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Cyclic climate and vegetation change in the late Miocene of Western Bulgaria

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A B S T R A C T

A late Miocene paludal to lacustrine sequence from a carbonate basin in NW Bulgaria (Staniantsi Basin) is analysed displaying up to 27 rhythmically bedded sedimentary cycles. In the lower part of the sequence, the cycles consist of alternating autochthonous brown coal and marls containing diverse mollusc assemblages. The upper part of the sequence is characterized by alternating dark to light grey clays and calcareous silts. A palynomorph record comprising 163 samples is analysed by statistical means to reconstruct vegetation changes. The Coexistence Approach is used to calculate quantitative palaeoclimate records for 6 parameters. The studied section displays hierarchical cyclicity patterns. Longer-term cycles possibly related to eccentricity (period ~100 kyr) are present in the palynomorph record and show climate changes of warmer/wetter and cooler/drier periods in combination with frequency oscillations of thermophilous elements. Short-term cycles most probably related to precession (period ~21.7 kyr) are expressed by alternations of brown coal and marl/shell beds and show cyclic change in peat-forming vegetation related to oscillations of the groundwater level. As a triggering mechanism, wetter/warmer and drier/cooler climate phases related to orbital precession are probable. In addition, sections sampled at high resolution display small scale climate and vegetational variability.

As is shown by the analysis ferns were an important component of the peat-forming vegetation, while outside the mire, a wetland vegetation consisting of pioneers and a mixed mesophytic forest with evergreen shrubs existed. An oligo- to mesotrophic slightly alkaline lake became repeatedly established with a diverse mollusc fauna and a dense hydrophytic vegetation with characean meadows.

In the upper part of the section, a spreading of herbaceous vegetation is observed, also known from other contemporaneaous palynomorph records in Bulgaria and surrounding areas. The increase of Asteraceae in the upper part of the section, combined with a marked decrease in woody taxa, points to an opening of habitats and a decrease in mean annual precipitation. This trend is paralleled by the mollusc fauna which yields several terrestrial, partly xerophilous taxa.

1. Introduction

Late Miocene to early Pliocene climate variability and short-term climate changes related to orbital forcing have recently been studied. Various sections from small continental basins in the Tethyan and Paratethyan realm, such as continental carbonate/mudstone cycles from the Teruel Basin (Spain), dark and light-coloured marls in the Servia Basin (NW Greece), and brown coal/marl sequences from Ptolemais (NW Greece) have been discussed in this context (van Vugt et al. 1998; Steenbrink et al., 1999, 2000; Aziz et al., 2004). The cyclicity in those records is basically referred to alternation of dry and wet periods generating oscillations in the lake level producing rhythmic deposition of dark layers rich in organic matter during lake-level lowstand and of light marls during lake-level highstand (Steenbrink et al., 2006). A correlation with the polarity timescale has been proposed for the eastern Carpathian Foredeep, a large basin with high depositional rates (Vasiliev et al., 2004).

For some of the studied sections there is evidence that the observed climatic variations are correlated with vegetation changes documented as shifting proportion of key elements like altitudinal or thermophilous components in the pollen spectra (Popescu,

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2001, 2006). It has become quite clear that global climate change and small-scale climate variability are important driving forces in the evolution dynamics of regional vegetation. However, more detailed studies on the impact of climate change on specific associations and phytocoenoses are required. Moreover climate changes observed are not really quantified.

In the present study, a palynological record of a late Miocene paludal to lacustrine sequence from a carbonate basin in NW Bulgaria (Staniantsi Basin) which displays up to 25 rhythmically bedded sedimentary cycles is analysed to study causal connections of climate and vegetation change. Plant communities are reconstructed from the palynomorph record using statistical methods. Climate change and variability are analyzed with the Coexistence Approach (CA) (Mosbrugger and Utescher, 1997).

As a small graben structure in Triassic carbonate rocks, the Staniantsi Basin had no connection with neighbouring Cenozoic depositional areas (Vatsev, 1999). Therefore the terrestrial ecosystems that existed around the lake and in the mire potentially were very sensitive to environmental changes, such as water level and climate. Thus the present study contributes to a continuing effort to improve our understanding of the evolution of modern Mediterranean vegetation and climate in western Bulgaria, a very important key-region located at the transition of the Tethys and the Paratethys realms (Ivanov et al., 2007b, 2008).

2. Regional setting

2.1. Geology and sedimentary succession

The Staniantsi Basin is located in western Bulgaria and adjacent Serbia (Fig. 1). The structure represents a small NW–SE trending intramontane graben with a width of ca. 3 km and a length of about 10 km (Vatsev, 1999). The basement of the basin is formed by limestones of middle Triassic age. According to Yovchev (1960) the Neogene fill of the basin can be subdivided into four units. The basal unit (unit 1) consists of lacustrine clays reaching up to 40 m in thickness. This is followed by a brown coal unit (unit 2) with variable thickness up to 25 m in the central part. Unit three, about 25 m in thickness, is composed of lacustrine clays and marls, with single limestone beds occurring in the upper third of the unit possibly representing paleosols. Unit four (ca. 25 m thick) consists of comparatively homogenous light-green calcareous clays containing carbonate nodules. In some parts of the open cast mine is an erosional contact with sandy sediments deposited by incised fluvial channels. The whole succession is terminated by red Pleistocene sands and conglomerates discordantly resting on the Neogene strata. While unit one is only known from drillings, all the other units are well exposed in the Staniantsi open cast, where brown coal has been mined since 1966.

The approximate position of the measured section discussed is shown in Fig. 2. The measured section (Fig. 3) starts with 27.5 m brown coal of unit 2, displaying ca. 18 cyclic alternating layers of brown coal and marl (cf. example on Fig. 4(A)). The thickness of the cycles varies between 1.8 m and 2.0 m in most of the cases. In some parts of the brown coal, no cyclic sedimentary pattern is visible over several profile meters, but here colour changes of about the same thickness are observed, so that a total of 27 cycles exists. The thickness of the single marl layers commonly is within the range of a few centimetres. In only rare cases does a greater thickness occur (e.g., up to 40 cm at the depth level of 9.5 m). The overlying unit 3 in the profile consists of 20.5 m of ca. 14 alternating dark- and light-coloured calcareous clays and marls (cf. example on Fig. 4(B)). The darker clays may be replaced by thin brown coal layers, especially in the lower part of the unit, and the light-grey layers of the upper part have a silty component. The cycles of unit 3 are thicker when compared to unit 2 (between 2 m and 3 m). The uppermost 5 m of the unit show coarsening upward cycles, with sandy fluviatile channels intercalated. The marls and silty marls from level 47 m to 69 m belong to unit 4.

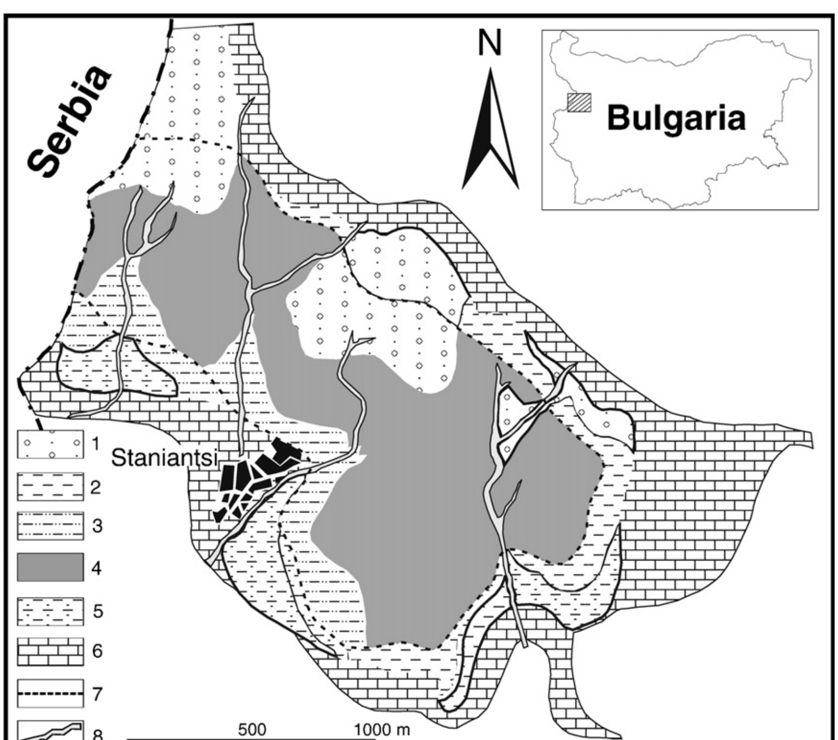

Fig. 1. Sketch map of the Staniantsi Coal Basin (redrawn from Yovchev, 1960 with corrections). 1) Quaternary conglomerates, sands and sandy clays; 2) grey to green clays; 3) grey fine clays and sandy clays; 4) brown coal; 5) dark grey clays; 6) Triassic basement; 7) assumed limit of the coal-bearing layer; 8) erosional valleys of temporary streams.

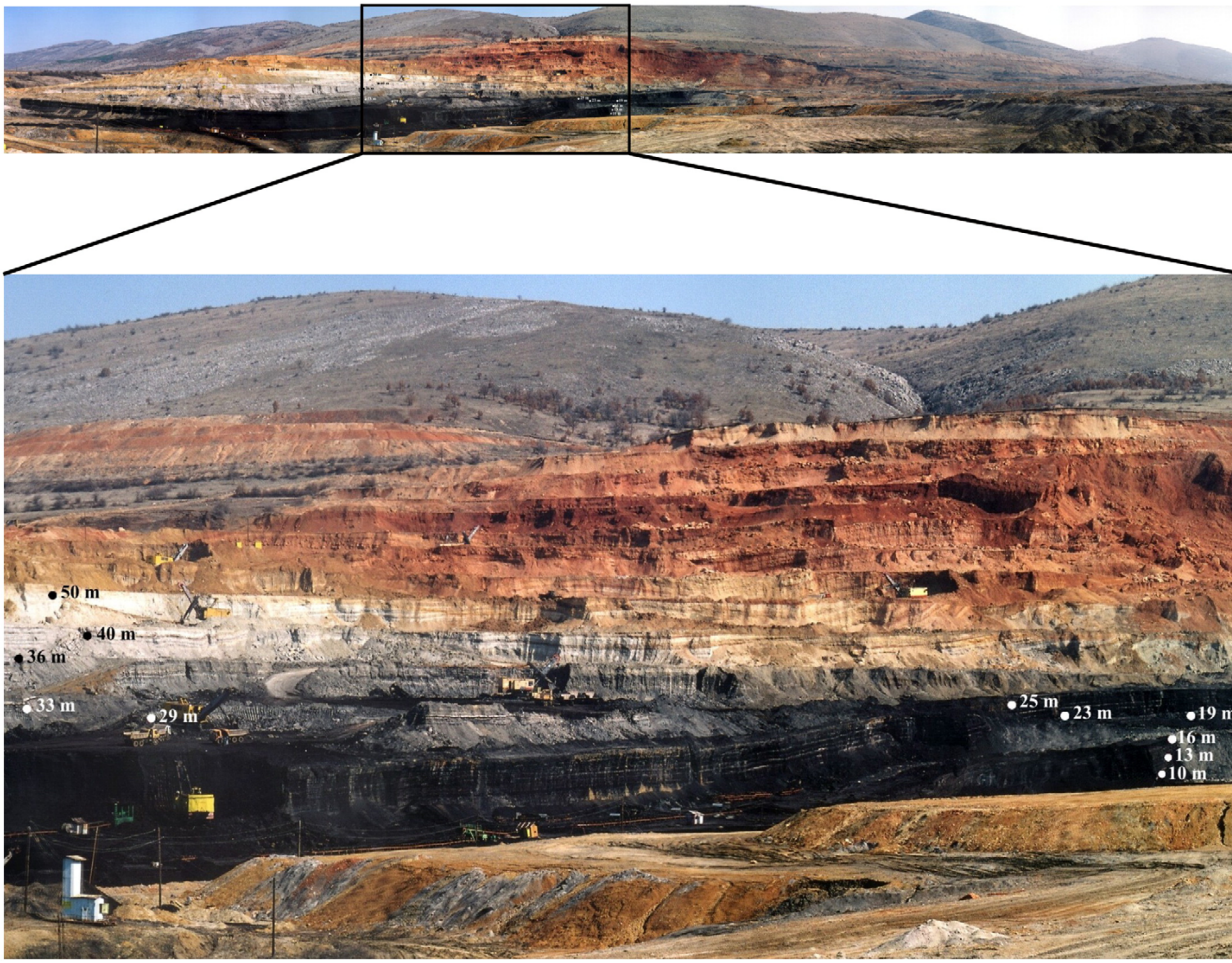

Fig. 2. Staniantsi Basin and open cast mine showing the position of the profile studied. The approximate depths are indicated.

