

Silicification and organic matter preservation in the Anisian Muschelkalk: Implications for the basin dynamics of the central European Muschelkalk Sea

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Anisian Muschelkalk carbonates of the southern Germanic Basin containing silicified ooidal grainstone are interpreted as evidence of changing pH conditions triggered by increased bioproductivity (marine phytoplankton) and terrestrial input of plant debris during maximum flooding. Three distinct stages of calcite ooid replacement by silica were detected. Stage 1 reflects authigenic quartz development during the growth of the ooids, suggesting a change in the pH–temperature regime of the depositional environment. Stages 2 and 3 are found in silica-rich domains. The composition of silica-rich ooids shows significant Al₂O₃ and SrO but no FeO and MnO, indicating that late diagenetic alteration was minor. Silicified interparticle pore space is characterized by excellent preservation of marine prasinophytes; palynological slides show high abundance of terrestrial phytoclasts. The implications of our findings for basin dynamics reach from paleogeography to cyclostratigraphy and sequence stratigraphy, since changes in the seawater chemistry and sedimentary organic matter distribution reflect both the marine conditions as well as the hinterland. Basin interior changes might overprint the influence of the Tethys Ocean through the eastern and western gate areas. Stratigraphically, such changes might enhance marine flooding signals. Ongoing research needs to address the complex interaction between an intracratonic basin and an open-ocean system by comparing local and regional biotic and abiotic signals.

Keywords: silicification, organic matter, Muschelkalk, Anisian, Germany

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Introduction

Muschelkalk carbonates of the Triassic Germanic Basin, a peripheral basin of the western Tethys Ocean, cover large parts of central Europe. During the Anisian, the basin was bordered by landmasses and open to the Tethyan shelf by three tectonically controlled gates in the south and southeast: the East Carpathian, Silesian–Moravian, and Western Gates (Fig. 1). The East Carpathian Gate was already active in the Late Induan, the Silesian–Moravian Gate opened in the Olenekian (Szulc 1999, 2000), and the westernmost communication to the Tethys was effective during the Anisian (Feist-Burkhardt et al. 2008a; Götz and Gast 2010). Major transgressive phases are

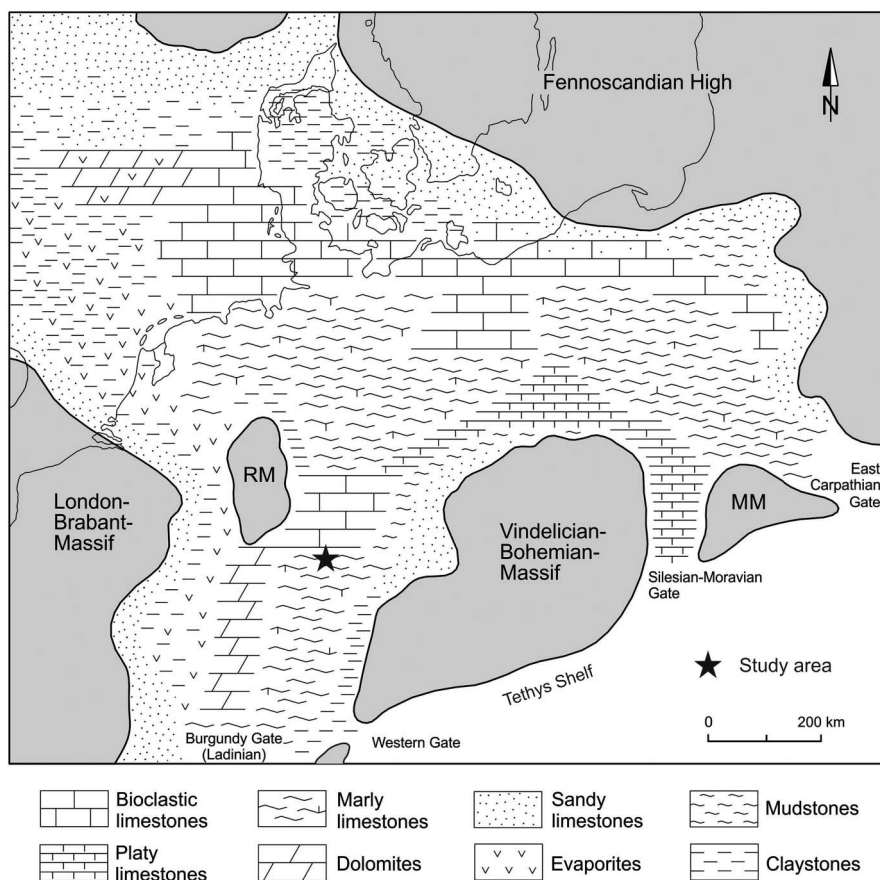


Fig. 1
 Paleogeography of the Lower Muschelkalk Basin during Pelsonian times (from Götz and Feist-Burkhardt 2012, based on Ziegler 1990) and location of the study area. MM – Małopolska Massif; RM – Rhenish Massif

