Conditions of Diatomites Formation in the Primorye (South of the Russian Far East)

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ABSTRACT: The analysis of diatomaceous deposits in the South of the Russian Far East allowed us to determine the taxonomic composition of the diatom flora and assess its qualitative (ecological) and quantitative traits. The obtained data enabled us to identify biofacies associated with the conditions of diatomite formation and reveal the reasons behind it. Possible reasons of high diatom productivity in the Neogene could be: development of a dense system of rivers and lakes; a long growing season under the conditions of a tropical monsoon climate with mild winters; active volcanic activity, the eruption products of which were the source of materials needed for the formation of valves and supporting the life activity of diatoms.

KEY WORDS: diatomite, Neogene, sediment genesis, South of the Russian Far East

INTRODUCTION

Miocene and Pliocene deposits of Primorye contain a rather large amount of tuffaceous diatomites and diatomites, which are a valuable biogenic source of mineral raw materials with widespread use in industrial and economic activities. All known deposits are almost totally depleted, while the search for new deposits requires a scientifically justified and focused approach. To date, the conditions, under which diatomite deposits formed, remain mostly unclear (Evzerov, 2011). Reconstruction of these conditions, elucidation of the factors that provide diatom biogenic sedimentogenesis will allow establishing cause-and-effect relationships in the "sedimentogenesis-diatomite" system, and determining criteria for diatomite deposit prospecting in the future. The diatomite formation period is characterized by the paleoclimate changes related to several geological events including tectonic activation and uplift of the Tibetan Plateau.

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The climate change accounted for a sharp difference in heating temperature of the plateau and surface waters of southern marginal seas (Royden et al., 2008). These events caused the formation of the East Asian monsoon and increase in aridity in Central Asia (Jiang, Ding, 2008; Likhacheva et al., 2009; Pushkar et al., 2019). Another global event of the time was a long-lasting positive carbon isotope excursion known as the “Monterey Excursion”, which coincided with the early to Middle Miocene warming after 16.9 Ma (Flower, Kennett, 1993; Wang et al., 2003; Holbourn et al., 2007). The latter might have also influenced the thermal maximum during this period (Miocene Climatic Optimum) and approximately at 13.5 Ma was replaced by a cooling trend, which continued in the Pliocene, temporarily ceasing only in the earliest (5.2–5.0 Ma) and middle (3.3–3.0 Ma) Pliocene (Dowsett et al., 2005). Similar climate changes in Primorye during the Miocene-Pliocene were also established (Korotky et al., 1996; Pavlyutkin, Petrenko, 2010; Pushkar et al., 2019).

This research is aimed at analyzing the spatio-temporal distribution of diatoms, their taxonomic composition, and high concentrations of valves in Miocene–Pliocene sediments of South Primorye lakes, and at establishing the reasons behind these processes.

MATERIALS AND METHODS

This paper presents analysis results of diatom samples collected from three diatomites located in different areas of Primorye (Figure): 1) western shore of the Khanka Lake, between settlements of Tury Rog and Novokachalinsk (Khanka District); 2) upper reaches of the Sergeevka River (Partizansk District); 3) near Terekhovka village (Nadezhdinsky District).

1. On the western shore of the Khanka Lake we collected forty samples from interbeds of brown, much brighter when dried, laminated tuffaceous diatomites of the Novo-kachalinsk formation, in the sections Nos 9149, L-03/2 (45°09'14,3"N, 132°00'23,3"E), L-04/1 (45°09'11,8"N, 132°00'126,00"E), L-02 (45°01'00,0"B, 132°00'25,1"E). Diatomite bed thickness is 4 to 11 meters. The Novokachalinsk formation age has been determined to be the beginning of the Middle Miocene or, possibly, late Early Miocene (Pavlyutkin et al., 2004). The studied sediments are hereinafter referred to as the Novokachalinsk diatomite.

2. Five samples were collected from the tuffaceous diatomite layer in a quarry near the Terekhovka village, site 4131 (43º20'N, 131º52'E), where the Shufan horizon sediments are exposed. The diatomite thickness is 4 m. It is white with a yellowish shade and is weakly cemented. The diatomite is of Pliocene age (Pavlyutkin, Petrenko, 2010). These sediments are hereinafter referred to as the Terekhovka diatomite.

3. Three samples were collected at a depth of 89.5–82.5 m from opoka-like clay

interbeds exposed by the boreholes 27 and 28 made by OOO "Primorgeologiya" in 1988 (Kovalenko, 1989). Total diatomite thickness and length were not determined. The diatomite is light, weakly cemented, and its color varies from white to yellow. According to the diatom analysis data, the diatomite is of the late Pliocene age (Avramenko et al., 2015). The described sediments are hereinafter referred to as the Sergeevka diatomite.