These strata contain numerous horizons with carbonate concretions but no distinct primary sediment structures are observed nor any evidence for cyclic sedimentation. Unit 4 is overlain by alluvial fans and coarse-grained gravel of Pleistocene age resting on an erosional surface

2.2. Stratigraphical dating

Mammal remains at several levels of the profile in combination with palaeomagnetic data obtained from a total of 22 samples allow the section to be dated of (Fig. 3). According to mammal findings, the lower part of the brown coal (unit 2; levels 0 to 27 m in the section) belongs to the mammal zone MN13, most probably to its earlier, latest Maeotian part (Nikolov, 1985; Spasov, Daxner-Hoeck, pers. comm.). Between the depths of 30 and 45 m the clastic overburden of the brown coal seam also contains large and small mammal fauna, indicating a late (Pontian) MN13 age (Vrubljanski et al., 1959). Palaeomagnetic studies indicate reverse magnetisation for the major part of unit 2 (cf. Section 4.2). Combining these results, units 2 and 3 in the section cover the time-span from the latest Maeotian (ca. 6.5 Ma) to the Pontian (>5.3 Ma), whereas the silty marls with carbonate concretions on top (unit 4; level 47 m to 69 m) might be of Pliocene age.

3. Materials and methods

One main section and three higher resolution sub-sections were logged in detail in order to document sedimentary structures and facies. From the main section, sediment samples were collected at various levels and processed to study the fossil mollusc and mammal fauna. For palaeomagnetic studies two sample series with a total of 32 samples were collected and processed. For pollen analysis 85 samples were taken. Each sample contains homogenized bulk material of a single layer or of 50 cm sediment at a maximum in case of a constant lithology. 22 of the samples did not provide enough palynomorphs to obtain reliable results. For the 63 samples analysed in the main section, an average of 775 pollen grains per sample were counted (σ=407), yielding a mean taxa diversity of 33.2 (σ=6.9). In addition, 78 samples were collected from three cycles at a sample rate of 0.05 m (Fig. 4A; B).

For vegetational reconstructions, pollen frequencies of all the 141 reliable samples were analysed by factor analyses (software: SPSS 12.0). In this procedure, the Varimax method is used, an orthogonal rotation method minimizing for each factor the number of variables with high loading. The number of factors is determined from the Kaiser criterion (Backhaus et al., 2005). Component diagrams are shown in rotated space (Fig. 5). For this procedure 46 palynomorph

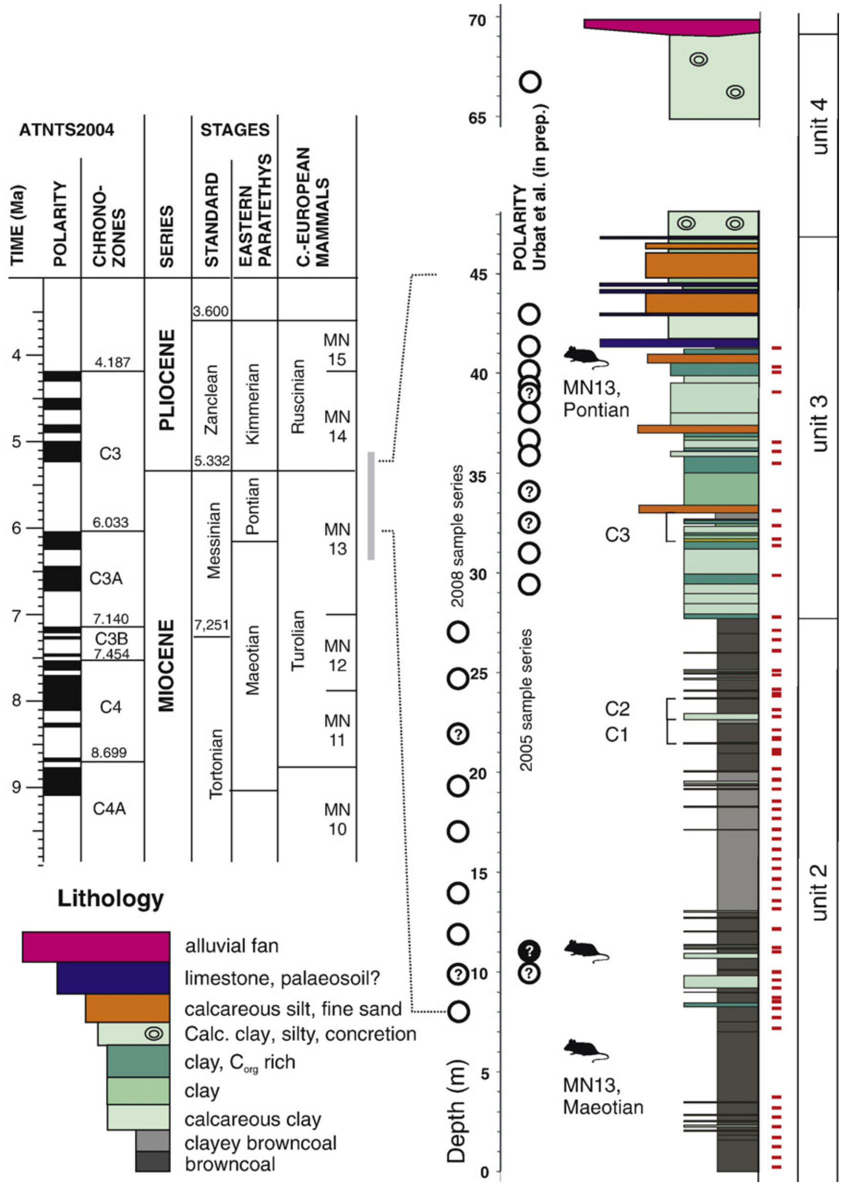

Fig. 3. Staniantsi section with stratigraphic data and polarity measured in the sample series from 2005 and 2008 (correlated). Question marks indicate samples with uncertain polarity. Circle: reverse polarity; dot: normal polarity. The bars next to the profile indicate the position of samples containing palynomorphs. C1–C3: Position of high resolution sections.

taxa are considered to be the most common in the sections. Palynomorphs sporadically occurring and attaining only minor percentages are not included because they do not provide any statistically significant correlations in the procedure. Factor analysis allows one to identify groups of taxa that show significant correlations and co-variance in the sections. These groups are then interpreted ecologically, plant communities are established, and their frequency distribution and cyclicity in the profiles are discussed.

To reconstruct quantitative palaeoclimate data we apply the Coexistence Approach (CA) (Mosbrugger and Utescher, 1997), a method based on climate requirements of all Nearest Living Relatives known for a fossil flora. The CA provides quantitative data for various climate variables, and it has been successfully applied to Neogene microfloral records (e.g., Bruch et al., 2002; Ivanov et al., 2002; Syabryaj et al., 2007; Ivanov et al., 2007a,b). The method is applied to a total of 141 samples, and 6 different variables are calculated: mean annual temperature (MAT), cold month mean (CMM), warm month mean (WMM), mean annual precipitation (MAP), precipitation of the driest month (MPdry), and precipitation of the warmest month (MPwarm). Seasonality of temperature, indicated for selected samples, is calculated as the difference between WMM and CMM.

Studies by Pross et al. (2000) have outlined the significance of the means of CA intervals. Following these considerations, means are used to visualize climate variability and its evolution.

4. Results

4.1. Sedimentary facies

The brown coal/marl cycles of unit 2 represent repeated changes of lacustrine to paludal conditions most probably related to a changing groundwater table. There is no evidence for any fluviatile input in this part of the succession. The brown coal can be regarded as dominantly autochthonous. It commonly contains numerous cuticles, small wooden fragments and charcoal, dispersed and stratified. Tree trunks with diameters of up to 15 cm are present and may also occur in distinct layers. However, stems from bigger trees are rare. Small root marks, most probably from herbaceous plants occur in rarer cases.

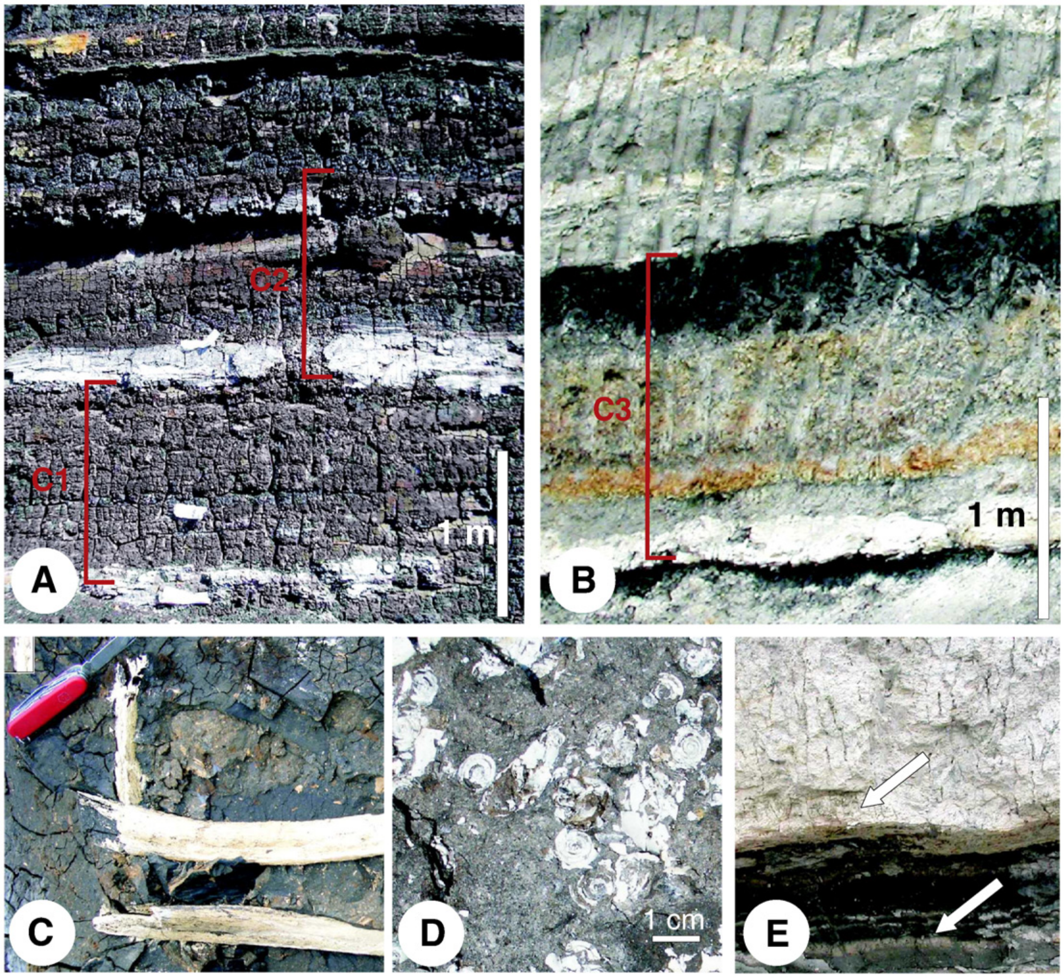

Fig. 4. Details of the Staniantsi section. A: brown coal/marl cycles at ca. 21.5 to 24 m of the section with position of high resolution sections C1 and C2 indicated (cf. Fig. 7); B: Silt/dark clay alternations in the upper part of the section (at ca. 32 m), with position of high-resolution section C3 indicated; C: Disarticulated remains of large mammals in the brown coal (at ca. 6 m), dispersed cuticles and small wood fragments; D: Horizon with planorbids and small shell fragments (at 10.80 m); E: Horizons with thin root marks (diameter ca. 1.5 mm) at depth level 36 m.