recognized by phytoplankton peaks (Götz and Feist-Burkhardt 2000, 2012), documenting the interaction between a restricted, intracratonic basin and an open-ocean system. Furthermore, changes in the basin interior are well displayed in lateral phytoplankton distribution patterns, pointing to a stratified water body in the basin center and well-oxygenated marginal and gate areas. However, beside the interpretation of distinct spatial patterns of phytoplankton assemblages reflecting the basin configuration, the effect of increased bioproductivity and sedimentary organic matter supply from the basin's hinterland on the seawater's pH conditions has not yet been addressed.

The new data presented herein on silicification of ooidal grainstone during maximum flooding and organic matter preservation in the Lower Muschelkalk of Franconia provide new insights into the basin dynamics of the Anisian Muschelkalk Sea and add important new biogeochemical parameters to constrain an early Mesozoic epicontinental ocean history.

Stratigraphy

The lithostratigraphic subdivision of the Lower Muschelkalk dates back to the 19th century (Bornemann 1886, 1888) and the current stratigraphic units following the international nomenclature (Hedberg 1976; Salvador 1994) using formations, members, and beds (Fig. 2) were introduced by Hagdom et al. (1993). The biostratigraphic framework is based on conodonts (Kozur 1974) and palynomorphs (Heunisch 1999). Radiometric dating lags behind due to the lack of volcanic ash layers. A sequence stratigraphic framework was provided by Aigner and Bachmann (1992) and revised by Szulc (1999); cyclostratigraphy was carried out by Götz (1996, 2002, 2004). The Muschelkalk stratigraphy of Lower Franconia was studied by Wilczewski (1967) and Hagdom et al. (1987); a cyclostratigraphic interpretation is given in Götz and Wertel (2002).

Material and methods

The Terebratel Beds of the Lower Muschelkalk are well exposed in natural outcrops as well as in abandoned and active quarries of Lower Franconia (Götz and Keller 1998) and were intersected in boreholes for raw material exploration (Götz and Ruckwied 2005). Here, we present sedimentological and palynological data from the Lower Muschelkalk Terebratel Beds (Terebratelbank Member, Pelsonian) of the Karlstadt section, exposed at the Klettergarten north of Karlstadt, southern Germany (Fig. 3). Sedimentological data for the study of silicification of ooidal grainstone originated from analyses of polished slabs and thin sections. Palynological samples for investigating the phytoplankton assemblage and sedimentary organic matter content were prepared using standard palynological processing techniques as described in

Anisian (pars)		Chrono- stratigraphy	Litho- stratigraphy	
		Illyr. (pars)		
Bithynian	Pelsonian	Lower Muschelkalk		Schaumkalk- bank Mb.
Röt L. M.				Wellenkalk-3 Member
				Terebratel- bank Mb.
				Wellenkalk-2 Member
				Oolithbank Mb.
Wellenkalk-1 Member				Wellenkalk-1 Member
		Grenz- gelbkalk Myophorien- schichten		

Fig. 2
Stratigraphy of the Lower Muschelkalk in Lower Franconia. L.M. – Lower Muschelkalk, Mb. – Member

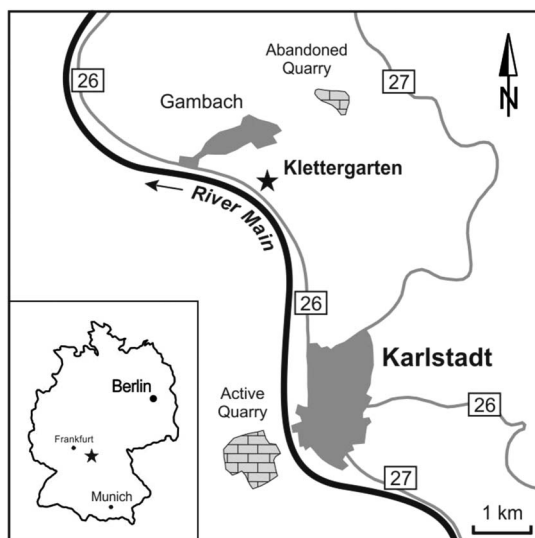


Fig. 3
Location of study area in southern Germany (Lower Franconia)

