofossil zones: the *Eiffellithus turriseiffelii* and the *Lithraphidites acutum* zones. The former is subdivided in two subzones, namely the *Prediscosphaera columnata* and the *Prediscosphaera cretacea* subzone. The boundary between them is used to define the Albian-Cenomanian boundary (samples 3–4). This study aims to demonstrate the higher stratigraphic potential of the FO of *P. cretacea* for the Albian-Cenomanian boundary, at least in the studied area as well as in the Mont Risou Cenomanian GSSP section in

Figure 1 Location of the Sanadinovo and the Tolovitsa sections on a schematic tectonic map of Bulgaria (after Dabovski and Zagorchev, 2009).

southern France (e.g. [Kennedy](#page-7-0) et al., 2004).

4. Material and methods

^A total of ²⁴ samples have been analyzed for their palynological content. Each sample of ¹⁰⁰ ^g dry weight was processed using standard preparation techniques including HCl and HF treatment, heavy liquid separation and KON treatment No oxidation was applied to the sample material. The residues were finally sieved through 6–8 µm nylon meshes. Strew mounts were made in ^glycerine jelly and are stored in the collections of the Sofia University 'St. Kliment Ohridsky'. The reader is referred to [Williams](#page-8-0) et al. (1998) for dinocyst taxonomy.

5. Palynostratigraphy

Palynological studies contribute important high-resolu-

tion stratigraphic information characterizing the Upper Albian-Cenomanian transition in the studied area, and several important dinocyst bioevents within this stratigraphic interval. Within the Albian, the most easily recognized biohorizon appears to be the first appearance datum (FAD) of *L. siphoniphorum* at the base of the *S. dispar* Ammonite Zone ([Williams](#page-8-0) et al., 2004) and the most important dinocyst event within the Cenomanian, is the last appearance datum (LAD) of *Ovoidinium verrucosum* at the early-middle Cenomanian boundary. *Xiphophoridium alatum* and *E. spinosa* make their FO at the top of the Dispar zone around or just below the Albian-Cenomanian boundary ([Leereveld,](#page-7-0) 1995; [Williams](#page-8-0) et al., [2004](#page-8-0)). These events are considered in our sections for the biostratigraphical interpretation of the dinocyst assemblages.

Dinoflagellate cysts have ^a continuous record through the Sanadinovo section, they predominate and indicate biostratigraphically the *Litosphaeridium siphoniphorum*

Dinocyst Zone ([Herngreen,](#page-7-0) 1978). The characteristic zonal association comprises cosmopolitan dinocyst taxa such as *Litosphaeridium siphoniphorum*, *Epelidosphaeridia spinosa*, *Xiphophoridium alatum*, *Ovoidinium verrucosum* and *Palaeohystrichofora infusorioides*. range of *L. siphoniphorum*, *X. alatum* and *Ovoidinium verrucosum* points to the lower par^t of the *L. siphoniphorum* zone and, thus demonstrates the high stratigraphical potential of this association for the latest Albian and early Cenomanian interval. Additional characteristic species are *Ovoidinium sabrosum*, *Microdinium distinctum*, *Wrevittia cassidata*, *Achomosphaera sagena*, *Epelidosphaeridia spinosa* and *Chlamydophorella nyei*.

The recently documented dinocyst succession in the section near Tolovitsa karst spring also indicates the lower par^t of the *Litosphaeridium siphoniphorum* Zone in this section. The most abundant species in the zonal association are: *L. siphoniphorum*, *Epelidosphaeridia spinosa*, *Palaeohystrichofora infusorioides*, *Pterodinium ^pterotum*, *Achomosphaera sagena*, *A. ramulifera*, *A. neptuni*, *Odontochitina costata* and *Ovoidinium scabrosum.* The characteristic marker species *Litosphaeridium siphoniphorum* and *Xiphophoridium alatum* first occur in sample ² and continue their ranges in the overlying samples, while *Rhynchodiniopsis cladophora* and *Systematophora* spp. are restricted to the lowermost samples [\(Figure](#page-5-0) 3).