Mammal remains may be disarticulated but are partly very well preserved.

The overlying unit three shows cyclic changes of light-coloured clays, silty clays and silts, and darker clays with higher contents of organic matter. Although the latter may show a fine lamination, the light clays show no bedding structures, probably due to bioturbation. In one case trace fossils occur. Although most layers contain shells of aquatic molluscs there is also evidence for terrestrial facies at certain levels. At depth 32 m to 37 m, filigree root marks occur at the base of several organic clay layers pointing to the existence of an autochthonous obviously herbaceous vegetation spreading in times of a low water level (Fig. 4E). The limestone layers occurring in the upper third of the unit commonly are partly red or show red and green spots. According to their positions in the cycles they might represent paleosols.

4.2. Palaeomagnetics

The Palaeomagnetic studies are based on the analysis of 23 samples collected in 2005 from the measured section (Fig. 3). The studies are complicated by a very weak remanent magnetisation of the sediments comprising complex signals of secondary components. Therefore, 9 additional samples were taken in 2008 from the lower part of unit 2 (Fig. 3).

In all the samples studied the mean intensity of natural remanent magnetisation (NRM) was below 0.05 mA/m. Consequently, a reliable measurement of the magnetisation was not possible using the long core cryogenic magnetometer of the Leibniz Institute for Applied Geosciences (GGA-Institut) in Grubenhagen (Rolf, 2000). High sensitive three-axes 2G cryogenic magnetometers (Institute of Geology and Mineralogy, Cologne University (samples from level 8 to 30 m); Institute for Geophysics; Ludwig Maximilians University/ Munich (samples from level 30 to 68 m)) were used to carry out demagnetisation experiments in order to find stable remanent magnetisations using Principal Component Analyses (PCA; Kirschvink, 1980).

According to these analyses, sediments reveal a stable reverse magnetisation in the basal part of the section at ca. 8 m, from 14 m to 43 m and at 66.7 m. Belonging to the later part of MN13, the profile section between 8 and 43 m most probably can be correlated with the lower part of the C3 Chron showing reverse polarity (ca. 5.2–6 Ma).

4.3. Mollusc fauna

Several samples yielded rich and well preserved mollusc assemblages (Table 1). At level 20–26 m a low diverse aquatic fauna with *Lymnaea* sp. and very rare planorbids is found along with *Carychium pachychilus*. The latter is a terrestrial gastropod whose extant relatives are hygrophilous, leaf-litter dwelling snails that require moist conditions. *Carychium* settles wetlands and forests but is also found along

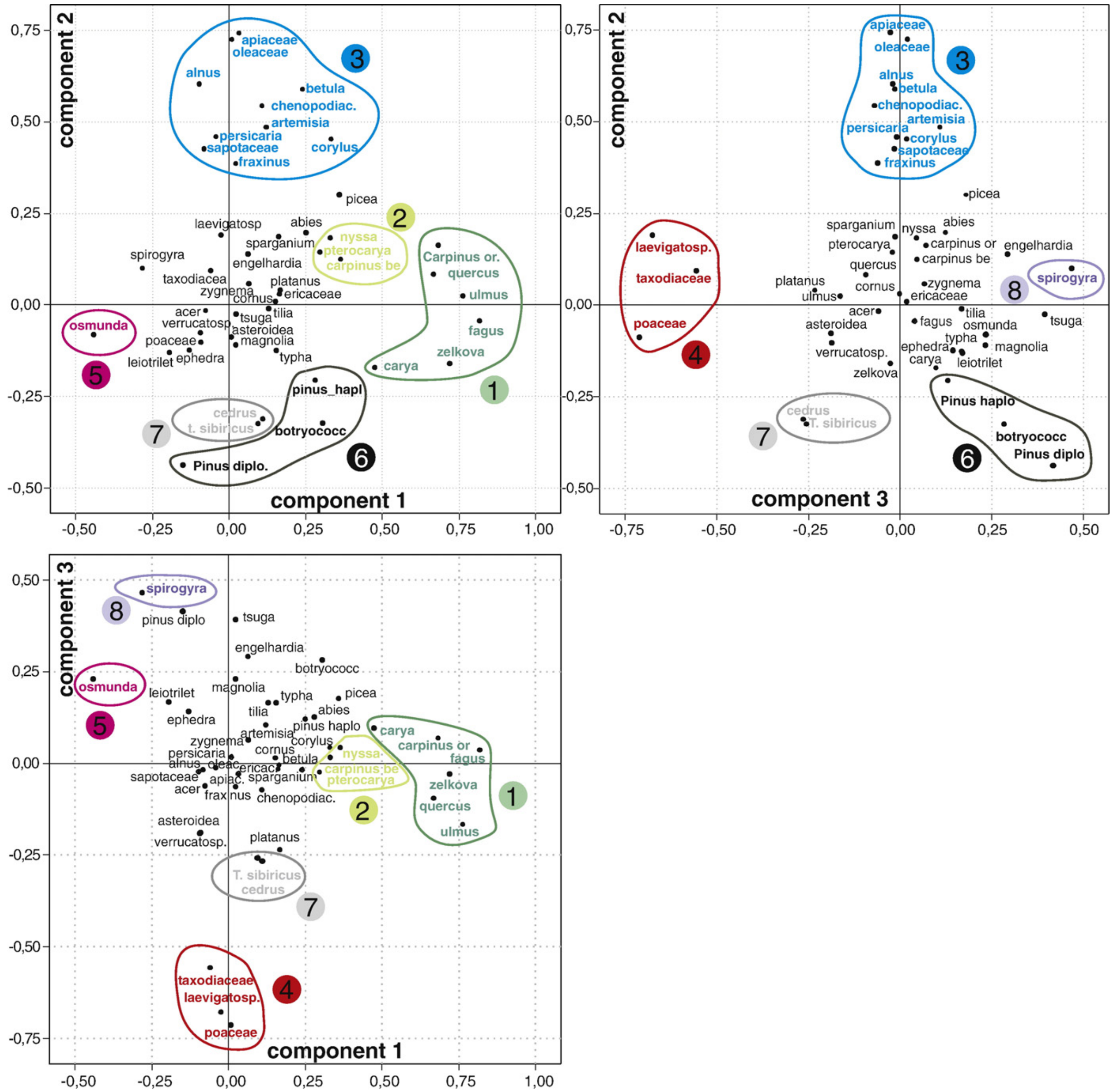

Fig. 5. Palynomorph groups obtained from factor analysis. 1) deciduous upland forest; 2) deciduous woods with *Nyssa*, *Pterocarya* and *Capinus betulus*; 3) wetland forest; 4) peat bog vegetation 1; 5) peat bog vegetation 2; 6) allochthonous community 1/*Botryococcus*; 7) allochthonous community 2; 8) *Spirogyra*.

lakes and ponds or even in tidal marshes (Ložek, 1964; Bruyndoncx et al., 2002; Frank, 2006).

From level 32–33.5 m only aquatic species are recorded. These samples are characterised by a high diversity of planorbids, which are represented by at least 6 species. Valvatids and lymnaeids are documented by 2 species each. All taxa are exclusively freshwater dwellers. Some of the species are also common elements in modern wetlands and thus allow direct comparisons: *Valvata piscinalis* prefers sand and mud bottoms with rich vegetation in lakes and slowly running rivers with pH-values between 6 and 9.6 (Glöer, 2002). Similar requirements are recorded for *Planorbarius corneus*, which needs temperatures above 18° and ph-values above 5 for reproduction. Like *Planorbis planorbis*, it may occur in slowly moving rivers but is most common in vegetated lentic environments (Wiese, 1991; Jopp, 2005). The high productivity of hydrophytic plants in planorbid inhabited ponds is partly even promoted by the nutrient release of the snails (Underwood, 1991). In addition, the occurrence of *Segmentina*, which avoids riverine settings, points to a purely lacustrine environment. Oligo- to mesotrophic conditions and hard water are favoured by all represented taxa also on the genus level (Lewin and Smolinski, 2006). Rich vegetation with *Chara* meadows is also proved by the common occurrence of Characeae oogonia in these samples. Decaying plant debris might also have been the trigger for periodic local low-oxygen conditions close to the sediment surface, as reflected by strongly pyritized sediment within some planorbid shells.

At level 35.0–35.5 m the fauna changes distinctly; the diversity of aquatic gastropods drops strongly and paludal conditions prevail. The composition with *Armiger* cf. *crista* and *Radix* sp. suggests a

Table 1
The mollusc fauna from Staniantsi. The first unit includes aquatic taxa, the second unit shows terrestrial elements

Staniantsi molluscs						
Molluscs taxon	20–26	32.7–33.0	33.0–33.5	35.0–35.5	40.0–40.4	41.0–41.5
Viviparus sp.	0	1	0	0	0	0
Bithynia sp.	0	1	0	0	1	1
Radix sp.	0	1	1	1	1	1
Lymnaea sp.	1	1	1	0	1	1
Gyraulus sp.	0	1	1	0	1	0
Planorbis planorbis (Linnaeus, 1758)	1	1	1	1	1	0
Planorbarius corneus (Linnaeus, 1758)	0	1	1	0	1	0
Armiger crista (Linnaeus, 1758)	0	0	1	1	0	0
Valvata (*Valvata*) *marginata* Michaud, 1855	0	1	1	1	1	1
Valvata (*Cincinna*) *piscinalis* (Müller, 1774)	0	1	0	0	0	1
Anisus cf. *mariae* (Michaud, 1862)	0	1	1	0	1	0
Segmentina filocincta (Sandberger, 1875)	0	1	1	0	1	0
Pisidium sp.	0	1	0	0	0	1
Carychium pachychilus Sandberger, 1875	1	0	0	0	0	0
Vertigo cf. *suevica* Gottschick and Wenz, 1919	0	0	0	1	0	1
Truncatellina cf. *cylindrica* (Férussac, 1807)	0	0	0	1	0	0
Gastrocopta (*Sinalbinula*) nov. sp.	0	0	0	1	0	0
Gastrocopta (*Albinula*) *acuminata* (Klein, 1846)	0	0	0	1	0	0
Limax sp.	0	0	0	0	1	1

relatively smaller, probably fragmented lake compared to samples 32–33.5 m (cf. Carlsson, 2001). This interpretation is supported by the occurrence of at least 4 species of terrestrial gastropods. Especially *Truncatellina* cf. *cylindrica* indicates an environmental change, being a xero- and calciphilous taxon that settles open habitats such as rocky slopes and screes (Ložek, 1964; Wiese, 1991). *Vertigo* and *Gastrocopta* also may be found in open drier habitats, but unlike *Truncatellina* they live also in a wide range of woodland habitats of varied humidity.