Vidal (1988). Electron probe micro-analyzer data acquisition [backscattered electron images, quantitative analysis, wavelength dispersive spectroscopy (WDS) element scans, line analysis, and WDS element maps] was performed at Rhodes University, Grahamstown, South Africa using a Jeol JXA 8230 Superprobe with four wavelength dispersive spectrometers. Analytical conditions employed were: acceleration voltage 15 kV, probe current 20 nA, dwell time for element mapping 200 ms and for line analysis 500 ms with 1 μm step. Mineral phases were analyzed with spot beam size (<1 μm). Eight elements were selected as most representative for the present study: Si, Al, K, Ca, Na, Fe, Mg, and Sr. The standards (st.) used for measuring the characteristic $K\alpha$ radiations were natural minerals: quartz st. for Si, orthoclase st. for Al and K, plagioclase An65 st. for Ca, albite st. for Na, fayalite st. for Fe, rhodonite st. for Mg, and celestine st. for Sr. The diffracting crystals used were: TAP for Si, Al and Mg; PETJ for K, large crystals with higher sensitivity TAPL for Na and Sr; PETL for Ca; and LiFL for Fe and Mn. The ZAF correction matrix was used for quantitative analysis of silicified ooids.

Results

The Terebratel Beds of the Karlstadt section are composed of bioclastic grainstone revealing distinct layers of ooidal grainstone (Fig. 4). In thin sections, these layers show silicification of ooids and interparticle silica cement (Fig. 5a–c).



Fig. 4
Polished slab of ooidal grainstone (Terebratel Bed), Karlstadt section (Lower Franconia)

Silicified interparticle pore space is characterized by preservation of prasinophytes (Fig. 5f). In palynological slides, these prasinophytes were identified as *Cymatiosphaera* sp. showing a typical reticulum (Fig. 5d and e).

Analysis of the silicified ooids was performed using high-resolution imaging and mapping of elements. The element map distribution shows three distinct stages of calcite ooid replacement by silica: Stage 1 (calcite stage), where small detrital quartz is present as dispersed grains in the central part of the ooid structure and/or as very discrete concentric rims of low concentration Si and Al (Fig. 6); Stage 2 (calcite–silica stage), where silica replaces certain concentric bands in the calcitic ooid structure, and where the concentric structure is broken by the silica band that is connected to the exterior of the ooid (Fig. 7); and Stage 3 (silica stage), where thin Ca-rich zones are still preserved in the structure of the ooid, and where most of it is replaced by silica (Fig. 8).

Figure 9 shows that Na and K have similar patterns. Furthermore, the analyzed transect (see section A–B in Fig. 9) shows similar behavior for the following pair of elements: Ca–Mg, Si–Sr, and Na–K. Other elements, such as Fe, Mn, and Al, do not show any relevant variations along the A–B section and are therefore not shown here. The composition of silica-rich ooids (Table 1) shows significant Al_2O_3 and SrO but no FeO and MnO.

Discussion

The Terebratelbank Member of the Lower Muschelkalk was previously identified as recording a third-order maximum flooding zone by independent sedimentological, paleontological, and geochemical studies (Aigner and Bachmann 1992; Szulc 1999, 2000; Rameil *et al.* 2000; Götz *et al.* 2003, 2005; Conradi *et al.* 2007; Feist-Burkhardt *et al.* 2008a; Götz and Török 2008; Götz and Lenhardt 2011). High primary bioproductivity during maximum flooding is reflected in phytoplankton peaks within the basin and the gate areas connecting the Germanic Basin with the Tethys shelf.