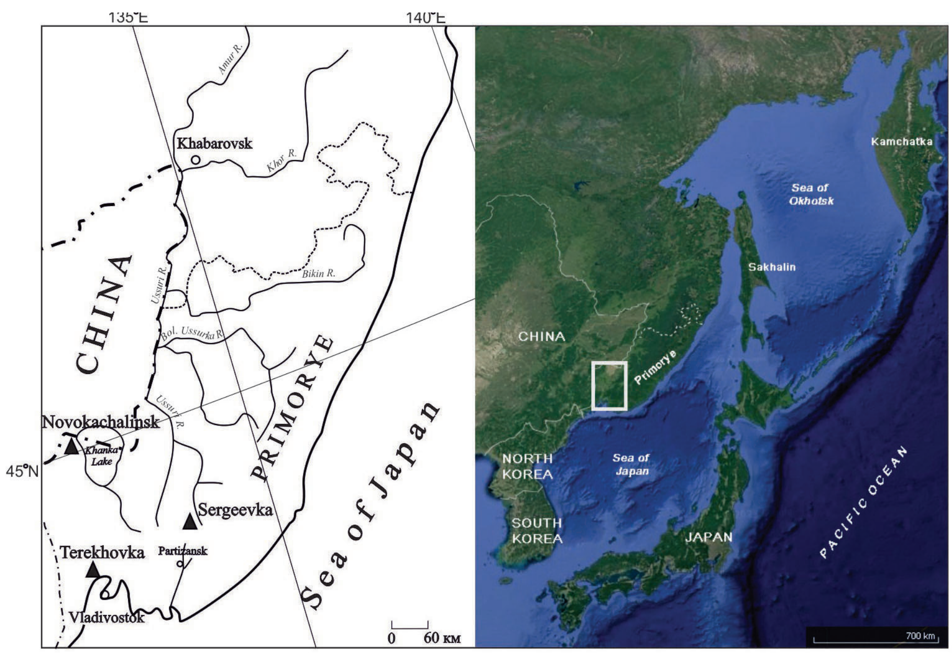

FIGURE: Map of the study area. ▲ – Locations of the diatomites

The sample processing was performed in accordance with the accepted standards (Diatoms…, 1974). The taxonomic analysis, calculation of diatom valves, and measurement of their parameters: diameter, length, width were conducted using the Carl Zeiss Amplival and Carl Zeiss Axioskop 40 light microscopes (LM), and Carl Zeiss EVO 40 (magnification of up to ×15000) and Merlin (magnification of up to ×50000) scanning electron microscope (SEM) at the Instrumental Centre of Biotechnology and Gene Engineering of the Federal Scientific Center of the East Asia Terrestrial Biodiversity FEB RAS. The diatom valve content in 1 g of sediment (N) has been determined via a method proposed by Avramenko and coauthors (2015).

RESULTS

The diatom analysis of the samples collected from three locations of siliceous organogenic sediments allowed us to describe the characteristics of Miocene–Pliocene diatom floras.

The diatom flora of the ***Novokachalinsk diatomite*** comprises 35 species and infraspecific taxa. Planktonic centric diatoms are the dominant species, the prevalence of *Aulacoseira praegranulata* var. *praeislandica* f. *praeislandica* (Jousé) Moiseeva, inhabiting freshwater lakes, reaches 84.7% (Plate I, *A*; Plate II, *A, D*, *G*). Other widespread species include the planktonic *A. praegranulata* var. *praeislandica* f. *curvata* (Jousé) Moiseeva constituting up to 7.27% (Plate II, *C*) and *A. praegranulata* var. *praegranulata* f. *praegranulata* (Jousé) Simonsen constituting up to 2% (Plate II, *B*). The presence of other taxa is very small (less than 1%). Among them: *A. praegranulata* var. *praegranulata* f. *curvata* (Jousé) Simonsen (Plate II, *F*), *A. praegranulata* var. *praeangustissima* f. *praeangustissima* (Jousé) Moiseeva*, A. praegranulata* var. *praeangustissima* f. *curvata* (Jousé) Moiseeva*, Alveolophora tscheremissinovae* Khursevich (Plate II, *J*), and *A. khursevichiae* Usoltseva, Pushkar et Likhacheva. The involvement of benthic taxa is minimal. This group is represented by centric diatoms *Melosira undulata* (Ehrenberg) Kützing (Plate II, *H*), *Ellerbeckia kochii* (Pantocsek) Lupikina (Plate II, *I*) and pennate diatoms of the *Achnanthes* Bory, *Eunotia* Ehrenberg, *Navicula* Bory, *Tetracyclus* Ralfs genera. The diatom valves are well-preserved. The valve content in 1 g of sediment is $14.04 \cdot 10^8$.

The study showed a wide variability in diameter and mantle height of the dominant taxon valve structure. The valve diameter of the prevailing *A. praegranulata* var. *praeislandica* f. *praeislandica* varies from 4.84 to 27 μm (coefficient of variation (CV) was 26.8), mantle height – 2.0–17.95 μm (CV = 28.3), the valve diameter of the coexisting *A. praegranulata* var. *praegranulata* f. *praegranulata* is 5.73–10 μm (CV = 14.1), its mantle height is 5.97–21.88 μm (CV = 21.3), and the valve diameter of *A. praegranulata* var. *praeangustissima* f. *praeangustissima*, the involvement of which was very small, ranges from 2.92 to 7.36 μm (CV = 18.0), its mantle height is 14.04–23.5 (CV = 14.9).