Lacustrine conditions similar to level 35 m became re-established at least at level 40.0–40.4 m. Planorbids are frequent along with numerous *Bithynia*. Oogonia of *Chara* indicate that *Chara* meadows were developed as well. The terrestrial component is strongly reduced aside from limacid slugs. These prefer forests with moderate moisture and avoid exposed habitats (Kerney et al., 1979).

At level 41.0–41.5 m lymnaeids and valvatids predominate in the assemblage, accompanied by the terrestrial gastropods *Limax* and *Vertigo*.

4.4. Vegetation analysis

A comparatively rich spore and pollen flora comprising 88 fossil taxa is recorded in the Staniantsi sections, (Appendix 1). It consists of plants from different taxonomic groups: lacustrine algae, pteridophytes, gymnosperms, and angiosperms. The latter provides the highest taxonomic diversity, namely about 70% of the taxonomic composition of the flora. Regarding biodiversity the comparatively low proportion in thermophilous taxa and palaeotropical elements is noteworthy. These elements are only sporadically present in the pollen spectra. Species allocated to the genera *Betula, Quercus, Fagus, Carya, Ulmus*, and the Oleaceae family are the most abundant among the angiosperm pollen recorded and point to the existence of a dominantly deciduous vegetation forest with some evergreen components in the undergrowth. In the major part of the samples, herbaceous plants are well represented in taxonomic diversity, but they only reach small proportions in the palynomorph spectra. However, at depths between 39 m and 41 m, non-arboreal pollen may reach up to 80% of the palynomorphs recorded. Regarding the spores, two pteridophyte species played an important role in the vegetation, apparently also in the peat-forming process — *Osmunda* and *Laevigatosporites* (cf. Thelypteridaceae) — both growing today in wet and swampy places. As lacustrine periphyton *Spirogyra* is most important, reaching up to 14% of zygospores at single levels. Today this genus of filamentous green algae is common in clean eutrophic freshwater, shallow stagnant water, or even on wet soil (Berry and Lembi, 2000). The abundance of zygospores is regarded as an important environmental indicator. Zygnemataceae produce spores during conjugation, when sexual reproduction occurs. Sexual reproduction seems to be triggered by fluctuations in nutrients or water level (Chmura et al., 2006). Thus higher frequencies of *Spirogyra* zygospores may correspond to higher water levels during mire formation.

In factor analysis for 46 palynomorphs frequent in the records (cf. chap. 3) three components explain 25% of the total variance in the data (Fig. 5). With respect to these three components, 8 groups are established comprising a total of 30 taxa. 15 taxa remain ungrouped because they show no specific affinities to one of the three components. The percentage proportions obtained for the groups are shown in pollen diagrams in Figs. 5 and 7 and are described in the following. All percentages cited below refer to the total frequency of these 46 taxa.

4.4.1. Component 1, positive correlation

Two groups are established that are both positively correlated to the component 1.

4.4.1.1. Deciduous upland forest vegetation. This group comprising *Carpinus orientalis, Carya, Fagus, Quercus, Ulmus*, and *Zelkova* most probably represents the deciduous upland vegetation growing on the foothills surrounding the basin. Although some of the taxa also have an affinity to floodplain vegetation the majority does not tolerate longer periods of inundation.

4.4.1.2. Deciduous woods with Nyssa, Pterocarya and Carpinus betulus.
This association potentially represents wetter stands of the forest, growing at the fringe of the basin, closer to the lake.

Group 1) commonly reaches 5 to 25% in the sections. Its frequency variability indicates short-term and longer-term cyclicity (ca. 1–2 m; ca. 6–8 m). In the main section, highest percentages are observed in the upper part. The pollen diagrams of the high resolution cycles show a clear affinity of group 1 to the brown coal facies. The wetter group 2 is sporadically present in the record, rarely exceeding 5%.

4.4.2. Component 2, positive correlation

4.4.2.1. Wetland forest. This association comprises trees (*Alnus, Betula, Fraxinus*), deciduous and evergreen shrubs (*Corylus, Fraxinus*, Oleaceae, Sapotaceae), and herbaceous plants (Apiaceae, *Artemisia*, Chenopodiaceae, *Persicaria*). *Alnus* may tolerate a permanently high groundwater level. The other genera include pioneer species. For some Sapotaceae, an affinity to peaty soils is known (e.g. Larson et al., 1981). The presence of *Persicaria* pollen clearly points to the existence of very wet edaphic conditions. Consequently, the record of Chenopodiaceae and *Artemisia* does not necessarily indicate dry conditions but may point to the presence of pioneer herbaceous communities on deforested areas and open shrubland. The wetland forest community has a pointed affinity to the peat bog facies in the sections where it may attain up to 50% of the palynomorphs recorded. Its habitat probably was the marginal parts of the mire and the aggradational zone most sensitive to water-level change (presence of pioneers).

4.4.3. Component 3, negative correlation

4.4.3.1. Peat-bog vegetation 1. The association combines *Laevigatosporites* (cf. Thelypteridaceae), attaining very high proportions, grasses, and pollen of Taxodiaceae reaching minor percentages. As the wetland forest (component 2), this group is clearly related to the peat facies and, according to the ecology of many Taxodiaceae and polypodiaceous ferns, interpreted as peat-forming vegetation. As shown in Figs. 5 and 7, the component shows a very pointed short-term cyclic variability, with percentage proportions shifting from 0 to over 50% (Figs. 5 and 7).

4.4.4. Component 1, negative correlation

4.4.4.1. Peat-bog vegetation 2 (Osmunda). Peat-bog vegetation 2 consists of a single member (*Osmunda*). Together with peat-bog vegetation 1, its changing proportions define the short-term cyclic variability in the brown coal part of the pollen diagram. The spores, reaching percentages of up to 60%, are more frequent in times of lake phases in the basin, replacing community 3. Today *O. regalis* is known as indicator for wet habitats growing around ponds, lakes, and streams (Schmeil et al., 2006).

4.4.5. Component 2, negative correlation; component 3, positive correlation

4.4.5.1. Allochthonous community 1 and Botryococcus. The association comprises *Pinus haploxylon* type (incl. *Cathaya*), *Pinus diploxylon* type, and *Botryococcus*, a green colonial alga. *Pinus* is more common in the upper siliciclastic part of the section, where it may dominate the spectra. Its high frequency may be explained by the absence of autochthonous palynomorphs and/or transport with sediment. *Botryococcus* is only frequent in the clastic overburden of the brown coal. High percentages of this alga are characteristic for oligotrophic lakes and estuaries. It blooms under elevated levels of dissolved inorganic phosphorus (Testa et al., 2001).

4.4.6. Component 2, negative correlation; component 3, negative correlation

4.4.6.1. Allochthonous community 2. This cluster combines cedar pollen and *Tricolporopollenites sibiricum.*, a palynomorph with unknown botanical affinity. In the brown coal of the main section, percentages of this group are commonly low, but may attain up to 20% in the clastic part. The high resolution records display cyclic variations of the percentages at a frequency of ca. 0.3 m, ranging between 5 and 25%. *Cedrus*, a high pollen producer commonly is interpreted as an indicator of seasonally drier conditions.

4.4.7. Component 3, positive correlation

4.4.7.1. Spirogyra. *Spirogyra* occurs up to the depth of 33 m in the main section, commonly attaining percentages below 5%. From 28 m to 33 m it co-occurs with *Botryococcus*, but above it is completely replaced by the latter. In contrast to *Botryococcus*, *Spirogyra* points to more eutrophic conditions. In the swamp environment these algae

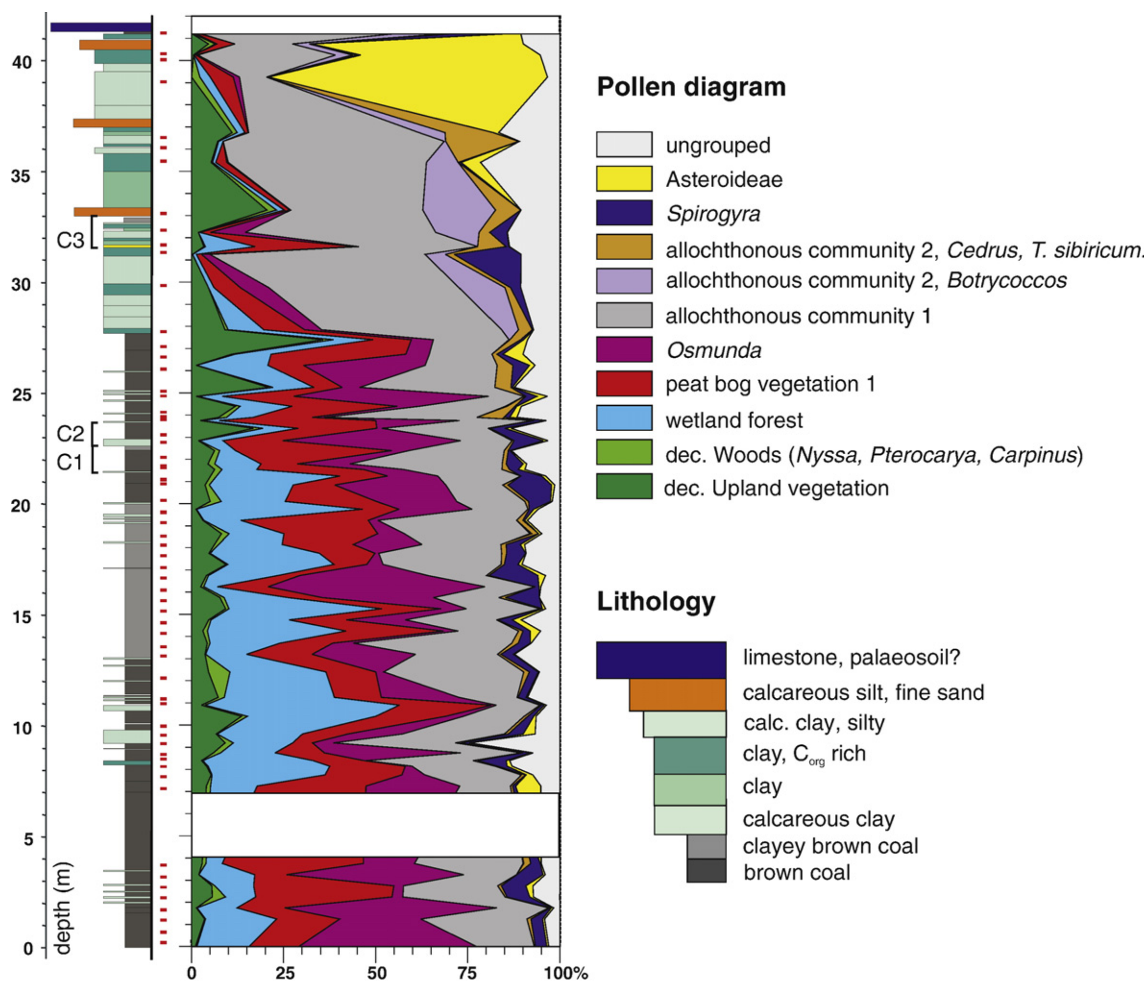

Fig. 6. Staniantsi section and diagram for groups obtained from factor analysis. The bars indicate the position of samples containing palynomorphs. C1–C3: Position of high resolution sections (cf. Fig. 8).

may occur in small stagnant but aerobic ponds as floating or submerged mats (McCourt and Howshaw, 2002).