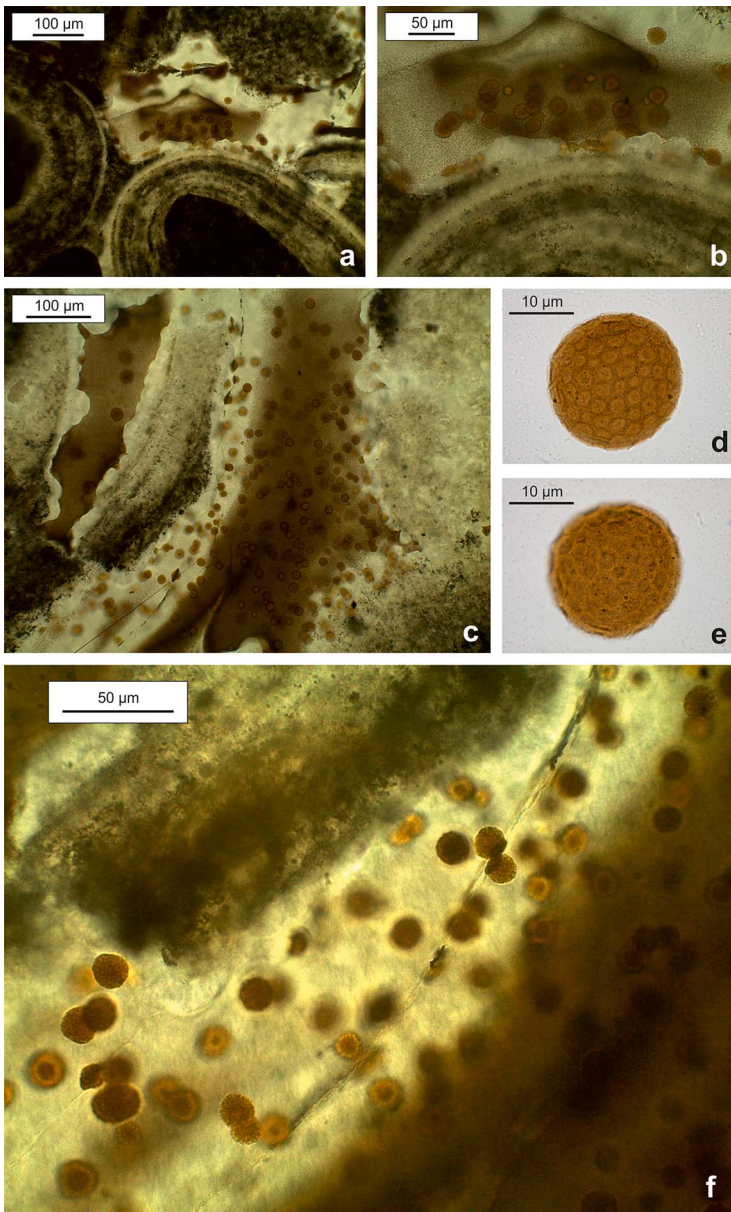


Fig. 5
Silicified ooids and prasinophytes preserved in interparticle silicified pore space. (a) Overview showing ooids and prasinophytes in thin section. (b) Detail of (a) showing the preservation of prasinophytes in the silicified pore space. (c) Massive occurrence of prasinophytes in silicified interparticle pore space. (d) and (e) *Cymatiosphaera* sp. (HF/LF). (f) Detail of (c) highlighting prasinophytes (*Cymatiosphaera* sp.) in silicified interparticle pore space

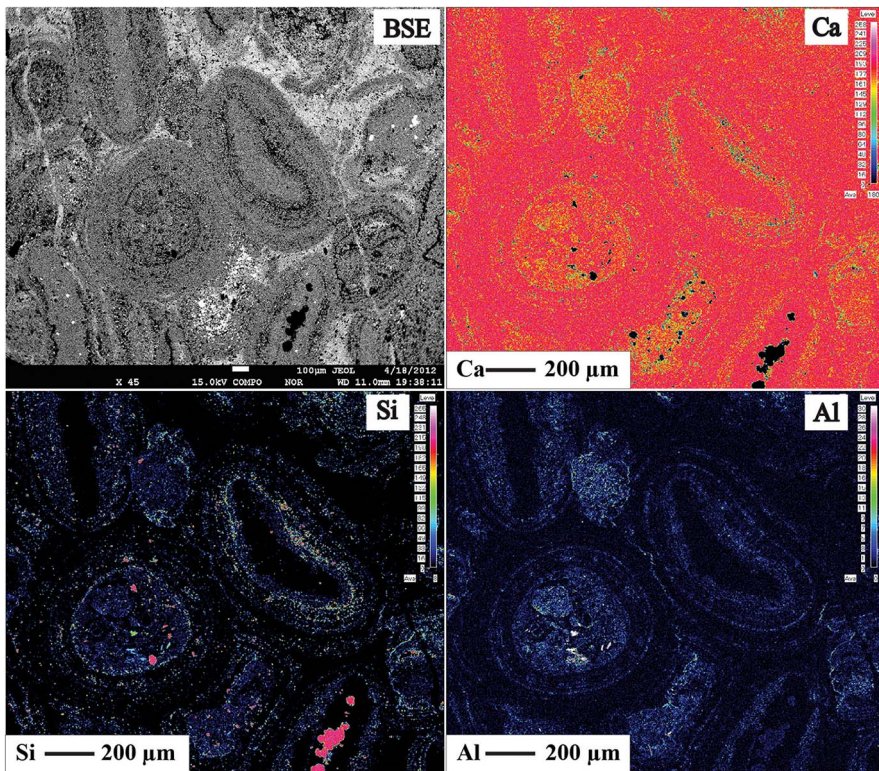


Fig. 6
Stage 1 of calcite ooid replacement by silica: detrital quartz is present as dispersed grains in the central part of the ooid and/or as very discrete concentric rims of low concentration Si and Al

The well-oxygenated gate areas and basin margins are dominated by phytoplankton of the *Micrhystridium* group, whereas the central basin parts show peak abundance of *Veryhachium* spp. and prasinophytes including *Tasmanites* spp. and *Cymatiosphaera* spp. (Rameil et al. 2000; Götz and Feist-Burkhardt 2012). Fine-grained, pyrite-bearing mudstone and a high abundance of prasinophytes in the central basin point to a stratified water column during deposition of the Terebratelbank Member. While high input of phytoclasts and plant debris from the landmasses bordering the Germanic Basin occurs throughout the Lower Muschelkalk, much higher amounts of land plant particles are detected in marginal and lagoonal settings (Götz et al. 2001).

