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Source: Paleontological Research, 26(3) : 301-313

Published By: The Palaeontological Society of Japan

URL: https://doi.org/10.2517/PR200029

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Paleontological Research, vol. 26, no. 3, pp. 301–313, July 1, 2022
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doi:10.2517/PR200029

Late Cretaceous diatoms (Bacillariophyta) from the Teshio-Nakagawa area, Hokkaido, northern Japan: Significance for their origin and biostratigraphy

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Received July 28, 2020; Revised manuscript accepted March 3, 2021

Abstract. Well-preserved marine diatoms are documented for the first time from authigenic carbonate rocks induced by cold methane (hydrocarbon) seepage in the Omagari Formation (latest Santonian to earliest Campanian in age, Late Cretaceous Epoch; around 83.6 Ma) of the upper part of the Yezo Group in the Teshio-Nakagawa area, northern Hokkaido (northern Japan). The diatom flora is rich in species of *Hemiaulus* and *Triceratium*, associated with a few other extinct diatom genera. An araphid genus (*Sceptroneis*) was also observed; this is one of the earliest fossil records of "pennate" (Bacillariophyceae) diatoms. Although valve ultrastructures have been mostly dissolved, the preservation of these diatoms is much better than that in the few previous reports of Cretaceous siliceous photosynthetic organisms from Japan and adjacent regions in the northwestern Pacific Ocean. Because of their common generic composition in the mid- to high-latitude regions in both hemispheres, diatoms are suggested to have experienced a global radiation by early Campanian time. Thus, our diatom records provide insights into the Late Cretaceous floral adaptive radiation around the northwestern Pacific margin, where the radiation history of diatoms is yet unclear. As *Gladius antiquus* was confirmed but *Basilicostephanus* species were absent from the present material, the flora from the Teshio-Nakagawa area is tentatively regarded as belonging to the *G. antiquus* Concurrent Range Zone, an interval extending from an undetermined Late Cretaceous Epoch to the latest Santonian Age. However, this floral correlation is inaccurate because our materials are latest Santonian to earliest Campanian in age, as dated by the molluscan (ammonoids and inoceramids) biostratigraphy. Hence, further research is required to clarify the sensitivity of different chronological proxies and the stratigraphic ranges of age-diagnostic diatoms in different geographic provinces.

Keywords: authigenic carbonate rocks, biostratigraphy, cold methane seepage, diatoms, Late Cretaceous, northwestern Pacific Ocean

Introduction

The diatoms are one of the most successful, and thus important, primary producers of the Cenozoic hydrospheric ecosystem (Armbrust, 2009). Their rise and early history, inferred to date back to the latest Paleozoic to early Mesozoic eras on the basis of fossil and molecular evidence (Rothpletz, 1896, 1900; Kooistra and Medlin, 1996), is, however, still unclear because of the paucity of fossil diatom records and the poor preservation of early diatoms as a result of severe diagenesis in the sediment.

A silicoflagellate specimen from a Cretaceous carbonate concretion from an uncertain locality in the central area of Hokkaido, northern Japan, was informally reported as the first fossil record of a siliceous photosynthetic organism in the Japanese Archipelago (Koizumi, 1979; Sawamura, 1979; Sawamura and Otowa, 1979). In previous studies, diatoms were unexpectedly encountered in residues after extraction of radiolarian shells from Upper Cretaceous materials. These occurrences, illustrated by means of scanning electron microscope images, have been formally presented from several localities across Japan. Those Late Cretaceous diatoms have been documented from the Osoushinai Formation, Hokkaido Prefecture, northern Japan (Iwata *et al.*, 1998), the Shoya Formation, Nagano Prefecture, central Japan (Takahashi *et al*., 1999), and the Hiraiso Formation, Ibaraki Prefecture, also central Japan (Ando *et al*., 2014). Unfortunately, further taxonomic investigations of those Late Cretaceous diatoms have not been carried out because the material is very poorly preserved or recrystallized; thus, it is difficult to precisely describe and identify them. However, we recently found well-preserved diatoms in Upper Cretaceous deposits in the Teshio-Nakagawa area, Hokkaido, and preliminarily documented the flora (Shimada *et al*., 2013). The flora was obtained from authigenic carbonate rock bodies formed as a result of cold methane seepage and associated with an *in situ* chemosynthetic fauna (Hikida *et al*., 2003; Jenkins and Hikida, 2011). In this paper, we extend the work of Shimada *et al*. (2013) by presenting additional floral data from a new locality, also in the Teshio-Nakagawa area, (Jenkins *et al*., 2007; Jenkins and Hikida, 2011), and discuss the significance of for their origin and biostratigraphy.

Geological setting

The Yezo Group, which consists mostly of Middle to Upper Cretaceous marine deposits, is widely exposed in the Teshio-Nakagawa area, Hokkaido, northern Japan. The unit consists chiefly of Cretaceous, and some Paleocene, forearc-basin deposits that are distributed zonally and meridionally in the axial ridge area of Hokkaido and northward to Sakhalin, Far Eastern Russia, as a result of subduction along the western margin of the Pacific Ocean (e.g. Ueda, 2016). The Yezo Group is characterized by very abundant molluscan megafossils (ammonoids and inoceramids), which are useful for construction of a refined biostratigraphy (e.g. Toshimitsu *et al.*, 1995; Takahashi *et al*., 2003).

In the upper part of the Yezo Group, the Omagari Formation is mainly composed of alternations of turbiditic sandstones/mudstones, intercalated with mudstones, sandstones, and conglomerates that were probably deposited on the continental shelf margin to slope (Figure 1). The thickness of the formation is estimated to be 550 to 810 m, becoming thinner northward in the Nakagawa Town area. The formation conformably overlies the Nishichirashinai Formation and underlies the Osoushinai Formation. Trace fossils, fragmented carbonaceous material, and molluscan shells sometimes occur in the Omagari Formation, but their preservation is generally poorer there than in the overlying and more fossiliferous Osoushinai Formation. *Gaudryceras tenuiliratum* (ammonoid) and *Inoceramus* (*Platyceramus*) *japonicus* (inoceramid) were found in the formation and are chronology indicators for the latest Santonian to earliest Campanian in age (*ca.* 83.6 Ma, Okamura, 1977; Toshimitsu *et al*., 1995; Takahashi *et al*., 2003; International Commission on Stratigraphy [ICS], 2020).