According to the dinoflagellate bio-events listed by [Williams](#page-8-0) et al. (2004) for the northern and mid latitudes, the assemblage of *L. siphoniphorum*, *E. spinosa*, *X.alatum*, *P. infusorioides*, *P. cingulatum* and *O. costata* defines an age of 100.95–96.7 Ma, using the time-scale of [Gradstein](#page-7-0) et al. [\(2012\)](#page-7-0). This corresponds to ^a late late Albian-early Cenomanian age and could be directly applied to the age determination of the studied section.

In both sections the group of land-derived sporomorphs is dominated by ^pteridophyte spores and gymnosperm pollen. Pteridophyte spores are presen^t in considerable high diversity and are typically represented by taxa assigned to *Cicatricosisporites*, *Bikolisporites*, *Gleicheniidites and Concavisporites.* Gymnosperm pollen from the *Araucariasites* com^plex is common whereas bissacate pollens are comparatively rare. These sporomorphs have more palaeoecological significance, rather than ^a stratigraphic value. Their combination indicates that the studied area was still par^t of the Southern Laurasian floral province ([Brenner,](#page-7-0) 1976) which was characterized by warm temperate to subtropical humid climate ([Hochuli,](#page-7-0) 1981).

Angiosperm pollen accounts on average for 7–8% of the total palynoflora and shows relatively low diversity, including undescribed forms. This pollen consistently occurs throughout this well-dated interval and is represented by triaperturate (tricolpate) forms of presumed eudicotyledon affinities. Most profuse species are *Tricolporoidites distinctus* and forms of the *Tricolpites* genus (Figure 4).

6. Discussion

6.1 Dinocyst productivity indicators and nutrient levels

Based on quantitative dinocyst data, productivity levels and nutrient availability can be reconstructed using the P/G (peridinioid/gonyaulacoid cycsts) ratio and the presence of palaeoenvironmental sensitive dinocysts. Dinoflagellates are very useful in estimation of surface-water nutrient levels because they possess groups that are either dominantly heterotrophic (the Peridinioid cysts or P-cysts) or ^phototrophic (the gonyaulacoid type cysts or G-cysts). Therefore, the ratio between these two groups (P/G ratio) provides an accurate measure of surface water nutrient availability (e.g. [Bujak,](#page-7-0) [1984](#page-7-0); [Powel](#page-8-0) et al., 1990; [Lewis](#page-7-0) et al., 1990; [Pearce](#page-7-0) et al., [2009\)](#page-7-0). The domination of P-cysts or high values of the P/G ratio are considered to be indicative of enhanced nutrient availability and to account for eutrophic conditions. In addition, certain peridinioid dinoflagellates are associated with elevated nutrient levels and thus are considered as high productivity indicators. For example, Pearce et al. [\(2009\)](#page-7-0) interpreted the Cretaceous species *P. infusorioides* to be ^a product of heterotrophic dinoflagellates and ^a prime indicator of nutrient-rich environments with high primary productivity.

Fundamental to better understanding the late Albian-early Cenomanian interval in the epicontinental settings is an estimation of surface water nutrient levels and productivity in the Sanadinovo section ([Figure](#page-4-0) 2). Such estimation is based on the P/G dinocyst ratio values and the presence of productivity indicators.

^A pronounced maximum in the P/G ratio is recorded in the uppermos^t Albian at the Sanadinovo section ([Figure](#page-4-0) 2), thus indicating ^a ^phase of enhanced nutrient availability. The marked domination of peridinioid dinocysts in this interval, namely *O. verrucosum*, *O. scabrosum* and *P. infusorioides* is considered to be indicative of eutrophic conditions. High nutrient levels and productivity are further supported by ^a pronounced dominance of the peridinioid dinocyst *P. infusorioides*, the product of heterotrophic dinoflagellates and prime indicator of nutrient-rich environments with high primary productivity ([Pearce](#page-7-0) et al., 2009). In our section, the top of the uppermos^t Albian ammonite Stoliczkaia dispar Zone (samples 455, 456) shows ^a higher representation of *P. infusorioides* within the dinoflagellate cys^t assemblages. These dinocyst data are documented within the uppermos^t par^t of the Dekov Formation and coincide with the increased ^phos^phorus mass accumulation occurring at this level ([Ivanov](#page-7-0) and [Stoykova,](#page-7-0) 1990).