4.4.7.2. Asteroideae. Asteroideae have no significant affinity to any of the components extracted by factor analyses. This is explained by the fact that their occurrence in the major part of the section is sporadic and does not exceed 10%. However, in three samples between 39 and 40.5 m in the main section, Asteroideae dominate the spectra reaching up to 75%, indicating an expansion of herbaceous communities.

4.4.8. Ungrouped palynomorph taxa

The following taxa are common in the palynomorph records but have no significant correlation with any component of the factor analysis and therefore remain ungrouped. These are *Leiotriletes* (pteridophyte), *Acer, Cornus, Ericaceae, Engelhardia, Magnolia, Platanus, Tilia* (angiosperm trees and shrubs), *Abies, Picea, Tsuga* (gymnosperms), *Sparganium, Typha* (angiosperm herbs), and *Zygnema* (algae).

4.5. Palaeoclimate analysis

The complete set of palaeoclimate data obtained for 141 samples of the main section and the three cycles sampled at high resolution (5 cm) is given in Appendix 2. For a total of 6 different variables (MAT, CMM, WMM, MAP, MPwarm, MPdry) lower and upper limit of the resulting CA intervals are presented. To visualize the results, means of CA intervals are used (Figs. 6 and 8).

In the analysis of the palynological record from the main section, 20 taxa at a mean contribute with climate data (Appendix 1). The CA commonly provides reliable results when more than 8 taxa are used (cf. Mosbrugger and Utescher, 1997). In some high-resolution samples the number of taxa used in the CA procedure may fall below this critical value (Appendix 2). As is evident from the data, MAT ranges between 12 °C and 18 °C, CMM between 0 °C and 12 °C, and WMM between 20 °C and 28 °C. Very warm conditions with MAT above 15.5 °C, CMM above 5 °C, and WMM above 25 °C results for several levels of the section. These shifts of the CA intervals to higher values basically are due to the occurrence of *Engelhardia* and/or *Reevesia*. CA intervals obtained for MAP cover a range between 370 mm and 1500 mm. Higher annual precipitation rates of at least 800 mm are related to the presence of *Engelhardia*, *Reevesia* and *Keteleeria*. While MPdry ranges between 10 and 40 mm for most of the samples MPwarm has a striking variability. Here shifts of Ca ranges between 50 to 60 mm and 80 to 170 mm are characteristic, with *Eucommia, Keteleeria,* and *Reveesia* bringing forth higher values.

Integrating over the different climate variables and the time-span regarded a warm temperate mostly humid type of climate existed in NW Bulgaria over the time-span concerned. With respect to precipitation a stronger seasonal imprint is obvious. The climate records obtained display cyclic changes of different order and magnitude. Details are discussed in the following chapter in the context of facies and vegetation change.

5. Discussion

5.1. Vegetation and climate evolution during the latest Miocene

The Maeotian/Pontian palynomorph record shown in Fig. 6 illustrates that the palynomorph spectra change along with the sedimentary facies, i.e., paludal versus lacustrine conditions. To analyze variations in the zonal vegetation during the time-span concerned the associations “deciduous lowland forest vegetation”, “deciduous woods with *Nyssa, Pterocarya* and *Carpinus betulus*”, “*Cedrus/Tricolporopollenites sibiricus*” and Asteroideae are plotted together with the “wetland vegetation” (cf. Section 3). In addition,

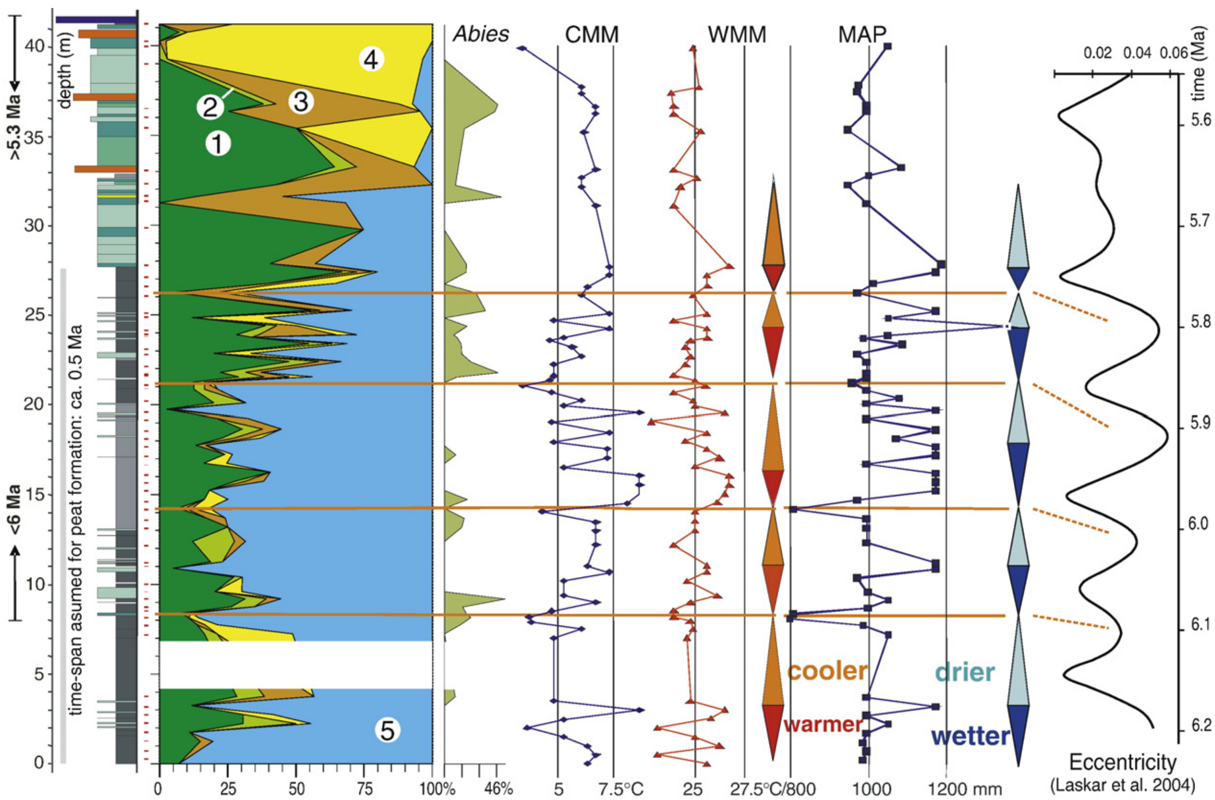

Fig. 7. Staniantsi section and diagram showing selected groups obtained from factor analysis. 1) deciduous upland forest; 2) deciduous woods with *Nyssa, Pterocarya* and *Carpinus betulus*; 3) *Cedrus–Tricolporopollenites sibiricum* group; 4) Asteroideae; 5) wetland forest. *Abies* percentages are calculated from total palynomorph sum. Longer-term vegetation and climate cycles with climate records for cold/warm month means (CMM/WMM) and mean annual precipitation (MAP) based on the means of coexistence intervals (cf. Appendix 2). Ages indicated refer to stratigraphic data shown in Fig. 3. Tentative correlation with the eccentricity curve (Laskar et al., 2004).

climate curves for CMM and MAP are given (Fig. 7). The record shows an overall increase in the proportion of the deciduous lowland vegetation. Also the *Cedrus*/*Tricolporopollenites sibiricus* association follows this long-term trend. This trend is continued up to the level of 37 m in the profile. There the pollen record shows an abrupt change expressed by the expansion of herbaceous vegetation mainly belonging to Asteroideae, as well as a sharp decrease in all woody taxa. This change is also indicated by the mollusc fauna, which points to a drop in lake level, a fragmentation of the lake system, and the spreading of drier and more open habitats settled by terrestrial gastropods. The expansion of herbaceous vegetation comprising the section between 37.7 and 41.0 m in the profile is evident from pollen counts of 3 successive samples. According to the available stratigraphical dating (C3 chron, upper part of MN13; cf. chap. 2) the interval represented in the section may be within the time-frame of the Messinian Salinity Crises.

The expansion of herbaceous vegetation is observed in various contemporaneous records from Bulgaria. Palynological data from the Tundzha Basin in SW Bulgaria indicate a wide distribution of herbaceous palaeocoenoses (NAP=48.2%) in that area (Ivanov et al., 2007a). There probably were at least two types of plant communities: A mesophytic herbaceous community of a wet prairie type growing in unforested and wet habitats along the basin (similar conditions are reported from the Pannonian Basin; cf. Hofmann and Zetter, 2005), and a community of xerophytic herbs of steppe character growing on dry terrains. Palynological data from the southwestern part of the Black Sea (DSDP hole 380A; cf. Popescu, 2006) also show an increase of the herbaceous component and the steppe/forest index (SFI) during the late Miocene to Pliocene. Also, a sharp increase and high percentages of the herbaceous component are recorded in the highest part of a late Miocene to Pliocene section in the Karlovo Basin (Ivanov and Slavomirova, 2004; unpublished data). The appearance and distribution of an *Artemisia* steppe in Turkey at the same time is discussed by Jimenez-Moreno et al. (2007). All these studies indicate a large-scale vegetation change at that time, most probably triggered by a changing climate.

Even though there is a clear vegetation trend observed along the section studied the temperatures and precipitations do not reveal such a well-defined long-term trend (Appendix 2; Fig. 7).

Just below the sudden increase of NAP, decreasing trend for MAP is apparent, but means do not significantly fall below 1000 mm. However, very high precipitation rates of at least 1000 mm (presence of *Reveesia*) and 800 mm, respectively (presence of *Engelhardia*) do not result for the samples above 31.6 m in the profile (Appendix 2). From 32 m on, *Carya* (>373 mm) *Fagus* (>422 mm), and two *Carpinus* species (>402 mm; 471 mm) delimit the ranges. Although these data point to a decline of precipitation, quantification remains difficult, because in microfloras taxonomic resolution is limited, and no Nearest Living Relatives can be specified to delimit the resulting precipitation ranges towards wetter conditions. Moreover, it has to be taken into account that for the vegetation growing close to the lake precipitation was not the only source of moisture. This could have a biasing effect on precipitation reconstruction based on the CA results.