The diatom flora of the ***Terekhovka diatomite*** comprises 36 species and infraspecific taxa. The dominant species are *Aulacoseira* Thwaites, planktonic centric diatoms (Plate II, *B*; Plate III). *Aulacoseira praegranulata* var. *praeislandica* f. *praeislandica* are dominant (up to 93.2%) (Plate III, *A–H, I*). Other species *Melosira undulata* (Plate III, *G*, *J*, *K*)-has a relatively high estimate of abundance (2.7%). Other taxa constitute less than 1%. These taxa represent genera of *Actinocyclus* Ehrenberg (Plate III, *L–N*)*, Tetracyclus, Ellerbeckia* Crawford (Plate III, *O*), *Eunotia.* The diatom valves are well-preserved in rocks. The valve content in 1 g of sediment is $1.5 \cdot 10^9$.

We have established a wide variability in diameter (CV = 15.83) and mantle height (CV = 10.84) of the dominant *A. praegranulata* var. *praeislandica* f. *praeislandica* taxon valves. The valve diameter varies from 4.45 to 22.8 μm, and the mantle height ranges from 3.25 to 12.34 μm.

The flora of the ***Sergeevka diatomite*** was formed by 25 species and intraspecies. The dominant are benthic pennate small-valved forms, less frequently planktonic diatoms (Plate I, *C*).

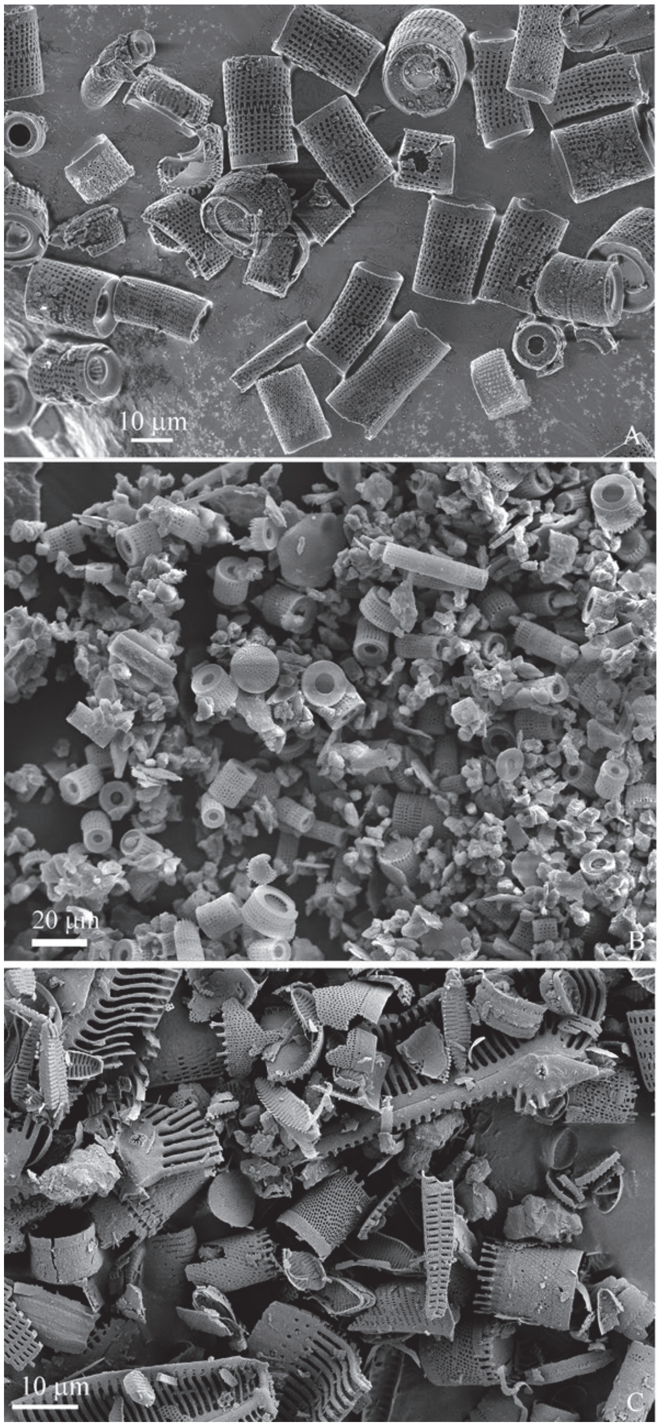

PLATE I: General view of the diatomites (SEM):
A – Novokachalinsk; *B* – Terekhovka; *C* – Sergeevka

