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Nitrogen Isotope Evidence for Changing Arctic Ocean Ventilation Regimes During the Cenozoic

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Key Points:

- Reduced mixing in a tectonically closed Arctic Ocean created the conditions for N₂ fixation by phytoplankton during the early Cenozoic
- A positive shift of 5‰ in the Arctic Ocean δ¹⁵N record implies a biotic response in surface waters to rapidly changing nutrient availability
- This shift indicates a switch to Atlantic-sourced nitrate and marks the opening to the North Atlantic during the early Neogene

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract In this work, I report on the coupling of dinitrogen (N₂) fixation and denitrification in oxygen-deficient waters of the Arctic Ocean during the Paleogene. This coupling fertilized marine phytoplankton growth and favored organic carbon burial. Reduced vertical mixing due to salinity stratification in a tectonically closed oceanic basin created conditions favorable for N₂-fixation by phytoplankton harboring diazotrophic bacterial symbionts. A positive shift of 5‰ in the δ¹⁵N record indicates a change in the main source of biologically available nitrogen due to rapidly changing nutrient availability. I interpret this shift as a switch to Atlantic-sourced nitrate as the main nitrogen source owing to the opening of the Arctic-Atlantic gateway to the northern North Atlantic. While the timing of the opening is still disputed among the available Arctic records, I use evidence from the northern North Atlantic to argue that the Arctic Ocean has been fully ventilated since the early Neogene.

Plain Language Summary A current key element of uncertainty is how the Arctic cryosphere will respond to and drive an increasingly warmer future climate. These questions can be addressed by investigating past climate states, yet paradoxically, the geological archives of the key region, the Arctic Ocean, are underexplored. I use material from a single borehole from the central Arctic Ocean (Lomonossov Ridge) to provide the first insights into Arctic Ocean nutrient utilization over the past 60 million years. The results show the prevalence of oceanic nitrogen fixation as the prime nutrient source to sustain marine productivity in an oxygen-depleted Arctic Ocean during the early Cenozoic (Paleogene—Middle/Late Eocene) with carbon dioxide concentrations significantly higher than today. Moreover, I provide evidence that the opening of the gateway to the North Atlantic caused major changes in nitrogen cycling expressed by a distinct shift to Atlantic-sourced nitrate as the prime source of new nitrogen to the surface ocean.

1. Introduction

Marine dinitrogen (N₂) fixation has received considerable attention in providing an additional source of new nitrogen to support oceanic production in oligotrophic regions (Karl et al., 2002). However, several lines of evidence exist that seasonal N₂-fixation also occurs adjacent to regions of intense subsurface denitrification, in nutrient-rich coastal waters, the deep sea, and cold Arctic waters (Benavides et al., 2018; Blais et al., 2012; Brandes et al., 1998). On longer geological time-scales, however, the role of N₂ fixation in both the global N and C cycles remains poorly constrained. Rau et al. (1987) introduced N₂-fixing cyanobacteria as the prime nutrient N source to explain low sedimentary δ¹⁵N values during Cretaceous black shale deposition. Since then, the coupling of denitrification and N₂ fixation has been frequently linked to short-term global/regional climate perturbations associated with anoxia such as the early Triassic extinction events (Grasby et al., 2016), oceanic anoxic events (Kuypers et al., 2004), and glacial/interglacial cycles in the Cariaco basin (Haug et al., 1998). Recently, Kast et al. (2019) concluded from δ¹⁵N measurements of foraminiferal shell-bound organic matter preserved in sedimentary records that while N₂ fixation is less important than subsurface nitrate as prime nutrient source today, higher rates of N₂ fixation occurred during the Paleogene in the North Pacific. This long-term coupling of denitrification and N₂ fixation in an oxygen-depleted ocean substantiates modern observations (Deutsch et al., 2007) that a decrease in the oceanic N reservoir is counteracted by an increase in the rate of N₂ fixation (and vice versa) within an individual ocean basin implying that the fixed N inventory of the ocean is self-regulating on timescales of million years.

For the Cenozoic Arctic Ocean, the history of the marine nitrogen (N) cycle is poorly constrained. A lack of data prior to the late glacial (Farmer et al., 2021) and considerable uncertainties in determining sources of various