The upper par^t of the section ^gives evidence for ^a declining marine productivity through the early and middle Cenomanian evidenced by the decreasing values of the P/G ratio

Figure ² Lithology and distribution of dinoflagellate cysts in the Sanadinovo section.

and the reduced abundance of *P. infusorioides*. However, the terrestrial input was high during the deposition of the Sanadinovo Formation documented by abundant spores, continental cuticles and tissues in the palynofacies.

The Tolovitsa karst spring dinocyst association also ^gives indications concerning nutrient availability and neritic productivity especially during the uppermos^t Albian interval. The encountered P/G ratio values in this interval, namely samples ¹ and ² ([Figure](#page-5-0) 3) as well as the abundance of *P. infusorioides* (less than 10%) indicate less eutrophic conditions, including nutrient availability and primary productivity in this deeper outer neritic environment than in the shallower epicontinental settings.

6.2 Palynofacies data and indications for OAE 1d

Palynofacies data from the Tolovitsa karst spring section corroborate well and complete the stratigraphic conclusions about the presence of uppermos^t Albian sediments in its lowermost part. Palynofacies documented in samples ¹ and ² are similar. Major constituents of the particulate OM in these samples are composed of grey greenish granular amorphous organic matter (AOM) which accounts up to 60% in the slides ([Sinnyovski](#page-8-0) and Pavlishina, 2014). The ^phytoclast fraction is represented by small equidimensional opaque particles, with single small translucent ^phytoclasts among them. According to Tyson ([1993](#page-8-0), [1995\)](#page-8-0) large amounts of AOM result from ^a combination of high preservation rate and low energy environments and are directly related to anoxic environmental conditions. Such palynofacies points to deposition in ^a distal, low-energy, anoxic environment with moderate terrestrial input in the basin. This information could be considered to indicate the well-documented ^global latest Albian oceanic anoxic event (OAE 1d) occurring at that time (99.5 Ma) across the Tethys ([Leckie](#page-7-0) et al., 2002; Ogg et al., [2004](#page-7-0)).

Biostratigraphically, this latest Albian Oceanic Anoxic Event (OAE 1d) spans the upper par^t of the ammonite Stoliczkaia dispar Zone, the *Rotalipora appenninica* ^planktonic foraminifera Zone and the *Eiffellithus turriseiffelii* NC

Figure 3 Lithology and distribution of dinoflagellate cysts in the Tolovitsa section.

¹⁰ nannofossil Zone ([Leckie](#page-7-0) et al., 2002; Ogg et al., [2004](#page-7-0); [Kennedy](#page-7-0) et al., 2004). The typical black, organic-rich interval correlates with the Breistroffer level at the Mont Risou Cenomanian GSSP section in southern France ([Kennedy](#page-7-0) et al., [2004](#page-7-0); Gale et al., [2011\)](#page-7-0), which occurs at ¹³⁵ to ¹²⁴ ^m below the top of the base Cenomanian GSSP. There, it comprises ^a set of thin, organic-rich beds, interbedded with pelagic marls ([Kennedy](#page-7-0) et al., 2004). This event has been identified not only in southern France, but also in Switzerland [\(Strasser](#page-8-0) et al., 2001), in southwestern North America ([Scott](#page-8-0) et al., 2013), the South Atlantic, and the southern Indian and eastern Pacific Ocean basins ([Wilson](#page-8-0) and Norris, [2001](#page-8-0); [Leckie](#page-7-0) et al., 2002). OAE 1d is associated with marine organic matter, high turnover in marine biota and radiation events, corresponding to an interval of major oceanic environmental changes and collapse of the water mass stratification [\(Erbacher](#page-7-0) et al., 1996; [Leckie](#page-7-0) et al., 2002).