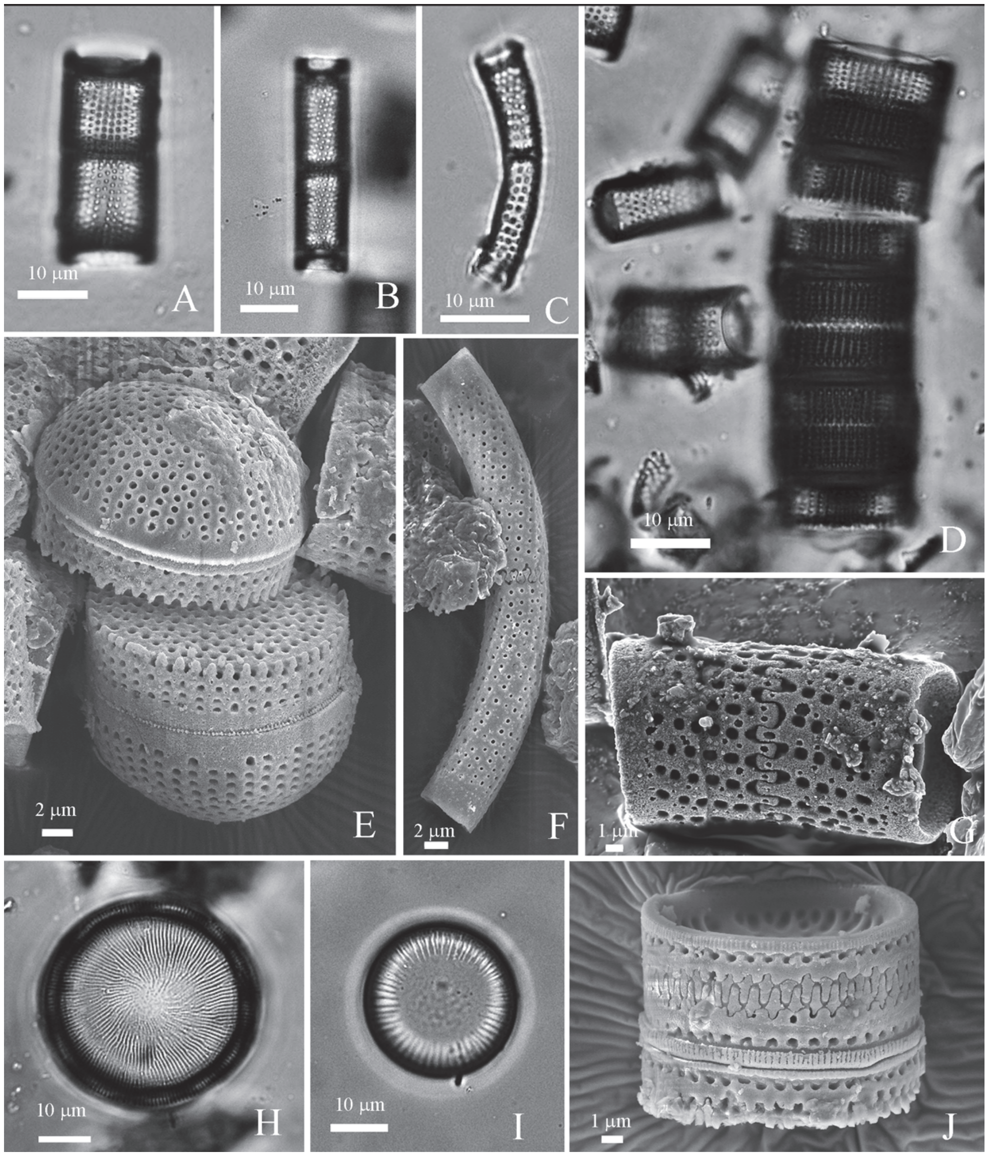

PLATE II: Rock-forming taxa of the Novokachalinsk diatomite: *A*, *D*, *G* – *Aulacoseira praegranulata* var. *praeislandica* f. *praeislandica* (Jousé) Moiseeva; *B* – *A. praegranulata* var. *praegranulata* f. *praegranulata* (Jousé) Simonsen; *C* – *A. praegranulata* var. *praegranulata* f. *curvata* (Jousé) Moiseeva; *E* – resting spores of *Aulacoseira*; *F* – *A. praegranulata* var. *praegranulata* f. *curvata* (Jousé) Moiseeva; *H* – *Melosira undulata* (Ehrenberg) Kützing; *I* – *Ellerbeckia kochii* (Pantocsek) Lupikina; *J* – *Alveolophora tscheremissinovae* Khursevich. *A–D*, *H*, *I* – LM; *E–G*, *J* – SEM