Two outcrops of fossiliferous carbonate rocks have been discovered hosted in slightly different horizons within the middle to upper part of the Omagari Formation (Figures 1 and 2; Hikida *et al*., 2003; Jenkins *et al.*, 2007; Jenkins and Hikida, 2011). The lower site, the Omagari Carbonate Islet Site (OCIS; “carbonate lens”, Hikida *et al*., 2003; “Omagari hydrocarbon seep carbonate”, Kaim *et al*., 2009), is exposed (*ca.* 10 m × 6 m in plain view) as a lens-like body situated in the channel of the Abeshinai-gawa River (Figure 2; 44°39′26″N, 142°2′25″E). This carbonate rock body is divided into two different zones—marginal brecciated and core boundstone facies—on the basis of lithology. The marginal brecciated facies occurs at the edge of the body, consists mainly of dark gray muddy carbonate breccia, and is rich in chemosynthetic infaunal molluscan shells in addition to small ammonoids. The core boundstone facies is composed of light grayish carbonate rocks with a dominance of exceptionally well-preserved vestimentiferan worm tubes and it lacks clastic material. The occurrence of such chemosynthetic biota and the very negative isotopic δ^{13}C ratios of the carbonate from both lithofacies ($\delta^{13}C = -41‰$ to $-45‰$ vs. Pee Dee Belemnite [PDB]) suggest that the OCIS rock body probably formed authigenically by a cold methane seepage process related to microbial sulfate reduction and anaerobic methane oxidation (Hikida *et al*., 2003; Jenkins and Hikida, 2011). Unfortunately, information on the stratigraphic relationship between the host deposits and the carbonate rock body is difficult to obtain because ongoing erosion by the Abeshinai-gawa River has caused the islet to become fully isolated in the channel during the past few decades (Figure 2).

In the upper horizon of the Omagari Formation, at a site 3 km downstream of the OCIS, four smaller carbonate rock bodies (maximum diameter *ca.* 0.5 m) were found exposed inside the channel of the Abeshinai-gawa River, surrounded by dispersed tiny carbonate concretions in the host sediments (Figures 1 and 2; Yasukawa Site, YS; “Yasukawa seep”, Jenkins *et al*., 2007; “Yasukawa seep carbonate”, Kaim *et al*., 2009; 44°40′37″N, 142°1′27″E). As at the OCIS, the carbonate bodies are divided into two lithological units. The lower facies consists dominantly of dark grayish micritic and clastic to tuffaceous layers, whereas the upper unit exhibits the networked radiaxial calcite rim-cementation texture typical of a biogenic or thermogenic methane origin. The upper facies is fossil-

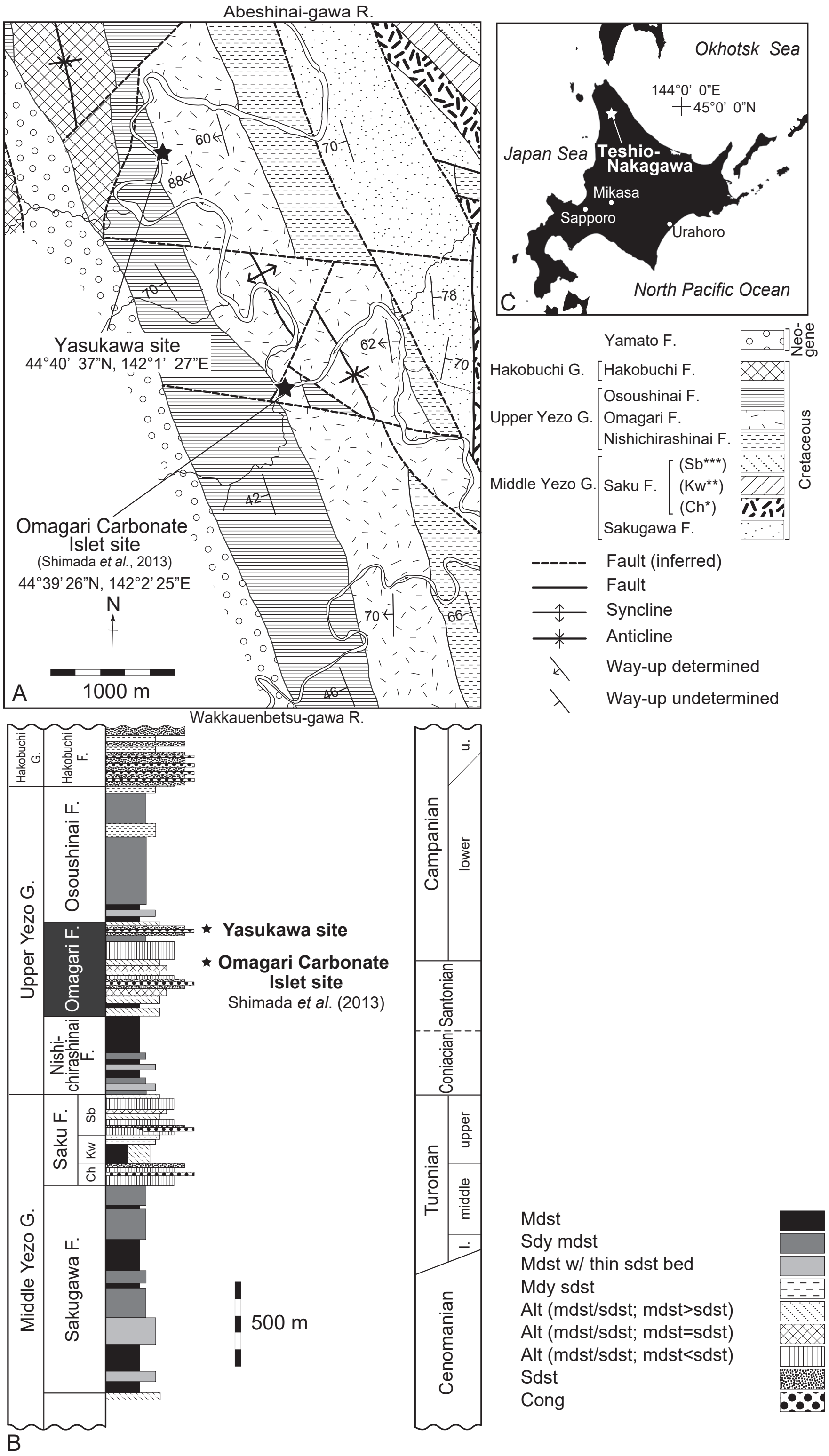

Figure 1. **A**, geological map of the studied area involving two sample localities; **B**, generalized columnar section. Both illustrations are modified after Takahashi *et al.* (2003). Ch*, Chirashinai-gawa River Sandstone Member; Kw**, Kyowa Mudstone Member; Sb***, Saku-bashi Bridge Alternating beds of sandstone and mudstone Member, Mdst, mudstone, Sdst, sandstone, Cong, conglomerate and Alt, alternation. **C**, Locality of the Teshio-Nakagawa area, Hokkaido, northern Japan.