Sources of silica in marine epicontinental basins are interpreted as biogenic (e.g., skeletal opal produced by radiolarians, diatoms, and siliceous sponges), volcano-genetic, and hydrothermal (DeMaster 1981; Packard et al. 2001; Flügel 2004). Another source is dissolved silica deriving from continental chemical weathering (Laschet 1984; Kump et al. 2000). However, extensive ferrallitic (humid tropical)

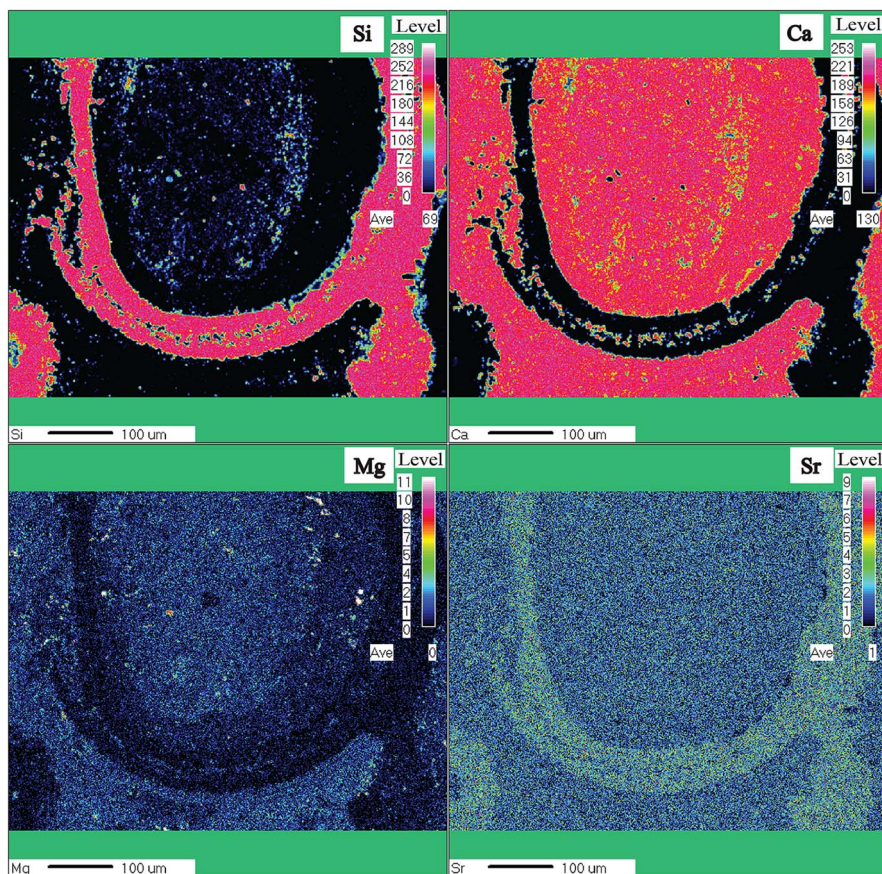


Fig. 7
Stage 2 of calcite ooid replacement by silica: silica replaces certain concentric bands in the calcitic ooid

weathering is necessary to dissolve silica and can be excluded for the overall arid to warm-temperate climate during Anisian times (Preto et al. 2010). Volcanic activity is not recorded during the Anisian of central Europe and limited to the Late Anisian–Ladinian of the western Tethyan realm (Budai and Haas 1997; Haas and Budai 1999; Szulc 2000; Feist-Burkhardt et al. 2008b; Kovács et al. 2011). Biogenic producers such as diatoms can be excluded, since they first occur in the Lower Cretaceous (Harwood et al. 2007). Radiolarians have not been reported from the shallow epicontinental Muschelkalk Sea and are still in the recovery phase after the P/T boundary event (De Wever et al. 2006); reef build-ups (e.g., siliceous sponges) are rare in the Lower Muschelkalk, including *Placunopsis* (bivalve) patch reefs in Germany (Hagdorn et al. 1999) and coral–sponge reefs in Poland (Hagdorn et al. 1999;