5.2. Cyclicity

As stated above the Staniantsi section displays — apart from non-cyclic long-term changes — cyclic changes of different orders and frequencies (Section 4.5). Longer-term cycles with a period of 6 m to 12 m, reflected in the palynomorphs and in the climate

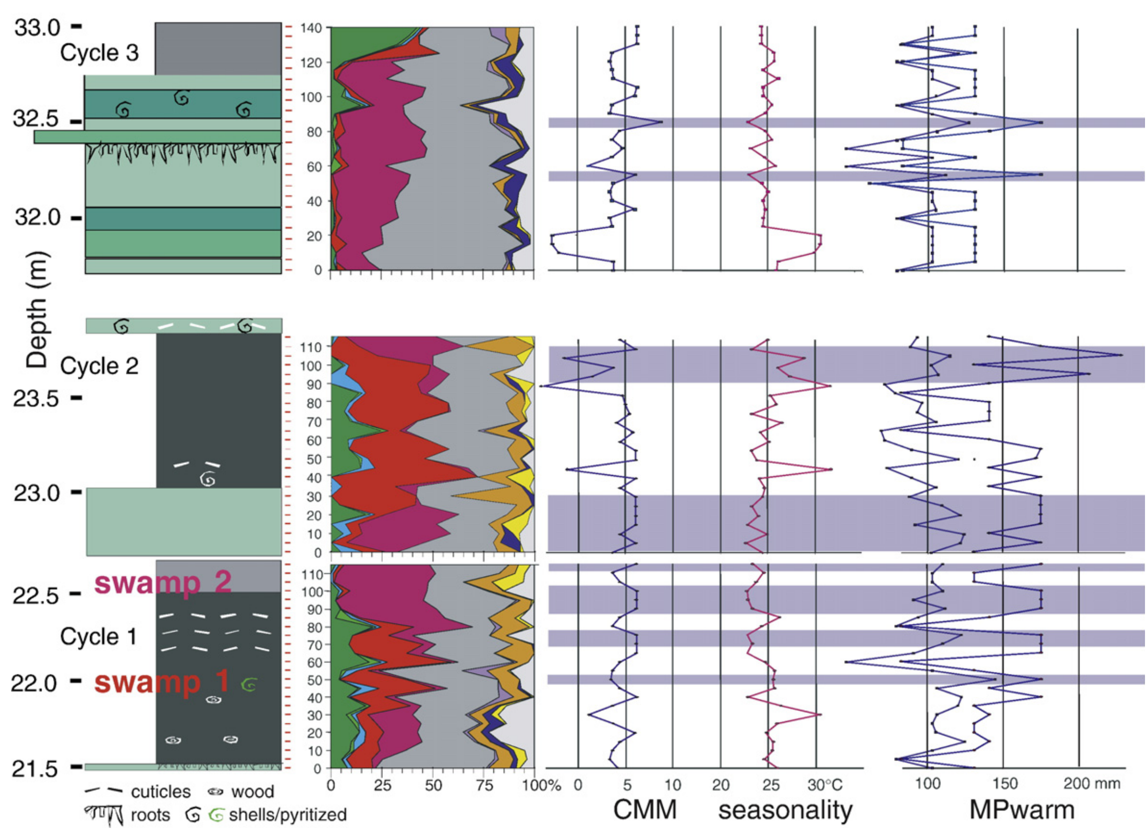

Fig. 8. High resolution sections (cf. Fig. 6 for positions in the main section) from the Staniantsi section. Grain size profile and diagram for palynomorph groups as obtained from factor analysis. The bars indicate sample positions. For lithology and pollen diagram cf. codes on Fig. 6. Climate records for cold month mean (CMM), seasonality of temperature, and precipitation in the warmest month (MPwarm). For the latter, means and upper limits of CA intervals are shown. Shaded depth levels in the climate records indicate wetter climate phases.

Table 2
Cyclicity observed in the brown coal (unit 2) of the Staniantsi section

Cycle type	Number of cycles observed	Cycle thickness (m)	Sedimentation rate in unit 2 as estimated from available stratigraphical data (m/kyr)	Periodicity (mean; min.; max.) (kyr)
Longer-term cycles	4–5	6.55 (4–7.5)	0.055 (~0.16 when decompacted)	119 (72–136)
Short-term cycles	ca. 27	1.3 (1.2–2)		23 (21–36)
Millennial scale variability	ca. 11	ca. 0.25		ca. 4.5

records are not correlated to any distinct lithological change (Fig. 7; see Section 5.2.1 for discussion) while short-term cyclicity with periods from 1 m to 2 m is related to facies change well expressed in the section (cf. Fig. 2; 5). A small scale climate and vegetational variability with a period of ca. 0.25 m is displayed in the high resolution records (Fig. 8).

Considering all the available stratigraphic information (Sections 2 and 4.2; Fig. 3) ca. 500 kyr is assumed to be represented by the brown coal seam in the section. Based on this assumption a sedimentation rate of ca. 0.16 m/kyr for unit 2 is obtained when the sequence is decompacted by factor 3. For the cycles periods of 119 kyr, 23 kyr and 4.5 kyr result (Table 2).

5.2.1. Longer-term cycles (6–12 m period)

Cyclic changes in the proportion of deciduous upland forest and wetland vegetation (Section 4.4) are observed (Fig. 7). In the brown coal seam (unit 2), these changes occur at a frequency of 6 to 7 m, in the upper, siliciclastic part of the profile thicker cycles of about 12 m are developed. These vegetational patterns can be correlated with temperature (MAT, CMM) and distinct precipitation (MAP) changes. Warmer and wetter phases in many cases correspond to a higher proportion of the upland vegetation, while in cooler, drier phases pollen from the local wetland vegetation dominates (Fig. 7). At that time possibly a less dense, patchy vegetation existed on the limestones of the surrounding uplands. As is shown by the temperature records the cooler phases are also characterized by increasing seasonality.

Factor analysis does not provide evidence for the existence of a typical altitudinal association in the palynomorph spectrum. This partially could be explained by the patchy occurrence of characteristic components in the section. To study a potential signal of the altitudinal vegetation and its connections to the cyclicity observed the *Abies* record is shown together with the climate record (Fig. 7). Although contributing with data in the CA analysis the taxon does not affect the results because of its very wide climatic range (cold boreal to tropical mountains). In the lower latitudes of Europe, however, this genus is presently confined to mountainous areas (e.g., Earle, 2008). It is shown that maxima in the *Abies* record are obviously connected to phases with cooler winters under a high eccentricity. As a possible source area for the component the main range of the Balkans comes into consideration, located in a distance of about 20 km.

As is evident from Table 2 the periods estimated for the longer-term cycles fall within the range of 100-kyr eccentricity cycles. Therefore, the eccentricity record according to Laskar et al. (2004) is tentatively plotted on Fig. 7 next to the pollen diagram. It is assumed that the cooler, drier phases are correlated with a high eccentricity, while phases with a lower eccentricity correspond to warmer, more equable climate phases.

100-kyr eccentricity cycles commonly are not well expressed in Neogene records (cf. Steenbrink et al., 2006). However, repetitive fluctuations referred to eccentricity are reported from an early Pliocene pollen record in Italy, close to the Apennines (Bertini, 1994), from a late Miocene to early Pliocene sequence from southern Romania (Popescu, 2001; Popescu et al., 2006), and from the early to mid-Miocene of the Pannonian Basin (Jiménez-Moreno et al., 2005). There maxima in the distribution of warm temperate elements are correlated with phases with a lower eccentricity, while cold winters under high eccentricity conditions are related to maxima in the distribution of altitudinal elements.

5.2.2. Short-term cycles (1–2 m period)

As is obvious from Fig. 6, the pollen diagram shows a pointed variability mainly due to proportion variations in the peat bog 1 and peat bog 2 (*Osmunda*) communities. This cyclic change has frequencies between ca. 1.2 m and 2 m and is well expressed in the brown coal part of the profile, where in many cases it is correlated with the changing sedimentary facies and the inundation events, respectively. In the brown coal part of the section, a total of 27 short-term cycles is developed. In the upper, clastic part of the section ca. 8 clay/marl cycles are present. In this part comparatively few samples have a diverse palynomorph record and hence this scale cannot be resolved.

Three subsections sampled at higher resolution (50 mm) show more details of the short-term variations (Fig. 8). In all the three cycles, the affinity of the peat bog 1 component (*Laevigatosporites*/*Taxodium*) to phases with peat forming is evident. Peat bog community 2 (*Osmunda*), in contrast, dominates the phases with marl sedimentation. Also the percentages of component 2 already increase in the upper centimetres of the brown coal layers, possibly reflecting a rise in the groundwater table. Cyclic frequency variations of *Osmunda* and monolete spores are known from Holocene sediments of the Everglades (Willard et al. 2001, 2006). There the increase in *Osmunda* spores reflects the onset of wetter conditions and higher water level, and the increase in monolete spores, in contrast, points to drier climatic conditions.

In order to reveal trends and variability in climate change, means of the CA intervals are used for the curves in Fig. 8. Apart from fluctuations interpreted as noise, consecutive data show periodic change in many of the cases. It is shown that in times of peat formation, the climate tended to be somewhat drier and cooler (cycle 1, 21.50–22.10 m; cycle 2, from 23.20 m on), whereas the presence of marls or coquina horizons is often related to slightly higher temperature and precipitation.

Although the use of CA interval means is well suited to the study of time series (cf. chap. 3), the quantification of climate change is problematic, because the CA intervals obtained for the samples largely overlap. However, dramatic climate changes can be excluded by the fact that taxa indicating a warm temperate climate persist throughout the sequences studied.

According to the inferred sedimentation rate and periodicity (23 kyr at a mean; cf. Table 2) the short-term cycles most probably reflect the impact of precession-induced fluctuations of the regional climate. Precession minima are supposed to correspond to episodes of increased precipitation, rise of lake-level, and distribution of the Swamp 2 vegetation (*Osmunda*-dominated pollen spectra, whereas precession maxima were coupled with decreased precipitation, lake-level decrease, and development of the Swamp 1 vegetation (*Laevigatosporites*-dominated pollen spectra). A correlation of organic layers with cooler and drier phases and marls with wetter, warmer phases in rhythmic sequences are assumed for other Neogene lacustrine deposits (e.g., Ptolemais Basin, N Greece; Terurel Graben, Spain) where they are referred to insolation changes caused by precession cycles (e.g., van Vught et al., 1998; Aziz et al., 2004). In the Teruel Basin climate change is derived from lithological characteristics and environmental interpretation of the carbonates. Qualitative interpretations of pollen spectra from Messinian brown coal/marl alternations in the Servia Basin (NW Greece) referred to the precession cycles support this assumption (Steenbrink, 1999).

Especially for the early Pliocene brown coal deposits of Ptolemais (northern Greece), there is evidence that the brown coal/marl alternations recorded are triggered by precession: marls correspond to precession minima (wetter phases) and brown coal to precession maxima (drier phases) (Van Vugt et al., 2001). The palynological analysis yields relatively high percentages of lowland trees (deciduous *Quercus*), herbs (Poaceae), and wetland elements (Taxodiaceae and Cyperaceae) in the (drier) phase with peat-forming (the first half of sedimentary cycle) (Kloosterboer-van Hoeve et al., 2006). The marl phase is characterized by high proportions of higher-altitude trees (mainly *Pinus*) in the second half of the cycle (wetter phase). This pattern reflects precession-controlled shifts from relatively dry to more humid conditions, with winter precipitation increasing during precession minima. Cyclic sequences from the Dacian Basin (southern Romania) covering the time-span from the late Miocene to the early Pliocene are also referred to orbital forcing (Popescu, 2001, 2006; Popescu et al. 2006). There the repeated increase in Cyperaceae percentages is assumed to be correlated with higher precipitation rates in times of precession minima favouring the spread of marshes at the flooded margins of the basin. Maxima of swamp forest vegetation, in contrast, are correlated to drier phases related to precession maxima.