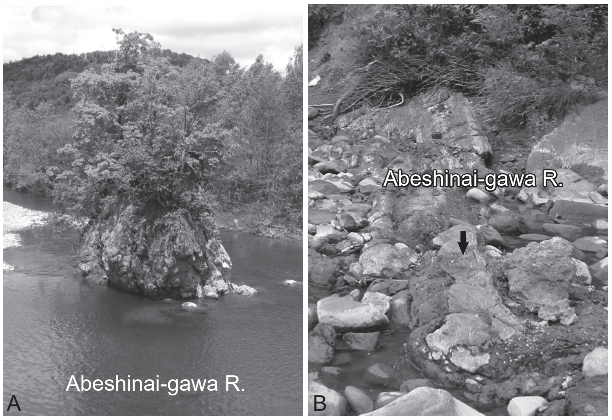

Figure 2. Photographs of the carbonate rock bodies studied in the present investigation. **A**, Omagari Carbonate Islet Site (OSIS); **B**, Yasukawa Site (YS). An arrow in the panel B indicates the rock body investigated in this paper.

iferous, with abundant chemosynthetic gastropods and bivalves (Jenkins *et al.*, 2007). Vestimentiferan worm tubes appear to be absent from the YS. Very negative δ^{13}C values (as low as −43.5‰ vs. PDB) were recorded from these carbonate matrices. Jenkins *et al*. (2007) concluded that the YS formed by cold methane seepage, as did the OCIS body, and that the distribution pattern of the chemosynthetic fauna and lithology at the YS were dependent on the methane discharge flux, i.e. the distance from the center of methane outgassing (Jenkins *et al.*, 2007). Biomarkers typical of anaerobic methane oxidizing archaea have been detected in the carbonate of the rock bodies (Jenkins *et al*., 2008).

Material and methods

Two rock samples from the core facies of the two carbonate bodies (more central; the boundstone facies at the OCIS; samples OCIS-CR and YS-CR) and two samples from the marginal facies were studied (samples OCIS-MR and YS-MR). Diatoms were extracted and concentrated from these carbonate materials by the following protocol; 5% formic acid and 15% hydrogen peroxide were sequentially added to a few grams of the sample at room temperature to gently remove carbonate and organic matter, respectively. When the reaction had finished, the generated suspension was neutralized by repeated decantation with purified water. UV adhesive or styrax was used as the mounting medium to prepare permanent slides. As fossil specimens of the diatoms usually occur fragmented in sedimentary rocks, a counting is conventionally performed when a halved piece of the valve is encountered under the light microscope; in case of an intact valve, two counts are given. In this study, at least 200 valves (more than 400 counts) were checked along traverses across each coverslip using oil immersion and 1000× magnification under a light microscope (OLYMPUS BX51). The count data were converted into percent abundances. In addition, the full areas of the coverslips of dozens of permanent slides of the four samples were observed to locate further well-preserved specimens for taxonomy. Taxa with very low abundance or composed of specimens too fragmented and difficult to identify were summed up as spp. In this paper, we compiled these extensively obtained data and those of Shimada *et al*. (2013) by checking additional areas of the permanent slides.

Photomicrographs of the diatoms were taken with a digital camera (OLYMPUS DP27 or CANON EOS X7). Light microscopic observation of thin-section slides of carbonate prepared from the core boundstone lithofacies at the OCIS was also performed to check the mode of occurrence of the diatoms. Scanning electron microscope (SEM) observation (JEOL JSM-6390 at Akita University or JEOL JSM-6390LV at the National Institute of Advanced Industrial Science and Technology; AIST) was also conducted.

Results

Diatoms were recognized in the samples from both carbonate rock bodies. Material originating from the core areas (OCI-CR and YS-CR) was more abundant and better preserved than that from the marginal lithofacies (Figures 3 and 4, Table 1). Specimens were rarely detected in the thin-section slides derived from the OCIS under a light microscope (Figure 5), and they had generally suffered from fragmentation; cribra and ends of long horns/processes are present, but thinly silicified structures are mostly lost. Silicoflagellates and chrysophyte cysts were commonly associated.

In the present study, *Cortinocornus rossicus* (Pantocsek) Glezer, *Hemiaulus polymorphus* v. *frigida* Grunow, *Hemiaulus* sp. A cf. *antiquus*, *Triceratium schulzii* Jousé sensu Strelnikova (1974), *Costopyxis schulzii* (Steineck and Schulz) Glezer, *Anuloplicata ornata* (Grunow) Glezer, and *Gladius antiquus* Forti and Schulz and its variety *G. antiquus* v. *tenuis* Gersonde and Harwood dominated in the samples (Table 1). As for other Late Cretaceous fossil assemblages reported in previous studies (e.g., Strelnikova, 1974), extinct or ancient diatom genera belonging to Hemiaulales, Triceratiales, or Melosirales predominated with high percent abundances. *Sceptroneis* was also observed as the one of the earliest fossil records of araphid genera (Figure 4, Table 1). Of the dominant species, *Hemiaulus* sp. A cf. *antiquus* and *Triceratium* sp. A (Figure 3) are unknown in previous Cretaceous