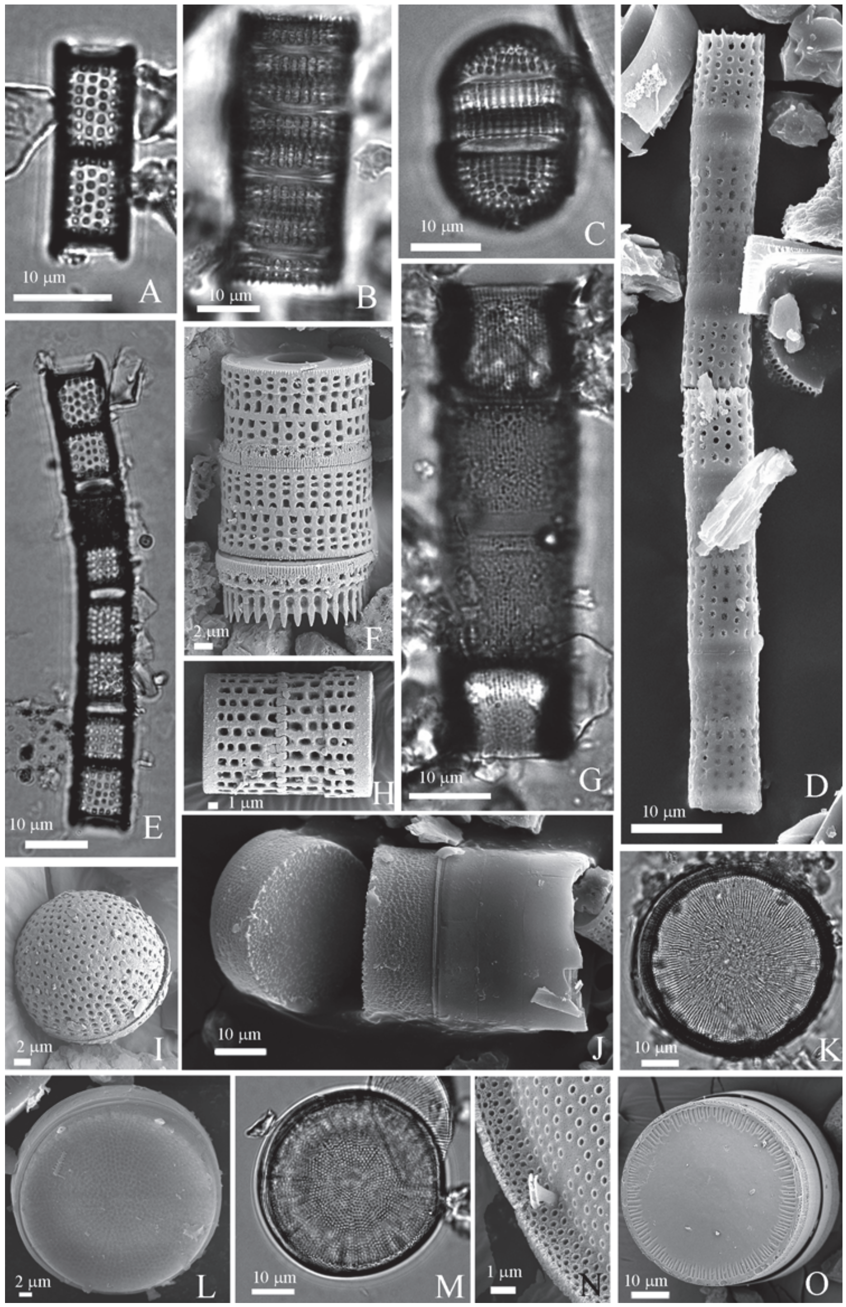

PLATE III: Rock-forming taxa of the Terekhovka diatomite: *A*, *B*, *D–H* – *Aulacoseira praegranulata* var. *praeislandica* f. *praeislandica* (Jousé) Moiseeva; *C*, *I* – resting spores of *Aulacoseira*; *G*, *J*, *K* – *Melosira undulata* (Ehrenberg) Kützing; *L–N* – *Actinocyclus gorbunovii* (Sheshukova-Poretskaya) Moiseeva & Sheshukova-Poretskaya; *O* – *Ellerbeckia kochii* (Pantocsek) Lupikina. *A–C*, *E*, *G*, *K*, *M* – LM; *D*, *F*, *H*, *I*, *J*, *L*, *N*, *O* – SEM