5.2.3. Millennial scale climate variability

Millennial scale climate variability is mostly known from Pleistocene and Holocene records (e.g., Heinrich events, Dansgaard–Oeschger-cycles) and is partly referred to internal forcings such as periodic ice-sheet collapse, partly to external drivers as solar energy flux (e.g., Bond et al., 2001). In the high-resolution climate records of the 3 cycles analysed, increasing and decreasing trends are supported by several consecutive data points. Considering the sedimentation rate in the brown coal as a basis, these climate oscillations are on a millennial scale (ca. 4.5 kyr at a mean; Fig. 8; Table 2). In the records a comparatively strong signal is observed for the precipitation of the warmest month, corroborating the above assumption. This is uncommon in climate records from the central European Cenozoic (Mosbrugger et al., 2005). There MPwarm constitutes an almost stable climatic variable. To a lesser extent, millennial scale variability observed at Staniantsi also affects temperature and its seasonality. Thus the Staniantsi cycles may be within the time range of millennial scale vegetation changes reported from the early Pliocene of the Ptolemais section (Kloosterboer-van Hoeve et al., 2006). There the changes are referred to shifts in orographic winter precipitation possibly triggered by teleconnections with the North Atlantic Oscillation (NAO).

6. Summary and conclusions

A summary model of the late Miocene environment of the Staniantsi Basin is shown in Fig. 9. The model reflects the situation in the lower part of the succession in the brown coal (unit 2) at the time of peat formation. At that time a deciduous forest grew on the foothills around the mire that developed in the basin. In the marginal parts of the mire, a wetland vegetation type was composed of pioneers, evergreen shrubs, and herbs. Ferns, in contrast, were most important as peat-forming vegetation (*Laevigatosporites*; Thelypteridaceae?), while the “lake” phases were characterized by the formation of an *Osmunda* lake shore (?) community. At that time an oligo- to mesotrophic, slightly alkaline lake became established and was settled by a diverse mollusc fauna and a dense hydrophytic vegetation with characean meadows.

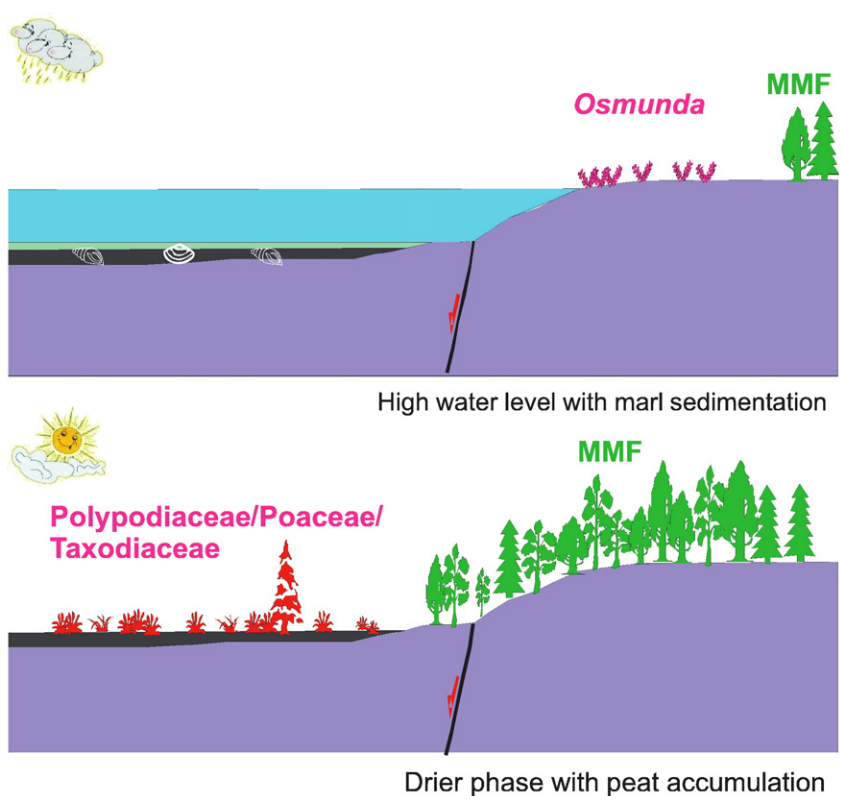

Fig. 9. Environmental model of the Staniantsi Basin for the time of peat-forming (brown coal unit).

Hierarchical cyclicity is expressed in the section. Longer-term cycles (period ~100 kyr) show frequency oscillations of thermophilous elements triggered by climate shifts from warmer/wetter to cooler/drier periods. Short-term cycles (period ~21.7 kyr) are expressed as brown coal/marl/shell layer alternations combined with cyclic change in vegetation types (swamp 1/2); these are interpreted as inundation cycles reflecting changes in groundwater level as is shown in Fig. 9. As a triggering mechanism, wetter/warmer and drier/cooler climate phases referred to orbital precession can be assumed, combined with local tectonic processes. The climate records reveal a distinct variability in temperature of the warmest month. This is uncommon in climate records from the central European Cenozoic and points to differing sensitivities of regional climates. The three subsections sampled at high resolution show short-term climate oscillations expressing changes in a sub-Milankovitch scale. These results show that the amplitudes of climate change were moderate, and that no dramatic changes occurred over the time span concerned.

The expansion of herbaceous vegetation in the upper part of the section can be correlated with other late Neogene records in Bulgaria and surrounding areas. The increasing abundance of Asteraceae, combined with a marked decrease in woody taxa, points to an opening of habitats, and most likely a distinct decrease in mean annual precipitation. This trend is paralleled by the mollusc fauna, which yields several terrestrial taxa. Phases of low water table led to a fragmentation of the lake, and open areas became settled even by some xerophilous gastropods.

Although the correlation of the hierarchical cycles observed at Staniantsi with orbital cycles in the moment is still tentative, the vegetation and climate changes observed fit overall with observations from other regions of the European late Miocene. In order to prove this hypothesis additional palaeomagnetic studies are planned to enhance the preciseness of the stratigraphical framework and, thus, of the time-control in the section.

Acknowledgments

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Appendix 1. Palynomorph taxa recorded in the Staniantsi sections; X: frequent taxa

Palynomorphs recorded in the Staniantsi section

Palynomorph taxon	Frequent in the record
Pinus diploxylon	X
Pinus haploxylon/Cathaya	X
Abies	X
Picea	X
Cedrus	X
Keteleeria	
Tsuga	X
Ginkgo	
cf. Podocarpus	
Tricolporopollenites sibiricum	
Platanus	X
Ericaceae	X
Betula	X
Carpinus orientalis–Ostrya	X
Carpinus betulus	X
Corylus	X
Cornus	X
Quercus	X
Fagus	X
Acer	X
Castanea	
Ulmus	X
Zelkova	X
Fraxinus	X
Tilia	X
Eucommia	
Carya	X
Magnolia	X
Pterocarya	X
Juglans	
Engelhardia	X
Reevesia	
Buxus	
Symplocos	
Sapotaceae	X
Araliaceae	
Anacardiaceae	
Ephedra	
Oleaceae	X
Pistacia	
Celtis	
Vitaceae	
Humulus	
Caprifoliaceae	
Rhus	
Taxodiaceae	
Alnus	X
Nyssa	X
Myrica	
Salix	
Liquidambar	
Inaperturate	
Chenopodiaceae	X
Poaceae	X
Cystaceae	
Asteroideae	X
Artemisia	X
Polygonum	
Caryophyllaceae	
Rosaceae	
Thalictrum	
Apiaceae	X
Persicaria	X
Lamiaceae	
Mentha/Salvia	
Knautia	
Viola	
Apocynaceae	
Rubiaceae	
Cyperaceae	
Typha	X
Arecipites	
Butomus	
Sparganium	X
Shagnum	
Laevigatosporites	X
Osmunda	X
Equisetum	
Polypodium	
Leiotriletes	X
Verrucatosporites	X
Echinatisporis	
Selaginella	
Botryococcus	X
Fungi	
Mougeotia	
Spirogyra	X
Zygnema	X

Appendix 2. Climate data obtained for the main section and the high resolved cycles (C1–C3) using the Coexistence Approach