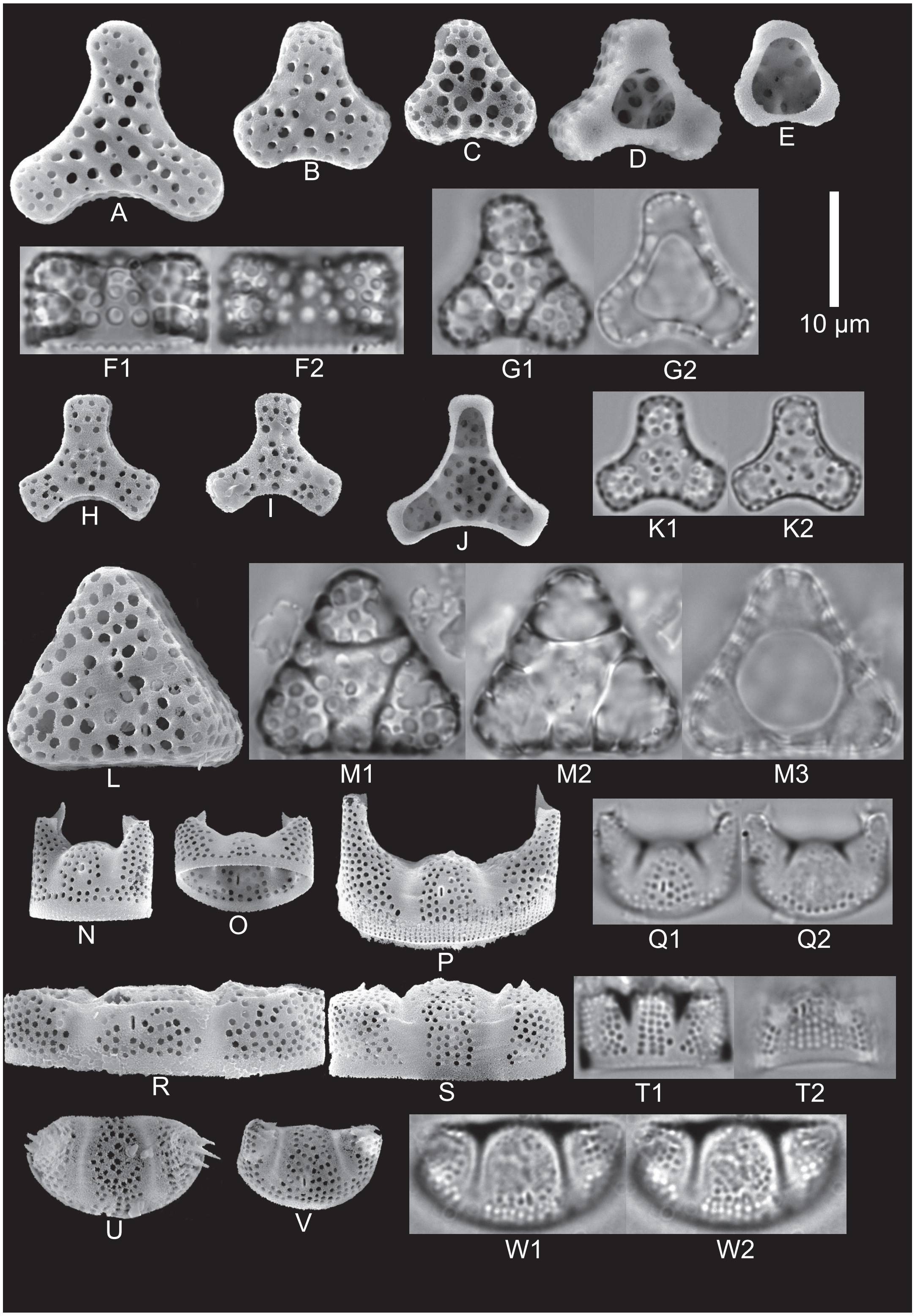

Figure 3. Photomicrographs of the Late Cretaceous diatoms from the Omagari Formation, Teshio-Nakagawa area, Hokkaido, northern Japan. **A**–**G**, *Triceratium schulzii* Jousé sensu Strelnikova (1974); **H**–**K**, *Triceratium* sp. A: **L** and **M**, *Triceratium planum* Strelnikova ?; **N**–**Q**, *Hemialus* sp. A cf. *H. antiquus* Jousé; **R**–**T**, *Hemiaulus* sp. D; **U**–**W**, *Hemialus* sp. E. Specimens C, D, M and W are from OCIS-CR. Others are from YS-CR. Specimens F, G, K, M, Q, T and W were taken in different focus by LM.

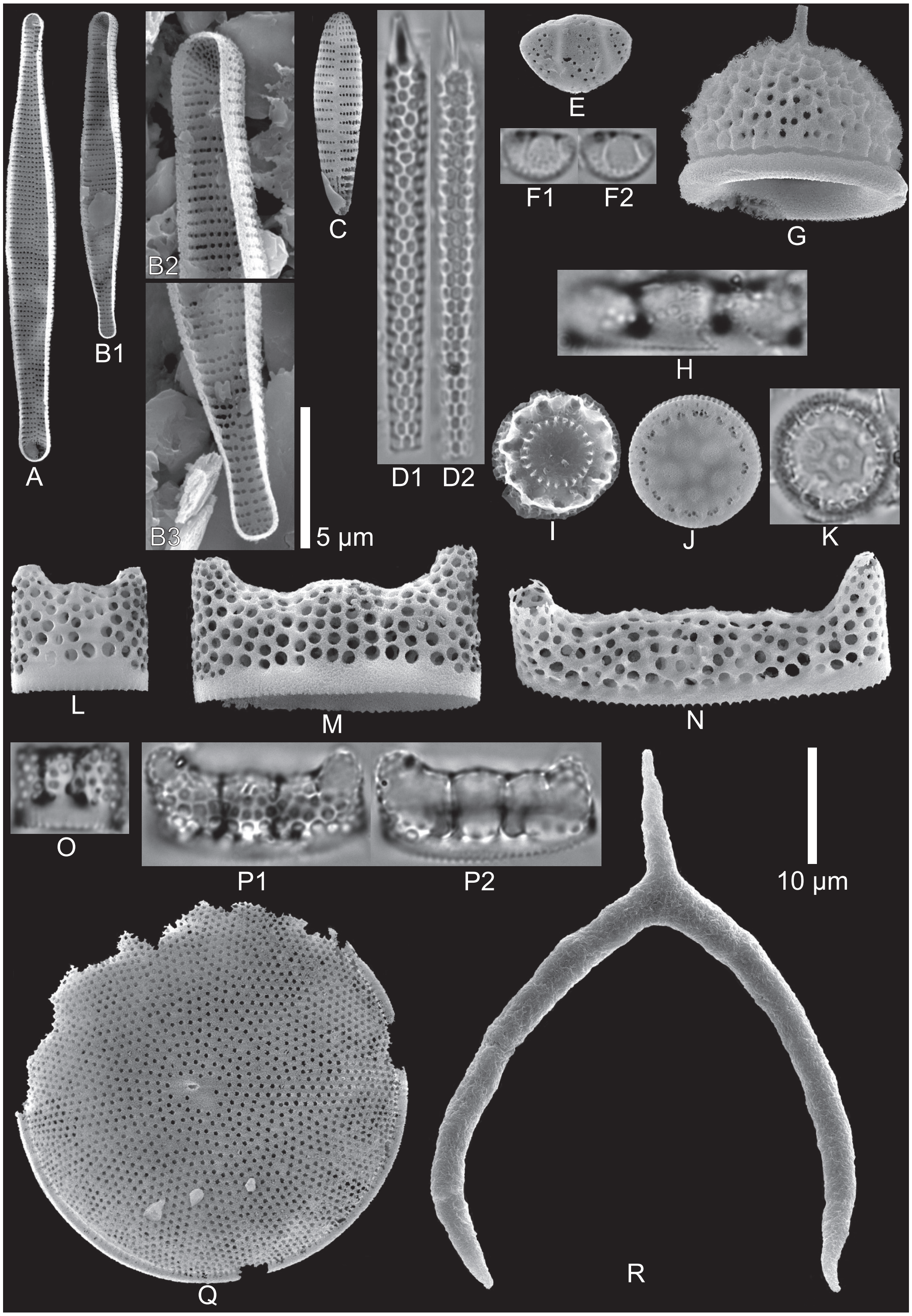

Figure 4. Photomicrographs of the Late Cretaceous diatoms from the Omagari Formation, Teshio-Nakagawa area, Hokkaido, northern Japan. **A** and **B**, *Sceptroneis* sp. A; **C**, *Sceptroneis* sp. C; **D**, *Gladius antiquus* v. *tenuis*; **E** and **F**, *Biddulphia sparsepunctata* Hajós and Stradner; **G**, *Costopyxis schulzii* (Steinek and Schulz) Glezer; **H**, *Anaulus sibericus* Strelnikova; **I**, *Paralia crenulata* (Grunow) Glezer; **J** and **K**, *Anuloplicata ornata* (Grunow) Glezer; **L**–**P**, *Cortinocornus rossicus* (Pantocsek) Glezer; **Q**, *Stellarima steinyi* (Hanna) Hasle and Sims (resting spore); **R**, *Lyramula furcata* Hanna (silicoflagellate). Specimens G, K, L, M and O are from OS-CR. Others are from YS-CR. Scale bar for specimens 2b and 2c indicates 5 µm. Specimens D, F and P were taken in different focus by LM.