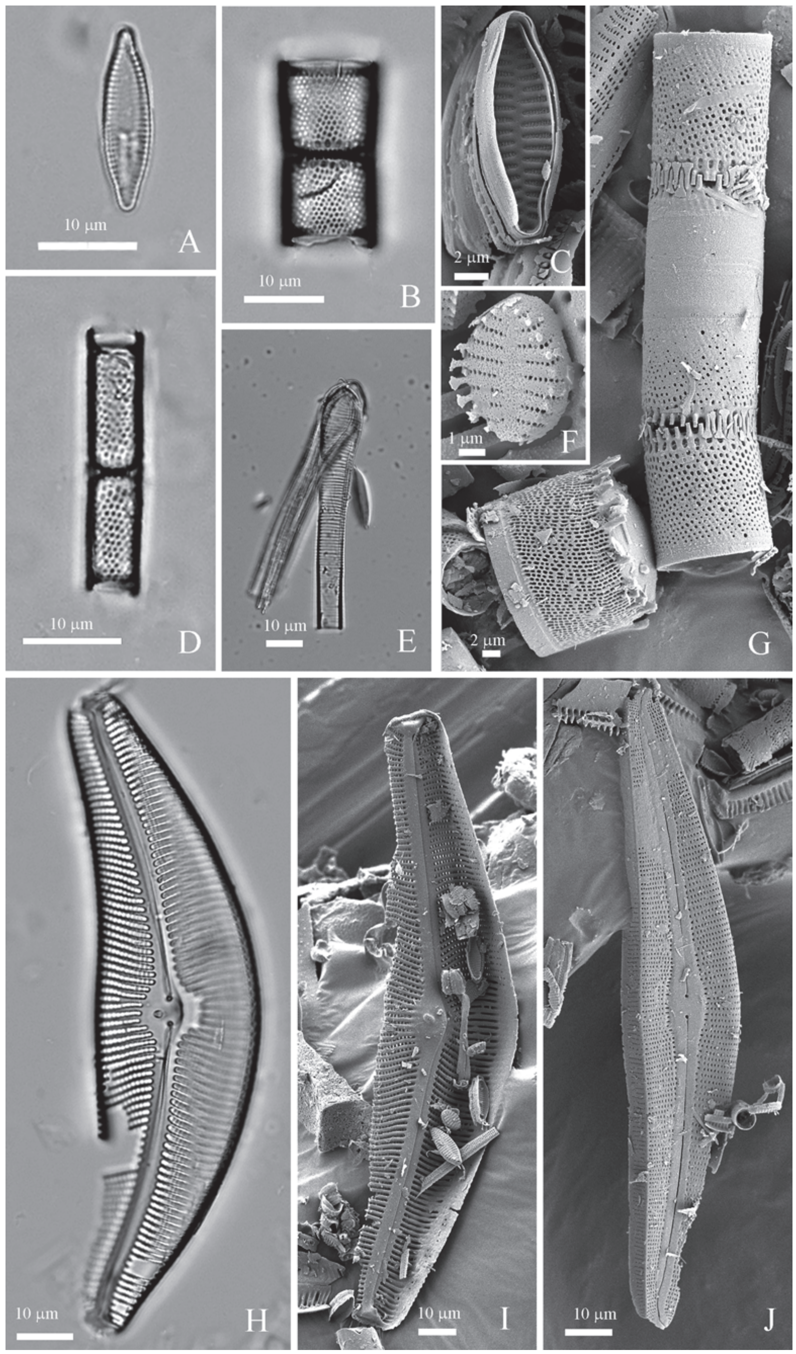

PLATE IV: Rock-forming taxa of the Sergeevka diatomite: *A*, *C*, *F* – *Staurosira venter* (Ehrenberg) Cleve et Möller; *B*, *D*, *G* – *Aulacoseira italica* (Ehrenberg) Simonsen; *E* – *Actinella brasiliensis* Grunow; *H*, *I* – *Cymbella* aff. *australica* (Schmidt) Cleve; *J* – *C. aspera* (Ehrenberg) Cleve. *A*, *B*, *D*, *E*, *H* – LM; *C*, *F*, *G*, *I*, *J* – SEM

Staurosira venter (Ehrenberg) Cleve et Möller is the rock-forming taxon (up to 84.8%) (Plate IV, *A*, *C*, *F*). *Aulacoseira italica* (Ehrenberg) Simonsen also has a high estimate of abundance, up to 15.6% to be precise (Plate IV, *B*, *D*, *G*). The presence of *Cymbella* Agardh species with a large valve size: *C. aspera* (Ehrenberg) Cleve (Plate IV, *J*) and *C.* aff. *australica* (Schmidt) Cleve (Plate IV, *H*, *I*) should be noted. Despite the latter not being abundant (around 2%), the large size of the valves of these taxa (up to 175 µm long) allows considering them, for our purpose, as rock-forming. The involvement of *Eunotia* and *Neidium* Pfitzer genera species is small. The diatom valves, found in sediments, are well-preserved. The valve content in 1 g of sediment $6.1 \cdot 10^8$.

The dominant *S. venter* species valve length ranges from 3.79 to 23.22 µm (CV = 38.2), valve width from 2.96 to 5.65 µm (CV = 10.94). The valve diameter of the coexisting *A. italica* species varies from 3.2 to 19.14 µm (CV = 33.27), its height from 6.53 to 20.47 µm (CV = 17.49). The valve length of the *C.* aff. *australica* ranges from 94.94 to 175.04 µm, and its width from 25.50 to 33.43 µm.