Depth (m)	Ntaxa	MATmin	MATmax	CMTmin	CMTmax	WMTmin	WMTmax	MAPmin	MAPmax	MMPdrymin	MMPdrymax	MMPwmin	MMPwmax
0.25	32	15.7	17.6	5	7.7	24.7	26.4	1096	1146	9	41	55	61
0.75	22	15.7	17.6	3.8	9.7	20.2	26.4	1096	1520	9	41	55	61
1.25	27	15.6	17.5	5	7.7	24.7	27.6	823	1146	9	41	55	95
1.75	22	13.3	17.6	0.9	9.7	23.6	26.4	471	1520	8	41	51	83
2.25	26	11.6	16.2	−0.3	7.5	20.2	26.4	581	1520	18	41	55	68
2.75	24	12.9	17.6	0.9	9.7	23.6	27.9	471	1520	8	41	55	61
3.25	21	15.6	18.4	5	12.5	24.7	28.1	823	1520	18	41	49	61
3.75	21	11.6	17.6	−0.1	9.7	21.7	27.9	471	1520	8	41	55	61
7.25	25	11.8	17.6	−0.1	9.7	21.7	27.6	581	1520	8	41	55	61
7.75	18	11.6	18.4	−0.3	12.5	21.7	28.1	453	1520	8	41	45	61
8.15	21	11.6	17.5	−0.1	7.7	21.7	27.9	451	1146	9	41	47	61
8.4	15	11.6	17.5	−0.3	7.7	21.7	26.4	471	1146	9	41	47	83
8.75	21	11.6	17.6	−0.3	9.7	21.7	26.4	473	1520	9	41	47	83
9.2	25	15.7	17.6	3.8	9.7	21.7	27.9	1096	1520	9	41	47	83
9.6	19	12.9	18.4	0.9	9.6	23.6	28.5	473	1520	8	41	108	172
10.4	18	11.6	18.3	−0.3	10.9	21.7	27.6	422	1520	8	41	47	122
10.9	19	15.6	17.6	5	9.7	24.7	26.4	823	1520	8	41	79	83
11.25	28	15.6	17.5	5	7.7	24.7	26.4	823	1520	8	41	79	83
12.4	22	15.7	17.6	3.8	9.7	21.7	26.4	1096	1520	8	41	55	83
13.2	21	15.7	17.6	3.8	9.7	23.6	26.4	1096	1520	8	41	51	83
13.7	21	15.7	17.6	3.8	9.7	23.6	26.4	1096	1520	9	41	51	83
14.25	19	12.9	17.5	0.9	7.7	23.6	26.4	471	1146	9	41	51	83
14.75	21	15.7	18.4	3.8	12.5	23.6	28.5	1096	1520	8	41	51	172
15.25	17	15.6	18.4	5	12.5	24.7	28.1	823	1520	9	41	79	172
15.75	18	15.6	18.4	5	12.5	24.7	28.5	823	1520	8	41	79	172
16.25	17	15.6	18.4	5	12.5	24.7	28.5	823	1520	8	41	79	172
16.75	20	12.9	18.3	0.9	9.7	23.6	26.4	471	1520	9	41	51	83
17.25	25	15.6	18.3	5	9.6	24.7	27.6	823	1520	8	41	108	122
17.75	23	15.6	17.6	5	9.6	24.7	26.4	1096	1520	9	24	45	172
18.15	26	11.6	17.6	−0.1	9.7	22.8	26.4	619	1520	9	41	47	83
18.65	22	15.6	17.6	5	9.7	24.7	26.4	823	1520	8	41	79	83
19.25	18	11.6	17.6	−0.3	9.7	19.6	26.4	471	1520	8	41	47	83
19.75	20	15.6	18.4	5	12.5	24.7	28.1	823	1520	9	41	79	172
20.1	16	12.9	17.6	0.9	9.7	23.6	26.4	471	1520	8	41	51	83
20.4	11	11.6	18.4	−0.3	12.5	21.7	28.1	578	1577	9	41	70	172
20.85	15	11.6	17.6	−0.3	9.7	21.7	26.4	471	1520	8	41	47	83
21.25	14	10.6	19.4	−2.7	9.6	21.7	29.3	396	1520	8	59	108	172
21.55	16	10.6	17.6	−0.3	9.6	21.7	28.3	471	1520	8	41	45	172
21.8	17	11.6	17.6	−0.1	9.7	21.7	26.4	471	1520	9	41	45	61
22.4	21	13.3	17.6	−0.1	9.7	22.8	26.4	471	1520	8	41	55	83
22.85	16	11.6	18.4	−0.3	12.5	21.7	27.9	422	1520	8	41	55	172
23.4	27	11.6	18.4	1.7	9.6	22.8	26.4	652	1520	16	41	55	61
23.75	21	11.6	18.4	−0.3	9.6	21.7	27.9	453	1520	9	41	108	172
23.9	26	12.9	17.6	0.9	9.7	23.6	27.6	578	1520	9	41	70	83
24.4	26	15.6	18.3	5	9.7	24.7	26.4	1187	1520	8	41	79	172
24.85	22	11.6	17.6	−0.1	9.7	21.7	26.4	581	1520	9	41	47	61
25.25	27	15.7	17.6	5	9.7	24.7	26.4	1096	1520	9	41	45	172
26.25	16	11.6	18.4	−0.3	12.5	21.7	28.1	422	1520	9	41	47	172
26.75	21	13.6	18.3	1.8	10.9	23.6	27.6	505	1520	5	41	47	172
27.4	28	15.6	17.6	5	9.7	24.7	26.4	823	1520	8	41	79	83
27.85	18	15.6	17.6	5	9.7	24.7	28.5	823	1548	8	41	79	83
31.25	17	15.7	17.6	3.8	9.7	21.7	26.4	1096	1520	8	41	47	122
32.3	8	11.6	18.4	−0.3	12.5	19.3	29.4	373	1520	8	41	45	175
32.8	13	11.6	18.4	−0.3	12.5	21.7	28.5	422	1577	8	41	47	175
33.25	24	15.7	17.6	3.8	9.7	21.7	26.4	1096	1548	8	41	55	175
35.35	11	11.6	18.4	−0.1	12.5	21.7	28.8	373	1520	2	41	47	61
36.35	18	15.7	17.6	3.8	9.6	21.7	26.4	1096	1520	9	41	45	122
36.75	23	15.7	17.6	3.8	9.6	21.7	26.4	1096	1520	9	41	45	122
37.5	8	11.6	18.4	−0.3	12.5	19.4	28.5	422	1520	8	41	47	175
37.85	10	11.6	18.4	−0.3	12.5	21.7	28.7	373	1577	3	41	13	172
40	8	7.6	20.8	−6.5	13.3	21.7	28.1	373	1724	8	43	45	195
Depth (cm)	C 1 cycle												
0	14	12.9	17.1	0.9	7.5	23.6	26.8	735	1347	24	41	74	131
5	17	11.6	16.6	−0.3	7	19.4	26.4	735	1347	25	41	74	83
10	18	11.6	17.1	−0.3	7.5	21.7	26.4	735	1347	24	41	74	131
15	11	11.6	18.4	−0.3	9.1	21.7	28.1	578	1577	9	41	108	141
20	14	15.6	16.6	5	7	24.7	26.8	823	1347	24	41	79	131
25	14	13.3	17.1	−0.1	7.5	22.8	26.4	735	1347	24	41	74	131
30	16	13.6	16.6	0.6	1.7	25.3	27.8	578	1577	25	41	70	141
35	11	11.6	17.1	−0.3	7.5	21.7	28.1	735	1347	24	24	108	131
40	10	11.6	18.4	−0.1	12.5	19.6	28.5	578	1520	5	41	70	175
45	13	11.6	18.4	−0.3	9.1	21.7	28.3	578	1520	5	41	70	141
50	8	11.6	16.6	−0.1	7	19.6	28.3	578	1577	25	41	115	175
55	16	11.6	17.1	−0.3	7.5	21.7	26.8	735	1347	24	41	74	131

Appendix 2 (*continued*)

Depth (cm)	C 1 cycle												
60	9	11.6	17.6	−0.3	9.1	21.7	26.4	471	1548	5	41	6	83
65	5	11.6	18.4	−0.3	12.5	19.4	28.3	422	1577	5	41	5	175
70	4	11.6	18.4	−0.3	12.5	19.4	29.4	373	1577	8	41	45	175
75	8	11.6	18.4	−0.3	12.5	19.6	28.1	578	1577	8	41	70	175
80	10	11.6	17.1	−0.3	7.5	19.4	26.4	735	1347	24	41	74	83
85	6	11.6	18.4	−0.3	9.1	21.7	29.5	413	1577	24	41	45	141
90	5	11.6	18.4	−0.3	12.5	19.4	29.4	413	1577	24	41	48	175
95	4	11.6	18.4	−0.3	12.5	19.4	28.5	422	1577	5	41	5	175
100	7	11.6	18.4	−0.1	12.5	19.4	28.5	373	1577	8	41	45	175
105	6	11.6	17.1	−0.3	9.1	19.4	26.8	735	1347	24	41	74	131
110	7	11.6	17.1	−0.3	7.5	19.4	26.8	735	1347	24	41	74	131
115	4	11.6	18.4	−0.3	12.5	19.4	29.5	373	1577	2	41	45	175
Depth (cm)	C 2 cycle												
0	8	11.6	17.1	−0.3	7.5	19.4	26.8	735	1347	24	41	74	131
5	5	11.6	18.4	−0.3	12.5	19.6	28.1	578	1577	9	41	70	175
10	9	11.6	18.4	−0.3	9.1	20.2	28.6	399	1577	8	41	108	141
15	4	11.6	18.4	−0.3	12.5	19.4	28.6	206	1577	2	41	8	175
20	8	11.6	18.4	−0.3	12.5	21.7	28.6	578	1577	8	41	70	175
25	3	11.6	18.4	−0.3	12.5	19.4	29.5	373	1577	8	41	45	175
30	1	11.6	18.4	−0.3	12.5	19.4	31.8	164	1577	0	41	0	175
35	5	11.6	18.4	−0.3	9.1	19.6	28.6	578	1577	2	41	70	141
40	2	11.6	18.4	−0.3	12.5	19.4	31.2	222	1577	0	41	3	175
45	6	4.4	19.2	−11.5	9.1	21.7	29.4	422	1864	5	59	5	141
50	11	11.6	18.4	−0.3	12.5	21.7	28.3	578	1577	5	41	70	172
55	3	11.6	18.4	−0.3	12.5	19.4	29.4	222	1520	0	41	3	175
60	3	11.6	18.4	−0.3	9.1	19.4	29.7	201	1577	2	41	2	141
65	15	13.6	17.6	1.8	9.7	23.6	26.4	505	1520	9	41	55	83
70	11	11.5	19.2	−1	9.1	23	28.1	619	1724	9	59	70	141
75	8	11.6	19.2	1.7	9.1	19.4	28.1	652	1520	16	41	45	141
80	12	12.9	18.4	0.9	9.1	23.6	28.3	619	1520	8	41	51	141
85	15	13.6	17.1	1.8	7.5	23.6	26.4	735	1347	24	41	74	83
90	3	0.2	19.2	−16.6	9.1	16.3	29.5	206	1864	2	59	2	141
95	3	7.4	16.6	−4	7	20	27.8	206	1864	2	59	8	207
100	9	7.6	17.1	−0.1	7.5	22.8	26.8	735	1347	24	59	74	131
105	2	0.2	22.2	−16.6	13.6	16.3	28.5	328	1864	2	59	2	228
110	2	11.6	18.4	−0.3	12.5	19.4	29.5	206	1577	2	41	2	175
115	4	11.6	18.4	−0.3	9.1	19.4	29.5	373	1577	8	41	45	141
Depth (cm)	C 3 cycle												
0	12	13.3	17.1	−0.1	7.5	22.8	26.4	735	1347	24	41	74	83
5	8	13.3	17.1	−0.1	7.5	22.8	26.8	735	1347	24	59	74	131
10	4	4.4	17.1	−11.5	7.5	18.9	26.8	735	1347	24	59	74	131
15	2	3.4	17.1	−12.9	7.5	18.9	26.8	735	1347	24	59	74	131
20	2	3.4	17.1	−12.9	7.5	18.9	26.8	735	1347	24	59	74	131
25	6	11.6	17.1	−0.3	7.5	19.4	26.8	735	1347	24	41	74	131
30	10	11.6	16.6	−0.3	7	19.4	26.4	471	1347	24	41	74	83
35	9	15.7	16.6	5	7	24.7	26.8	1096	1347	24	41	79	131
40	5	11.6	17.1	−0.3	7.5	19.4	26.8	735	1347	24	41	74	131
45	5	11.6	17.1	−0.3	7	20	26.8	735	1347	24	41	74	131
50	4	11.6	17.1	−0.1	7.5	19.4	26.8	735	1347	24	41	74	61
55	3	11.6	18.4	−0.3	12.5	19.4	28.8	338	1577	2	41	47	175
60	3	5.3	17.6	−7.5	9.7	17.4	26.4	373	1548	2	43	6	83
65	6	11.6	17.1	−0.3	7.5	19.6	26.8	735	1347	24	41	74	131
70	4	11.6	17.6	−0.3	9.7	19.4	26.4	471	1548	2	41	6	83
75	6	11.6	17.1	−0.3	7.5	21.7	26.4	735	1347	24	41	74	83
80	8	11.6	18.4	−0.3	9.1	19.6	28.6	576	1520	8	41	70	141
85	8	15.6	18.4	5	12.5	24.7	28.6	823	1577	8	41	79	175
90	10	11.6	17.1	−0.3	7	19.4	26.8	735	1347	24	41	74	131
95	13	11.6	17.1	−0.3	7.5	21.7	26.4	735	1347	24	41	74	83
100	21	15.6	17.1	5	7	24.7	26.4	823	1347	24	41	79	131
105	14	15.6	17.1	5	7.5	24.7	26.8	823	1347	24	41	108	131
110	11	11.6	17.1	−0.1	7.5	22.8	26.8	735	1347	24	41	74	131
115	7	11.6	17.1	−0.3	7.5	19.4	26.8	735	1347	24	41	74	131
120	19	11.6	17.1	−0.3	7	21.7	26.4	735	1347	24	41	74	83
125	17	11.6	16.3	−0.3	7.5	21.7	26.8	735	1347	24	41	108	131
130	16	15.6	17.1	5	7.5	24.7	26.4	823	1347	24	41	79	83
135	29	15.6	17.1	5	7.5	24.7	26.4	1096	1347	24	41	74	131
140	26	15.6	17.1	5	7.5	24.7	26.4	1096	1347	24	41	74	131

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