Table 1. Late Cretaceous diatoms from the Omagari Formation in the Teshio-Nakagawa area, Hokkaido, northern Japan.

Sample	OCI-CR	OCI-MR	YAS-CR	YAS-MR
Abundance	Moderate	Few	Moderate	Very few
Preservation	Moderate	Poor	Good	Very poor
Diatom taxon				
Actinoptychus tenuis Strelnikova			*	
Anaulus sibericus Strelnikova	0.23	0.24	0.93	
Anuloplicata ornata (Grunow) Glezer	8.49	18.12	3.73	
Benetorus sp.	*	0.24	*	
Biddulphia sparsepunctata Hajós and Stradner			*	
Cortinocornus rossicus (Pantocsek) Glezer	18.12	21.41	25.64	
Costopyxis schulzii (Steinek and Schulz) Glezer		0.71	3.50	
Euodiella sp.			0.47	
Gladius antiquus Forti and Schulz	*	*	*	
G. antiquus v. *tenuis* Gersonde and Harwood	1.38	1.88	1.86	
Hemiaulus altus Hajós and Stradner			0.93	
H. elegans (Heiberg) Grunow		0.24		
H. polymorphus v. *frigida* Grunow	4.59	2.12	2.10	
Hemiaulus sp. A cf. *H. antiquus* Jousé	32.57	16.94	15.15	*
Hemialus sp. B cf. *H. kittonii* Grunow	2.98	0.71	2.80	
Hemiaulus sp. C cf. *H. polymorphus* Grunow	5.96	4.00	5.59	
Hemiaulus sp. D	5.28	6.59		
Hemiaulus ? sp. E	0.92	0.24		
Hemiaulus ? sp. F	2.06	2.12	4.43	
Hemiaulus spp.	6.42	5.41	7.69	
Medlinia sp.			*	
Paralia crenulata (Grunow) Glezer	0.23	1.41	0.47	
Pterotheca evermanni Hanna	*		*	
Pterotheca spp.		0.94		
Pyxidicula weyprechtii Grunow			0.23	
Sceptroneis grunowii Anissimowa		0.24	0.47	*
Sceptroneis sp. A			*	
Sceptroneis sp. B			*	
Sceptroneis sp. C			5.59	
Sheshukovia sp.		0.24	0.23	
Stellarima steinyi (Hanna) Hasle and Sims	*		*	*
Stephanopyxis turris (Greville and Arnott) Ralfs	1.83	3.53	0.93	
Thalassiosiropsis? sp.	*	0.47		
Triceratium indefinitum (Jousé) Strelnikova	0.69	0.24	3.50	*
T. schulzii Jousé sensu Strelnikova (1974)	2.06	2.82	6.53	*
T. planum Strelnikova	*		0.23	
Triceratium sp. A	5.05	4.71	1.63	
Miscellaneous resting spores	0.46	1.88	3.26	*
Genus and species indet. A		0.71	0.70	
Genus and species indet. B	0.23	0.24	*	
Genus and species indet. C		0.47	0.93	
Genus and species indet. D	*		*	
Miscellaneous	0.46	1.18	0.47	
Total counted	100	100	100	–

* Present

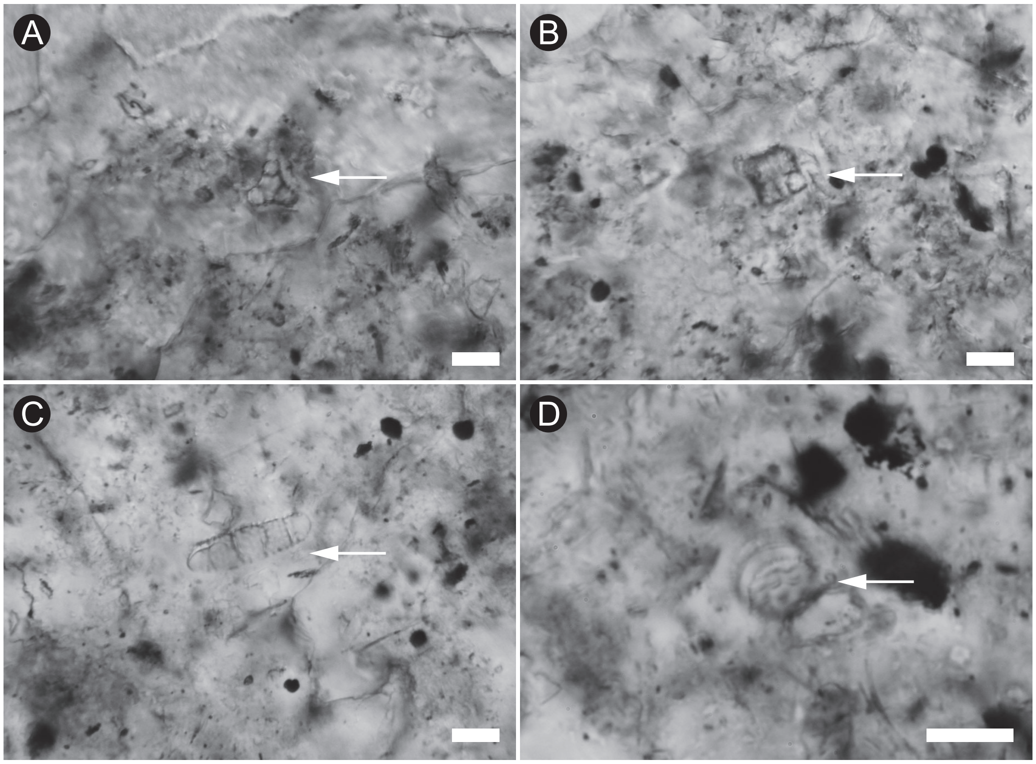

Figure 5. Photomicrographs of the Late Cretaceous diatoms in the thin section slides derived from the core (boundstone) lithofacies, the OCIS, Omagari Formation, Teshio-Nakagawa area, Hokkaido, northern Japan. **A** and **B**, *Triceratium schulzii* Jousé sensu Strelnikova (1974); **C**, *Cortinocornus rossicus* (Pantocsek) Glezer and **D**, *Anuloplicata ornata* (Grunow) Glezer. Scale bars indicate 10 μm.

fossil assemblages from other geographic provinces. Specimens of perhaps larger diatom taxa were sometimes encountered, however, not identifiable because they were fragmented into small pieces. These floral assemblages (OCIS samples and YS-CR) tend to be similar in taxonomic composition, except for YS-MR, in which the diatoms were very poorly preserved (Table 1).