DISCUSSION

Productivity of diatoms from the studied deposits

The valve concentration in sediments is often one of the indicators of diatom productivity in the geological past. The analysis of the valve content in the studied diatomites showed high concentrations in all sediments. The highest valve concentration is in the Terekhovka diatomite – 1.5 billion valves per 1 g of dry sediment, a lesser concentration is found in the Novokachalinsk diatomite – 1.4 billion, and the smallest concentration in the Sergeevka diatomite – 610 million.

Another specific trait of centric diatom valve division intensity, their productivity in other words, is the variability of such morphological parameters as diameter and height of the valve mantle. High variation ratios of the mentioned parameters of the dominant taxa from the studied sediments indicate an intensive vegetative reproduction of diatoms. This is also confirmed by long colonies preserved in sediments and made of 4–8 valves (Plate II, *D*; Plate III, *B*, *D*, *E*), and by frequently found initial cells (up to 0.9%) (Plate II, *E*; Plate III, *C*, *I*).

Taxonomic and ecological compositions of diatom communities as a reflection of their habitat characteristics

Taxonomic and ecological compositions of the diatom flora described above indicate that the diatoms actively developed in the Neogene lakes related spatially and genetically to the superimposed Cenozoic depressions that used to be sedimentary basins (Pavlyutkin, Petrenko, 2010). The dominance of the *Aulacoseira* genus, living in the plankton of modern lakes of different genesis, in the flora of the Novokachalinsk and Terekhovka diatomites (Trifonova, 1979), and the presence of small valve species of the *Staurosira* Ehrenberg, *Pseudostaurosira* Williams et Round, and *Staurosirella* Wiliams et Round genera in the

Sergeevka diatomite suggest that they accumulated in similar basins. Only the *Aulacoseira* flora developed in larger and deeper lakes, while the *S. venter* flora developed in a small, relatively shallow and, probably, oxbow lake. Small valve taxa of the Sergeevka diatomite are considered as the pioneering species, settling in reemerged modern oligotrophic arctic waters (Michelutti et al., 2003) and small forest lakes (Cherepanova et al., 2013). The Sergeevka diatomite accumulating in an oxbow lake of a river's valley is also evidenced by the presence of *Eunotia*, *Cymbella*, *Neidium* genera inhabiting swamps or adjacent biotopes. This lake, probably, dried out from time to time and later filled with water during an inundation or flood. The cross-section near the Terekhovka village has clay interbeds and sand inclusions, which may serve as indirect evidence of complex sedimentary conditions. It should also be noted that in the second half of the Miocene in Primorye the lake sediment genesis was replaced by an alluvial one (Pushkar et al., 2019).

Modern representatives of the *Aulacoseira* genus are often a part of the prevailing groups of planktonic algal communities in freshwater basins. This genus is considered to be cosmopolitan and can be found on all continents (Kociolek, 2018). Moreover, we have revealed seasonal fluctuations and high productivity of certain taxa. For example, *A. islandica* (Müller) Simonsen is a cold-water species, the thermal optimum of tolerance of which is +5 to +10 °C. It completely dominates plankton in spring and autumn and reaches highest productivity in the middle or end of May. The water mass heating causes the cessation of population growth (Trifonova, 1979). *Aulacoseira granulata* (Ehrenberg) Simonsen, which dominates plankton in summer and autumn at 20 °C in shallow waters (Barinova et al., 2008), is thought to be the most thermophilic species of this genus. We assume that wide taxonomic diversity of the *Aulacoseira* genus in the Novokachalinsk and Terekhovka diatomite can be indicative of sharply pronounced seasonal patterns during this time of year.

Influence of temperature on diatom productivity

In waters of temperate climate regions phytoplankton dynamics are characterized by clear seasonal trends. Diatoms in these waters often bloom in spring and late autumn, when the conditions for active algal growth, water mixing, nutrient content and light intensity are optimal (Reynolds, 2006). Relatively low temperature of water (Trifonova, 1979), in which other groups of phytoplankton do not develop so actively (Sommer et al., 1986), is considered to be one of the factors defining high productivity of diatoms in these seasons.

Korotky and coauthors (1996) used the palynological data to reconstruct the main parameters of the Neogene climate in Primorye. They took into account ecological preferences of modern taxa analogous to indicator taxa from Neogene spore and pollen spectra. The climate of the Middle Miocene, when the Novokachalinsk diatomite formed, was identified as typical subtropical with average annual temperature of +15–17 °C, in July +27–29 °C, in January +4–7 °C, and annual precipitation of 1200–1400 mm. The abundant precipitation and the forming monsoon climate accounted for the development of a dense lake system in the region, and a relatively warm winter could be the reason behind the long growing season of diatoms. An indirect evidence of diatom development in wintertime at relatively high winter temperatures is that diatoms demonstrate a relatively high growth rate and are usually adapted to low light intensity in waters with high concentrations of nutrients in comparison to other phytoplankton taxa (Reynolds, 2006).