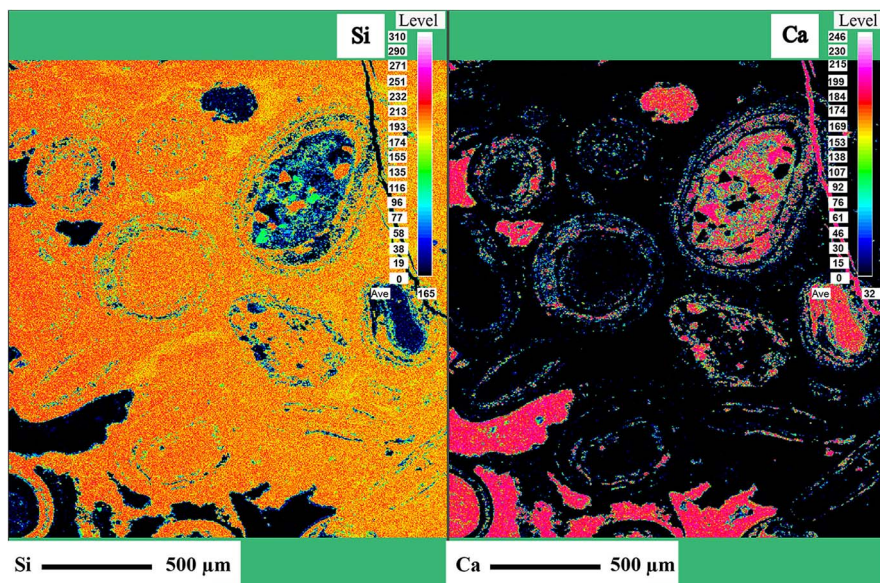


Fig. 8 Stage 3 of calcite ooid replacement by silica: thin Ca-rich zones are still preserved in the structure of the ooid, whereas most of it is replaced by silica

Szulc 2003). The best developed Pelsonian coral–sponge build-ups occur in Upper Silesia where they form bioherms of some 2–80 m across and several meters height (Szulc 2000). The hexactinellid sponges along with scleractinian corals gave rise to the oldest *in situ* reefs found in the western Tethys province (Szulc 2007). Further factors to be considered for the presence of silica in marine settings are the chemistry of the pore fluid, the pH value of the environment, the presence of clay minerals, and the amount of organic material (Flügel 2004).

Mechanisms for the replacement of carbonates by silica were discussed as follows: (1) local lowering of the pH by introducing CO₂ into the waters through respiration or by decomposition of organic matter; the lowering of pH would increase the solubility of calcite and silica would precipitate instead of the dissolved calcite (e.g., Siever 1962; Knoll 1985; Hesse 1989; Maliva and Siever 1989); (2) oxidation of hydrogen sulfide, reducing the pH at oxic/anoxic boundaries (Clayton 1986); (3) mixing of marine and continental waters, leading to dissolution of calcite and precipitation of silica (Knauth 1979); (4) mixing of saline lake waters with meteoric groundwaters (e.g., Nickel 1982) in continental environments. Part of the groundwater can be supersaturated with respect to quartz and undersaturated with respect to calcite; (5) microbial activity on sediment surfaces (Renaut et al. 1998) where the negatively charged OH and carboxyl groups on microbial surfaces would allow binding with silicic acid that can promote silicification.