Discussion

Authigenic carbonate rocks as a potential source of ancient diatoms

Carbonate concretions, which can consist of 40% to 70% carbonate, are spheres a few centimeters to several meters in diameter that occur as separate bodies within the surrounding sedimentary rock; such concretions are found in units of a variety of geologic ages. These concretions have sometimes grown around a nucleus provided by a megafossil. Some concretions are believed to form by aerobic microbial decomposition of soft tissues of dead organisms shortly after burial under anoxic conditions (Marshall and Pirrie, 2013). Spherical concretionary carbonate is suggested to have precipitated rapidly, in as surprisingly little as several weeks to a month for a concretionary body of several centimeters in diameter (Yoshida *et al.*, 2015). This process would very quickly shield and protect dead organisms from mechanical compaction or dissolution by interstitial water, thus allowing them to remain intact. Interestingly, abundant and exceptionally well-preserved siliceous microfossils sometimes occur in carbonate concretions in addition to the megafossils at the concretions' centers, despite the lack of microfossils in the surrounding host rock (for example Blome and Albert, 1985). The assemblages contained within concretions can be used for biochronology (Murakami *et al*., 2012).

Unusual chemosynthetic ecosystems composed of molluscs, arthropods, or vestimentiferan tube worms have been discovered on the modern deep-sea bottom, where hydrocarbon (= methane) outgassing occurs, in tectonically active settings such as plate margins (Majima *et al.*, 2005; Jenkins and Hikida, 2011). The primary producers in these ecosystems are complex symbiotic aerobic microbial communities. In such places precipitation of carbonate is methanogenetically and authigenically accelerated because of an increase of alkalinity. The carbonate thus produced displays extraordinarily negative δ^{13}C values (usually less than −30‰, indicating depletion of ^{13}C) as a byproduct of the metabolic processes of the microbial communities of methane-oxidizing archaea and sulfate-reducing bacteria (Jenkins and Hikida, 2011). Geologic evidence of past methane seepage activity is generally recognized by such very negative $\delta^{13}C$ ratios in carbonate rocks and associated occurrences of fossil chemosynthetic biota (Majima *et al*., 2005; Jenkins and Hikida, 2011). As for the many examples of spherical concretions, authigenic carbonate rocks induced by cold methane seepage are considered to have been formed by microbial processes. Therefore, the diatom valves documented in this study may have been well preserved as a result of rapid enclosure within the authigenic carbonate, even though their small size and delicate morphology would normally render them vulnerable to dissolution (Figure 5). As diatoms are incontestably organisms that depend on photosynthesis, not chemosynthesis, for their survival, the diatom thanatocoenoses in these authigenic carbonate rocks are allochthonous, and they would have sunk out from the surface euphotic zone and deposited in seeps on the sea bottom. The Omagari Formation, including the studied authigenic carbonate rock bodies, is inferred to have been deposited in semi-pelagic settings such as the continental margin to slope based on the bio- and lithofacies (Hikida *et al*., 2003; Takahashi *et al*., 2003; Jenkins *et al*., 2007). Our diatom flora supports these inferred sedimentological conditions in that the assemblage is dominated by probable planktonic taxa (Table 1; Witkowski *et al.*, 2011). Considering that the carbonate rock bodies at the two localities are at most several to ten meters in size (Figure 2), the diatoms from these different lithofacies (= different positions) within a single rock body should have experienced an identical sedimentation history. Therefore, dissimilarity or spatial heterogeneity of preservation in samples from different lithofacies can be explained by a preferential dissolution bias, i.e. variability in the precipitation rate of authigenic carbonate.

The Yezo Group consists of thick marine deposits of Cretaceous age, partially extending into the Paleocene, and crops out over a wide area in Hokkaido. The rocks of this group contain abundant carbonate concretions hosting well-preserved megafossils such as molluscs and vertebrates (e.g. Takahashi *et al*., 2003). In the Japanese Archipelago, authigenic carbonate rocks formed by cold methane seepage in the past have generally been discovered in Hokkaido, except for the Upper Cretaceous Nakamura Formation, Kochi area, southwestern Japan ("Sada Limestone"; Majima *et al*., 2005; Nobuhara *et al.*, 2008). As many as five localities of such authigenic carbonate rock bodies have been found exposed in Upper Cretaceous deposits in the Teshio-Nakagawa area (Kaim *et al*., 2009; Hikida, unpublished data). We concentrated on analysis of the authigenic carbonate rocks in Hokkaido, particularly as a potential source of ancient diatoms, records of which are sparse from the northwestern Pacific Ocean. So far, we have successfully found ancient (pre-Neogene) diatoms from the following seep carbonate localities: Teshio-Nakagawa (Figure 1; Omagari Formation, uppermost Santonian to Campanian, Upper Cretaceous; Shimada *et al*., 2013 and this study); Urahoro (Nuibestu Formation, Oligocene, Paleogene, eastern Hokkaido; Shimada *et al*., 2016), and Mikasa (Hikagenosawa Formation, Albian, Lower Cretaceous, central Hokkaido; Shimada *et al*., 2018).

The Cretaceous diatom flora of the northwestern Pacific Ocean

Our knowledge of ancient diatoms is extremely limited because these organisms were generally more fragile than shelled zooplankton such as radiolarians to survive diagenesis: processes of dissolution, alteration of biogenic opal-A to pyrite, and recrystallization to opal-CT and quartz have destroyed siliceous remains (for example Barron and Baldauf, 1995). Dating of such floras is also less reliable than that of assemblages from younger sedimentary rocks. Despite the scarcity of Cretaceous fossil records, several Campanian to Maastrichtian diatom fossil assemblages, which are markedly diverse and sometimes excellently preserved, have been reported from widely dispersed localities, particularly in mid- to high-latitude regions: the Ural Mountains, Russia (Jousé, 1949, 1951; Strelnikova, 1974; Oreshkina *et al*., 2013); the Arctic Ocean and adjacent regions (Barron, 1985; Tapia and Harwood, 2002; Davies *et al*., 2009; Witkowski *et al*., 2011); and the Southern Ocean (Hajós and Stradner, 1975; Harwood, 1988; Fourtanier, 1991). The common occurrence of several genera in distant geographic regions suggests that these diatoms were planktonic (Witkowski *et al.*, 2011), and that certain surface oceanographic processes may have been established by the Late Cretaceous that accelerated the diatom evolutionary radiation and subsequent colonization by seed populations.