From the Late Miocene and during the Pliocene, the climate in South Primorye became colder: the temperature in January was -2–3 °C, in July +22 °C, and annual precipitation was over 1200 mm (Korotky et al., 1996). These conditions defined the active development of diatoms in Pliocene waters, which led to the formation of the Terekhovka and Sergeevka diatomites. The presence of warm-water *Actinella brasiliensis* Grunow in the sediments near the Sergeevka village serves as indirect evidence.

Volcanic activity and its impact on diatom productivity

One more condition needed for increased diatom productivity is a sufficient amount of dissolved silica and nutrients – phosphates and nitrates, which are used in the formation of valves and are required for activity of these algae. Volcanic ash was the main source of silica and nutrient transport into the lakes of Primorye during the Miocene and Pliocene. This is evidenced by thick tuffaceous diatomite deposits of the Novokachalinsk formation and basaltic flows of the Shufan horizon (Pavlyutkin, Petrenko, 2010). Hard siliceous valves of almost all studied diatoms prove the high (or even excessive) concentration of silica in the water. In addition, the volcanic ash can inhibit the dissolution of biogenic (opal) silica and support the formation of diatom deposits (Riedel, 1959).

Pyroclastic material covered a large territory and was accumulated in lakes not only during eruptions, but also afterwards from the surface of drainage basins of these waters (Kovalenko, 1989). Small volcanic particles provided nutrients needed for the formation of diatom valves more actively than, for example, basaltic interbeds that cover the bottom of the sedimentary basin, in which the Sergeevka diatomite formed.

Influence of carbon concentration in the atmosphere on the diatom reproduction rate

Diatoms are photosynthetic organisms, thus, CO_2 content in the Earth's atmosphere is undoubtedly of great importance for their development. They are considered to be a biogeochemically significant group of phytoplankton, contributing largely to natural carbon fixation (Armbrust, 2009), and some researchers suggest cultivating them to prevent carbon dioxide concentration increase in the atmosphere (Sethi et al., 2020).

The formation of the Novokachalinsk diatomite was related to the Monterey carbon excursion in the Middle Miocene, when the CO_2 concentration in the atmosphere reached 470–630 ppm (Sosdian et al., 2018; Babbila, Foster, 2021), while its modern concentration is 300–450 ppm. This diatomite is marked by the maximum concentration of diatoms in sediments. It is possible that the high diatom productivity in this period was caused, among other factors, by the high content of this greenhouse gas in the atmosphere (Pushkar, 2020).

CONCLUSIONS

Diatoms develop actively under the influence of many factors, among which the most significant are: favorable paleogeographic setting with a specific climate, in which water basin with a diverse and abundant diatom flora form; and an environment with a sufficient amount of dissolved silica and other nutrients required for the formation of diatom valves.

The conducted diatom analysis showed that all studied diatomites have high valve concentrations and a wide morphological variability of the dominant taxa (the Novokachalinsk and Terekhovka diatomites) indicating high diatom productivity during the diatomite formation.

The Novokachalinsk and Terekhovka diatomites are clearly dominated by species of the *Aulacoseira* genus usually inhabiting the plankton in freshwater, and the Sergeevka diatomite is dominated by small valve species of the *Staurosira*, *Pseudostaurosira* and *Staurosirella* genera living in small oxbow lakes in river valleys.

A rather important factor effecting/causing the high diatom productivity is the relatively low water temperature. According to reconstructions, during the Neogene Primorye had a monsoon climate, which accounted for the development of a dense lake system in the region, and relatively warm winter could be the cause of a long diatom vegetation season, which provided the possibility for diatoms to develop actively in spring-autumn-winter period.

Intensive volcanic activity in South Primorye during the Miocene and Pliocene accounted for the high diatom productivity, as the waters were filled with a sufficient amount of silica, phosphates and nitrates that play a significant role in the development of diatoms and in formation of their valves. In the Middle Miocene volcanic ash served as a source of silica and other materials enhancing the development of diatoms of the Novokachalinsk diatomite, while basaltic lavas surrounding the Terekhovka diatomite supplied it with all necessary materials used by diatoms to form their valves. Hard siliceous diatom valves of these two diatomites indicate that a large amount of silica was transported into these basins. The dissolved silica of basaltic interbeds, composing the sedimentary basin bottom and destroyed by water, was a supply source for the Sergeevka diatomite.

The productivity of diatoms as photosynthetic organisms feeding on nutrients, the production of which requires carbon, depends totally on its concentration in the Earth's atmosphere. We therefore assume that productivity of the diatoms of the Middle Miocene Novokachalinsk diatomite could be influenced by the Monterey Excursion involving very high concentrations of CO_2.

Thus, the reasons for the active development of diatoms in the Middle Miocene (Novokachalinsk diatomite) include a long spring-summer growth season, intensive volcanic activity of the region and, probably, a high CO_2 concentration in the atmosphere during that period. In the colder Pliocene (the Terekhovka and Sergeevka diatomites) the high diatom productivity was ensured by two vegetative periods – spring-summer and autumn, which is confirmed by the presence in taphocoenoses of species preferring colder water.

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