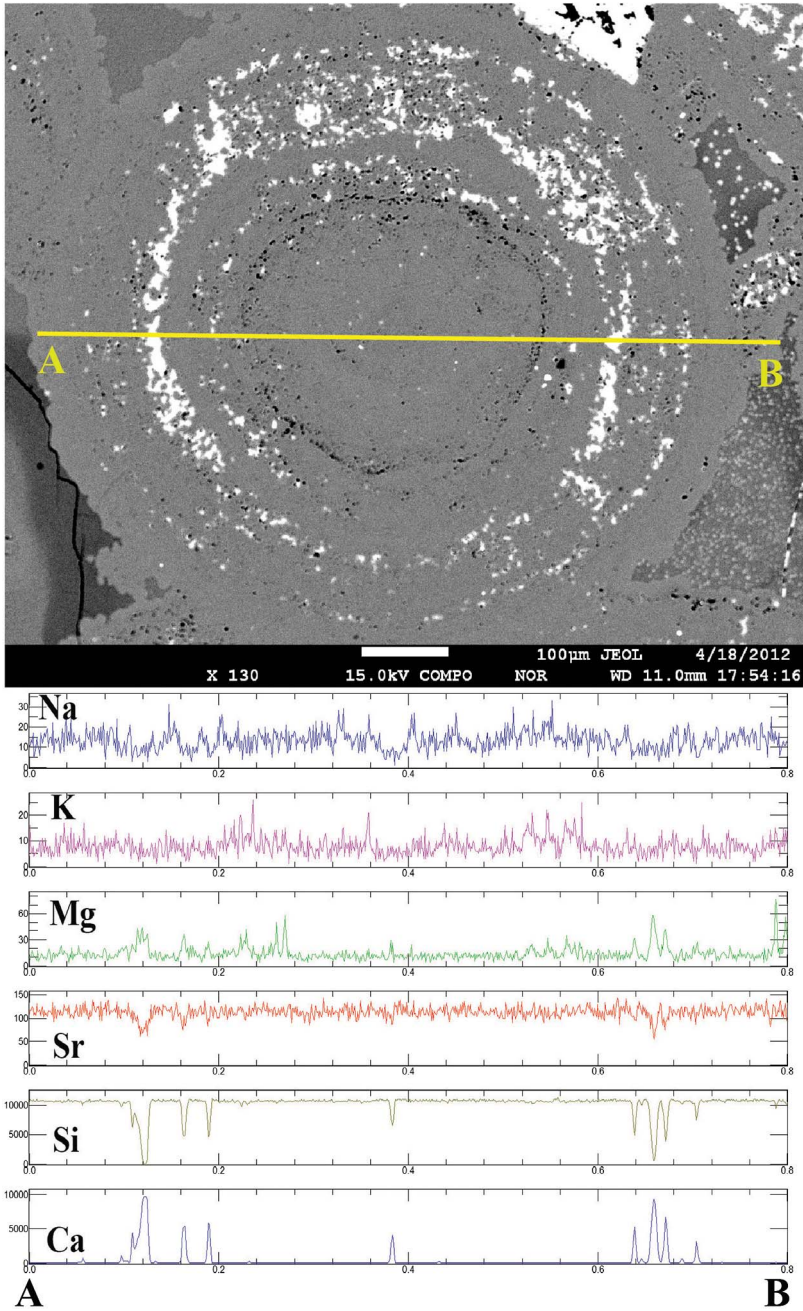


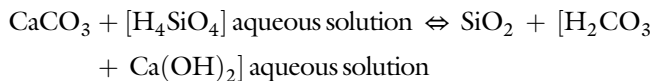
Fig. 9
Backscattered image of zoned ooid (Stage 3), with A–B compositional profile realized by WDS line analysis

Table 1
Average of 30 electron microprobe analyses of silica-rich ooids

Oxide	wt. %	StDev (%)	DL (ppm)
SiO ₂	98.399	0.06	70
Al ₂ O ₃	0.423	1.35	39
FeO	0.000	100	195
MnO	0.000	100	55
MgO	0.014	27.7	40
CaO	0.170	2.34	29
BaO	0.003	20.98	50
SrO	0.478	11.57	72
Na ₂ O	0.034	8.28	24
K ₂ O	0.048	4.46	18
Total	99.569		

StDev: analytical standard deviation 1 sigma; DL: detection limit

Maliva and Siever (1989) noted that none of the above mechanisms can explain why the volumetric rate of silica precipitation is equal to the calcite dissolution rate. However, assuming that the silicification occurs at the calcite–water interface, the replacement of calcite should be controlled by a quasi-isochoric metasomatic reaction. With pH increasing above 7, the silicic acid in aqueous solution gradually loses 1 or 2 proton(s) and forms H₃SiO₄[−] or [H₂SiO₄]₂[−]. In this speciation, a high concentration of silicic acid seems to be in equilibrium with quartz (the solubility of SiO₂ is high). With decreasing pH, the silicic acid gains the protons and becomes H₄SiO₄, which is a charge-balanced compound. It will react with calcite in the following manner:



In the studied ooidal grainstone of the Karlstadt section, Stage 1 of the calcite ooid replacement by silica most probably shows small grains of detrital quartz reprinting the nuclei of ooids. The low concentration silica present in the concentric bands of the ooid probably reflects authigenic quartz development during the growth of the ooids and might suggest a change in the pH–temperature regime of the depositional environment. Stages 2 and 3 are found in silica-rich domains.

The similar behavior for the element pairs Ca–Mg, Si–Sr, and Na–K suggests that the Si-rich fluid responsible for ooid replacement was also Sr-rich, and that the fluid

replaced Ca–Mg from the calcite. The MgO/CaO ratio is probably related to the initial Mg/Ca ratio in the replaced calcite.

The similar patterns of Na and K are probably related to the same fluid that was responsible for calcite replacement by silica. The absence of FeO and MnO in silica-rich ooids indicates that late diagenetic alteration was minor.