A prototypic diatom zonation scheme was proposed for Campanian to Maastrichtian strata in Arctic Canada (Tapia and Harwood, 2002). On the basis of the occurrence of *Gladius antiquus* in both of our study localities in the Teshio-Nakagawa area (Table 1), the diatom assemblages are unequivocally assigned to the *G. antiquus* Concurrent Range Zone (Tapia and Harwood, 2002), defined as the interval between the first occurrence of *Basilicostephanus* sp. 1 (*sensu* Tapia and Harwood, 2002) and the last occurrence of *G. antiquus*. *Costopyxis schulzii*, *Hemiaulus polymorphus* v. *frigida*, and *Paralia ornata* (= *Anuloplicata ornata*), documented as associated representatives of this zone (Tapia and Harwood, 2002), were coincidentally encountered; however, *Basilicostephanus* specimens were absent in our material (Table 1). Commenting that a "post Albian? to pre Campanian (late Cenomanian to late Santonian) age" is inferred for the *G. antiquus* Zone on the basis of molluscan and foraminiferal biostratigraphy, Tapia and Harwood (2002) interpreted the diatom assemblages of this zone to be transitional, including the probable last survivors from the Early Cretaceous (*Basilicostephanus*, *Bilingua* and *Gladius*; Gersonde and Harwood, 1990) and taxa typical of Late Cretaceous time. The overlying *Costopyxis antiqua* Partial Range Zone is defined as the interval from the last occurrence of *G. antiquus* to the last occurrence of *C. antiqua* (Tapia and Harwood, 2002). On the basis of correlation to another flora from an adjacent part of the Ural Mountains, Russia (the First Diatom Complex; Strelnikova, 1974), Tapia and Harwood (2002) determined the geologic age of the zone as latest Santonian at the least. However, as the geologic age range of our materials is suggested to be approximately latest Santonian into earliest Campanian (*ca.* 83.6 Ma; ICS, 2020), there is a slight disagreement in the floral correlation between these two different provinces (the northwestern Pacific Ocean and Arctic Canada). Accordingly, further research to clarify whether the disagreement may have arisen from sensitivity of the different chronological proxies, and whether the age-diagnostic species have different stratigraphic ranges in each geographic province, will be required to develop a Cretaceous diatom biozonation.

Conclusions

In this study, we demonstrate for the first time that cold methane-seepage-induced authigenic carbonate rocks can contain ancient diatoms, as do spherical carbonate concretions. The specimens from authigenic carbonate rocks are better preserved and taxonomically diverse compared to the Cretaceous floras previously reported from the northwestern Pacific Ocean (Iwata *et al*., 1998; Takahashi *et al*., 1999; Harwood *et al*., 2004; Ando *et al*., 2014). Although these authigenic carbonate rocks formed by biogeochemical processes are less abundant both temporally and spatially than drill cores of thick biosiliceous deposits formed under more stable and highly productive marine conditions, such carbonates may offer the possibility of obtaining diatoms from horizons in which fossil occurrences have been previously thought to be poor or absent. Similarly to floras reported from other oceans at higher latitudes, the diatom assemblages in this study are composed of planktonic genera, plus a few unknown (and thus new) associated species that were probably endemic to the region. This evidence suggests that adaptive migration and colonization of the diatom community had been established as a result of globally driven surface oceanographic processes during Late Cretaceous time. However, further research is needed to improve the biozonation of Late Cretaceous diatoms to yield greater spatial coverage with higher temporal resolution.

Acknowledgements

Fumio Akiba (Diatom Mini Lab Ltd.), an anonymous reviewer and Akihisa Kitamura (Shizuoka University), Journal Editor, gave fruitful comments to improve the manuscript. Kazuaki Nagaya and Chiaki Hayasaka helped us with curation at the Nakagawa Museum of Natural History. This research was funded by Grants-in-Aid (KAKENHI) to CS from the Japan Society for the Promotion of Science (nos. 26400497 and 18K03818).

We are grateful to the late Akinori Takahashi for his dedication to paleontological investigation and for his suggestion that we investigate the Cretaceous molluscan bio- and lithostratigraphy of Hokkaido.

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Anaulus sibericus Strelnikova [Figure 4-H]. Barron (1985), pl. 10.2, fig. 10; Harwood (1988), fig. 9.14; Dell'Agnese and Clark (1994), fig. 3.3.

Anuloplicata ornata (Grunow) Glezer [Figure 4-J and 4-K]. Fenner (1994), pl. 1, fig. 5; Oreshkina *et al*. (2013), pl. II, fig. (6). *Melosira ornata* Grunow. Strelnikova (1974), pl. I, figs. 5 and 6. *Paralia ornata* (Grunow) Grunow ex van Heurck, Tapia and Harwood (2002), pl. 1, figs. 3 and 4; Witkowski *et al*. (2011), figs. 6.E and 11.D. *Pseudopodosira bella* Posnova and Glezer, Fenner (1978), pl. 10, figs. 6 to 8 (figs. 2 and 3 are excluded)? Genus and species unknown, Shimada *et al.* (2013), figs. 3.6 to 3.8.

Benetorus sp.

Biddulphia sparsepunctata Hajós and Stradner [Figure 4-E and 4-F].

Cortinocornus rossicus (Pantocsek) Glezer [Figure 4-L to 4-P]. Witkowski *et al*. (2011), figs. 8.A to 8.E, 8.H, 8.R, 8.S and 12.A to 12.D; Oreshkina *et al*. (2013), pl. IV, figs. (10) to (12). *Hemiaulus polycystinorum* Ehrenberg. Jousé (1949), pl. I, fig. 10; Hajós and Stradner (1975), pl. 6, figs. 4 to 7. *Hemiaulus rossicus* Pantocsek. Glezer *et al*. (1974), pl. XI, fig. 6 and pl. XV, fig. 10; Strelnikova (1974), pl. XLIII, figs. 1 to 18; Ross *et al*. (1977), pl. 8, figs. 48 to 52; Harwood (1988), figs. 14.18 to 14.21, figs. 15.4 and 15.5; Tapia and Harwood (2002), pl. 3, figs. 5 to 8; Shimada *et al*. (2013), figs. 3-9 to 3-12.

Costopyxis schulzii (Steineck and Schulz) Glezer [Figure 4-G]. Glezer *et al*. (1988), pl. 31, figs. 15 to 19; Witkowski *et al.* (2011), figs. 7.E, 7.F, 7.M, 7.N and 11.K; Oreshkina *et al*. (2013), pl. I, fig, (2). *Stephanopyxis schulzii* Steineck, Strelnikova (1974), pl. IV, figs. 1 to 3 and 8; Fenner (1985), fig. 14.6 (referring to Strelnikova, 1974); Fourtanier (1991), pl. 5, fig. 8. *Stephanopyxis schulzii* v. *cretacea*, Jousé (1949), pl. I, figs. 1 and 2.

Euodiella sp.

Gladius antiquus Forti and Schulz. Gersonde and Harwood (1990), pl. 7, figs. 1 and 2, pl. 8, figs. 1, 2, 5, and 6; Tapia and Harwood (2002), pl. 1, fig. 2; Witkowski *et al.* (2011), figs. 7.Q and 7.R.

G. antiquus v. *tenuis* Gersonde and Harwood [Figure 4-D]. Tapia and Harwood (2002), pl. 1, fig. 1; Shimada *et al*. (2013), fig. 3.19.

Hemiaulus altus Hajós and Stradner. Fenner (1978), pl. 24, fig. 15.