Palynologically the OAE 1d event has been documented in the Mesilla Valley section, New Mexico, USA ([Scott](#page-8-0) et al., [2013](#page-8-0)) where it was located in the lower par^t of the upper Albian- Cenomanian *Ovoidinium verrucosum* dinocyst Zone, which correlates with the lower par^t of the *Litosphaeridium siphoniphorum* Zone ([Herngreen,](#page-7-0) 1978) and the uppermos^t Albian *Stoliczkaia dispar* ammonite Zone. The chronostratigraphic age of this event in the Messila Valley Formation was documented as uppermos^t Albian (97.39–97.30 Ma) and it

Figure ⁴ Dinoflagellate cysts from the Albian Cenomanian boundary interval in the Sanadinovo and Tolovitsa sections ca ^x 1000. (a) *Ovoidinium scabrosum*; (b) Ovoidinium verrucosum; (c) Litosphaeridium siphoniphorum; (d), (e) Microdinium distinctum; (f) Xiphophoridium alatum; (g) Canningia torulosa; (h) *Carpodinium obliquicostatum*; (i) *Psaligoyaulax deflandrei*.

was characterized by the consistent presence of the dinocyst markers *Ovoidinium verrucosum* and *Hapsocysta dictyota.* In contrast to the other, above mentioned oceanic pelagic basins, the organic-rich, anoxic signal in the Messila Valley Formation was deposited close to the shore and was strongly affected by terrigenous runoff and high rates of sediment accumulation ([Scott](#page-8-0) et al., 2013).

In contrast to the US section, the anoxic signal in the Tolovitsa karst spring section was most probably associated with elevated marine productivity and only moderate terrestrial input into the basin. The dinocyst species richness displays ^a relatively stable pattern throughout the sampled succession. The slight increase in diversity towards the top of the anoxic interval could be considered to indicate the response of dinoflagellate cys^t assemblages to ^a gradual sea-level rise following the relation between the dinocyst diversity and the eustatic sea-level change outlined by [Olde](#page-7-0) et al. [\(2015\)](#page-7-0). It could be also interpreted to be ^a P-OAE (P for productivity) according to [Erbacher](#page-7-0) et al. (1996).

7. Conclusions

The Albian-Cenomanian boundary interval is characterized based on dinoflagellate cysts, miospores and palynofacies data in two sections from North Bulgaria. Based on nutrient and productivity dinocyst indicators ^a ^phase of enhanced nutrient availability and high primary productivity is inferred for the uppermos^t Albian interval. The pronounced domination of peridinioid dinocysts in this interval, namely *O. verrucosum*, *O. scabrosum* and especially *P. infusorioides*, is considered to reflect eutrophic conditions. It coincides with the increased ^phosphorus mass accumulation occurring at the top of the Upper Albian Dekov Formation. OAE 1d is indicated in the Tolovitsa karst spring section. Palynofacies documented in the uppermos^t Albian interval from this section are characterized by ^a high amount of granular amorphous organic matter (AOM) which points to deposition in ^a distal, low-energy, anoxic environment with moderate terrestrial input into the basin. Our sections can serve as one more example suggesting ^a relationship between Cretaceous peridinioid cysts (including *Palaeohystichphora infusorioides*, *Ovoidinium verrucosum*, *O. scabrosum*) and anoxic/suboxic conditions and/or high primary productivity.

The miospore spectrum inferred relatively stable vegetation patterns during and after the Albian/Cenomanian boundary and the times of OAE1d formation. The hinterland vegetation integrated mainly ^pteridophyte spores and gymnosperms and the area was par^t of the Southern Laurasian floral province which was characterized by warm temperate to subtropical humid climate. Angiosperms were still minor par^t of this vegetation.

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