Partial and complete silicification of biogenic or abiogenic carbonate grains and chert formation have been described from Paleozoic to Cenozoic marine carbonate systems (e.g., Swett 1965; Zijlstra 1987; Martín Penela and Barragán 1995; Young et al. 2012). Reports on Muschelkalk chert date back to the 19th century (Seebach 1861; Sandberger 1864; Speyer 1875); it was first studied with respect to its paleogeographic significance by Trammer (1977) in Poland, where it was reported from Upper Silesia and the Holy Cross Mountains in papers dating back to the 1930s [for review see Kwiatkowski (2005) and references therein]. More recently, silicified oncoidal limestone and chert nodules were described from the Polish Muschelkalk by Kwiatkowski (2005). Chert nodules are interpreted as originating from early diagenetic limestone silicification. They occur within lagoonal and evaporitic settings in distinct horizons of the Lower Muschelkalk succession in Upper Silesia and the Holy Cross Mountains. The Pelsonian Terebratel Beds represent the only stratigraphic interval where no silicification is documented in the Polish Muschelkalk. In northern Switzerland, representing the western gate area during Anisian times, chert nodules are reported from dolomitic, partly stromatolitic limestone of the uppermost Anisian and at the Anisian–Ladinian boundary (Jordan 2016; Pietsch et al. 2016); however, no silicification occurs in the Pelsonian “Wellenmergel” (Kaiseraugst Formation).

The paleogeographic location of the studied Karlstadt section in the southern part of the Germanic Basin, with close landmasses in the northwest (Rhenish Massif) and southeast (Vindelician–Bohemian Massif), suggests high primary bioproductivity and high terrestrial input of plant debris during the deposition of the Terebratel Beds. This was observed in previous studies by the sedimentary organic matter content and phytoplankton assemblages of the Terebratel Beds in Lower Franconia (Götz and Ruckwied 2005). High organic matter content in a marine setting on the other hand has an impact on the pH of the water column which in turn favors silica precipitation. This effect is reflected in the Terebratel Bed samples from the Karlstadt section and demonstrates the high variability in seawater chemistry and basin dynamics of the Anisian Muschelkalk. In the southern basin, high organic matter content temporarily led to pH changes and silica precipitation and calcite replacement, whereas in the central part of the basin, a stratified water column developed, favoring the deposition of pyrite-bearing mudstone in the lower oxygen-depleted layer and prasinophyte blooms in the upper oxygenic layer (Rameil et al. 2000). Marginal sections close to the Rhenish Massif (Götz et al. 2001) show a much more diverse phytoplankton assemblage of acritarchs and prasinophytes, and a high phytoclast input from the hinterland; however, no silicified particles were encountered. The same applies for the well-oxygenated gate areas (Götz and Feist-Burkhardt 2012), where no silicification occurs during maximum flooding in the Pelsonian (Kwiatkowski 2005; Jordan 2016).

These different patterns in palynofacies composition seem to influence the pH conditions in the different parts of the basin, and thus strong local effects on the basin dynamics must be assumed. The complex interaction between the hydrodynamic regimes within the basin and gate areas, related to local differences in water depths (e.g., shoals, restricted bays), and times of lower or higher bioproductivity as well as lower or higher terrestrial influx of organic debris, seem to intensify local processes which in turn lead to local patterns in seawater chemistry. Stratigraphically, transgressive and maximum flooding phases are marked by increased bioproductivity; in the case of close landmasses, terrestrial input of plant debris as well as palynomorphs, especially wind-dispersed pollen grains, is high. Even slightly changing pH values, triggered by variations in organic matter content, are thus most probably responsible for silica precipitation and calcite replacement of non-biogenic components such as ooids in certain parts of the basin and at certain times. Independent of the mechanism for replacement of carbonates by silica, any consuming of carbonate will produce CO₂, which in turn triggers basin interior and regional paleoenvironmental changes. Ultimately, a complex superposition of local and regional effects seems to have caused distinct basinal patterns during Mid-Anisian times.

Conclusions

The present study of the Terebratel Beds adds another puzzle piece to decipher the complex basin dynamics of the Germanic Basin during Anisian times. The effect of increased bioproductivity (marine phytoplankton) and sedimentary organic matter supply from the basin's hinterland on the seawater's pH conditions has been so far overlooked. Changes in organic matter seem to have a strong influence on basin dynamics. Spatial basin interior changes might even overprint the influence of the Tethys Ocean through the eastern and western gate areas. Stratigraphically, such changes might enhance the marine flooding signal. Thus, silicified grainstone and sedimentary organic matter preservation are well suited as indicators of flooding phases and might add to the cyclostratigraphic and sequence stratigraphic interpretation of epicontinental seas.

To understand the complex interaction between an intracratonic basin and an open-ocean system during the early stage of the break-up of Pangea, integrated sedimentological–paleontological–geochemical studies, encompassing the western and easternmost Anisian Muschelkalk series in Spain and southeastern Europe, are needed in the ongoing research.

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