H. elegans (Heiberg) Grunow. Glezer *et al.* (1974), pl. 11, fig. 7 and pl. 19, fig. 6; Strelnikova (1974), pl. XLIV, figs. 1 to 15; Hajós and Stradner (1975), pl. 5, figs. 17 to 19; Barron (1985), pl. 10.2, fig. 3; Harwood (1988), fig. 13.18; Fenner (1994), pl. 5, figs. 1 to 3; Oreshkina *et al*. (2013), pl. IV, fig. (7). *Cortina elegans* Heiberg, Schmidt (1874–1959), pl. 76, figs. 54 and 55.

H. polymorphus v. *frigida* Grunow. Schmidt (1874–1959), pl. 143, figs. 35 and 36; Glezer *et al.* (1974), pl. XV, fig. 9 and pl. XIX, fig. 1; Strelnikova (1974), pl. XLV, figs. 1 to 19; Harwood (1988), figs. 13.29 and 14.7; Dell'Agnese and Clark (1994), fig. 4.2; Tapia and Harwood (2002), pl. 5, fig. 8.

Hemiaulus sp. A cf. *H. antiquus* Jousé [Figure 3-N to 3-Q].

Hemiaulus sp. B cf. *H. kittonii* Grunow.

Hemiaulus sp. C. cf. *H. polymorphus* Grunow.

Hemiaulus sp. D [Figure 3-R to 3-T].

Hemiaulus ? sp. E [Figure 3-U to 3-W].

Hemiaulus ? sp. F.

Medlinia sp.

Paralia crenulata (Grunow) Glezer [Figure 4-I]. Nikolaev *et al*. (2001), pl. 8, figs. 1 to 8; Sims and Crawford (2002), figs. 1 to 17; Tapia and Harwood (2002), pl. 4, figs. 14 and 15; Witkowski *et al*. (2011), figs. 7.H, 7.I and 11.E; Oreshkina *et al.* (2013), pl. II, fig. (2); Shimada *et al*. (2013), fig. 3.4. *Melosira fausta* Schmidt, Hanna (1927), pl. 3, figs. 12 to 14. *Melosira sulcata* (Ehrenberg) Kützing, Fenner (1978), pl. 13, fig. 6; Moshkovitz *et al.* (1983), figs. 4.(e) and 4.(f). *Melosira sulcata* (Ehrenberg) Kützing v. *crenulata* Grunow, Jousé (1955), pl. I, fig. 6; Glezer *et al*. (1974), pl. XVI, fig. 3; Strelnikova (1974), pl. I, figs. 7 and 8.

Pterotheca evermanni Hanna. Harwood (1988), figs. 18.13 and 18.14; Fenner (1994), pl. 4, fig. 9; Nikolaev *et al*. (2001), pl. 39, figs. 6 and 7; Oreshkina *et al*. (2013), pl. IV, fig. (6).

Pyxidicula weyprechtii Grunow. Oreshkina *et al.* (2013), pl. II, fig. 4.

Sceptroneis grunowii Anissimowa. Jousé (1951), pl. IV, fig. 8, Glezer *et al*. (1974), pl. XVII, fig. 12; Hajós and Stradner (1975), pl. 11, figs. 14 to 16; Dzinoridze *et al.* (1978), pl. 10, fig. 8; Harwood (1988), figs. 17.9 and 17.10.

Sceptroneis sp. A [Figure 4-A and 4-B].

Sceptroneis sp. B.

Sceptroneis sp. C [Figure 4-C].

Sheshukovia sp.

Stellarima steinyi (Hanna) Hasle and Sims [Figure 4-Q]. Sims and Hasle (1987), figs. 1 to 18; Harwood (1988), figs. 20.3 and 20.4; Dell'Agnese and Clark (1994), fig. 4.8; Nikolaev *et al*. (2001), pl. 20, figs. 1 to 6; Tapia and Harwood (2002), pl. 7, fig. 2.

Stephanopyxis turris (Greville and Amott) Ralfs. Strelnikova (1974), pl. VIII, figs. 1 to 13; Fenner (1978), pl. 12, fig. 7; Moshkovitz *et al*. (1983), fig. 7.(l); Barron (1985), pl. 10.3, fig. 5; Harwood (1988), figs. 19.26 and 19.27; Dell'Agnese and Clark (1994), fig. 5.1; Fenner (1994), pl. 2, fig. 12; Nikolaev *et al*. (2001), pl. 7, figs. 5 and 6; Tapia and Harwood (2002), pl. 9, figs. 9 and 10; Shimada *et al*. (2013), fig. 3.18.

Thalassiosiropsis sp. cf. *T. wittiana* (Pantocsek) Hasle.

Triceratium indefinitum (Jousé) Strelnikova. Glezer *et al*. (1974), pl. XI, fig. 5; Strelnikova (1974), pl. XXX, figs. 1 to 29, pl. XXXI, figs. 1 to 6; Barron (1985), pl. 10.3, figs. 11 and 12; Harwood (1988), fig. 20.11 and figs. 21.4 to 21.7; Tapia and Harwood (2002), pl. 4, figs. 1, 2, 5 and 6. *Trinacria indefinita* Jousé, Dell'Agnese and Clark (1994), fig. 5.3; Witkowski *et al*. (2011), figs. 8.I and 8.J.

T. planum Strelnikova ? [Figure 3-L and 3-M]. Barron (1985), pl. 10.3, fig.7; Fenner (1985), figs. 13.8 and 13.9; Dell'Agnese and Clark (1994), fig. 4.10; Oreshkina *et al*. (2013), pl. III, figs. (1) and (2).

T. schulzii Jousé [Figure 3-A to 3-G]. Strelnikova (1974), pl. XXIX, figs. 1 to 7; Fenner (1978), pl. 30, figs. 1 to 11; Fourtanier (1991), pl. 5, fig. 6; Tapia and Harwood (2002), pl. 2, figs. 5, 12 and 13; Shimada *et al*. (2013), fig. 3.1 to 3.3. *Triceratium kanayae* Fenner. Fenner (1985), fig. 8.24 to 8.27. *Triceratium* sp. a, Kanaya (1957), pl. VII, figs. 5 to 7. *Triceratium* sp. 1, Fourtanier (1991), pl. 5, figs. 4 and 5.

Triceratium sp. A. [Figure 3-H to 3-K]. This species resembles *T. schulzii* Jousé sensu Strelnikova (1974) or *Triceratium kanayae* Fenner described by Fenner (1984), but is differentiated by the presence of a circular elevation on the valve face. In addition, the projecting corners of the valve are more acutely constricted and the septum under the corners is narrower than in *T. schulzii*/*T. kanayae.*

Genus and species indet. A. Connecting structure of *Gladius antarcticus* f. *alta* Harwood ?

Genus and species indet. B.

Genus and species indet. C.

Genus and species indet. D.

Author contributions

CS and MSK performed the diatom analysis. RGJ and YH contributed to the local geological survey and faunal analysis. All authors participated in integrated discussion of the Cretaceous paleoecology and paleoenvironment. CS was totally responsible for designing the research and wrote the manuscript.