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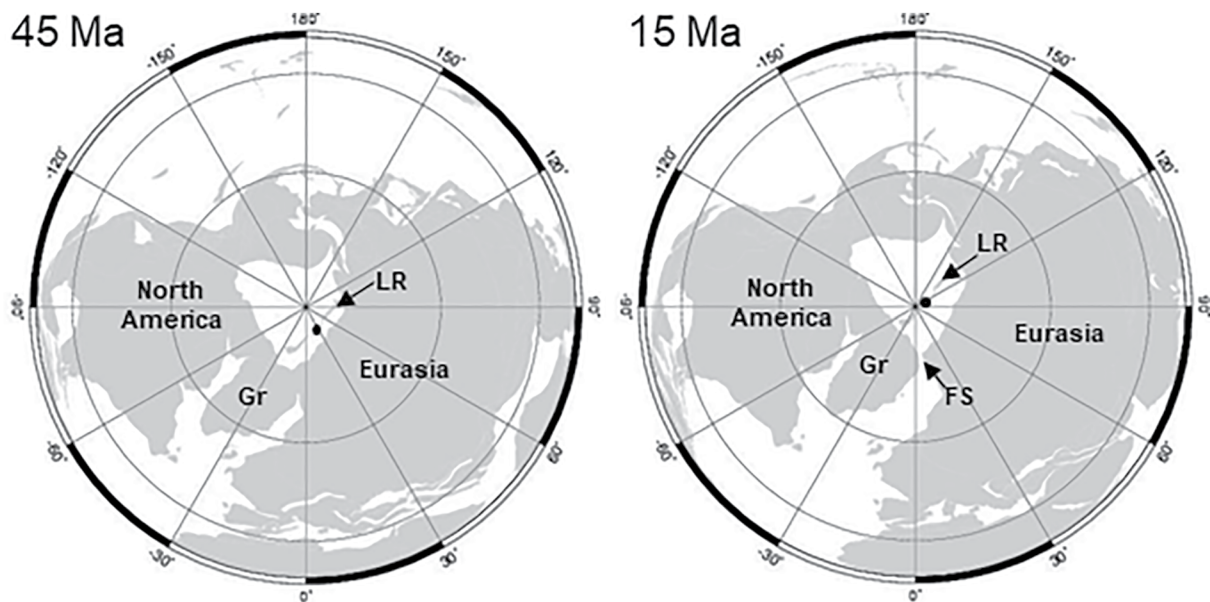


Figure 1. Paleogeographic maps of the Northern Hemisphere at approximately 45 and 15 Ma (source: www.odsns.de). Arctic Coring Expedition borehole: black dot. Gr = Greenland, LR = Lomonosov Ridge, FS = Fram Strait.

nitrogenous fractions in fossil-free deposits (Schubert & Calvert, 2001) hamper inferences on past biogeochemical changes of the N-cycle in the Arctic Ocean. Generally, the oceanic $\delta^{15}\text{N}$ signal depends on the isotopic composition of nitrate and the degree to which this inorganic nitrogen (IN) pool is utilized (Altabet & Francois, 1994). It records the combined isotopic signals of the inputs (mainly N_2 fixation) and outputs (mainly denitrification) of fixed nitrogen to and from the ocean. Today's cryosphere reflects a climate state that developed during a stepwise global cooling during the Cenozoic greenhouse-to-icehouse climate transition (Zachos et al., 2001). The Arctic Ocean was isolated from the global oceanic thermohaline circulation system during most of its geological history (Moran et al., 2006). This oceanographic configuration gradually changed when Greenland and Svalbard began to move apart from each other, initiating the opening of the Arctic-Atlantic gateway (AAG) through the Fram Strait (Figure 1); however, the exact timing of AAG opening, widening, and deepening remains poorly constrained (Jakobsson et al., 2007). Here, I examine Arctic Coring Expedition (ACEX) (International Ocean Discovery Program (IODP) Expedition 302) $\delta^{15}\text{N}$ data from the Lomonosov Ridge (Figure 1), along with productivity (total organic carbon (TOC)), and other nutrient (phosphorus (P)) proxies to reconstruct changes in the Arctic Ocean ventilation regime during the Cenozoic. I show that N_2 -fixing organisms (diazotrophs) fueled marine phytoplankton growth in an oxygen deficient, semi-enclosed Arctic environment during the Paleogene. The high rates of N_2 fixation during the Paleogene ended abruptly with the opening of the AAG and improved ventilation of the Arctic Ocean during the early Neogene.

2. Methods

The sediment sequence (ca. 430 m) recovered from the Lomonosov Ridge (Figure 1) during IODP Expedition 302, is divided into four major lithologic units (Moran et al., 2006) (Figure 2). Age model 1 (AM 1) (Backman et al., 2008) places the upper ~ 198.7 mcd (meter composite depth) in the Early Miocene (~ 18.2 Ma) through the present, while the sequence between 198.7 and 404.8 mcd encompasses the Late Paleocene to early Middle Eocene (~ 44.4 – 56.2 Ma). A hiatus spanning the period ~ 44.4 – 18.2 Ma is located at 198.7 mcd. In an alternate age model (AM 2) Poirier and Hillaire-Marcel (2011) proposed a continuous sedimentary section rather than a hiatus with low mean sedimentation rates (0.2–0.8 cm/kyr) (Figure 2). The new data in this study are discussed regarding both age models. In addition, I include new stratigraphic and geophysical evidence from south of the AAG (Dumais et al., 2021; Rydningen et al., 2020) to substantiate my inferences. Bulk stable nitrogen isotopes ($\delta^{15}\text{N}_{\text{tot}}$) were analyzed for the upper 400 mcd of the record. The data are supplemented by additional TOC (Stein et al., 2006) (Figure 2), and phosphorus data (Figures 2 and 3) (März et al., 2010), as well as paleo-productivity

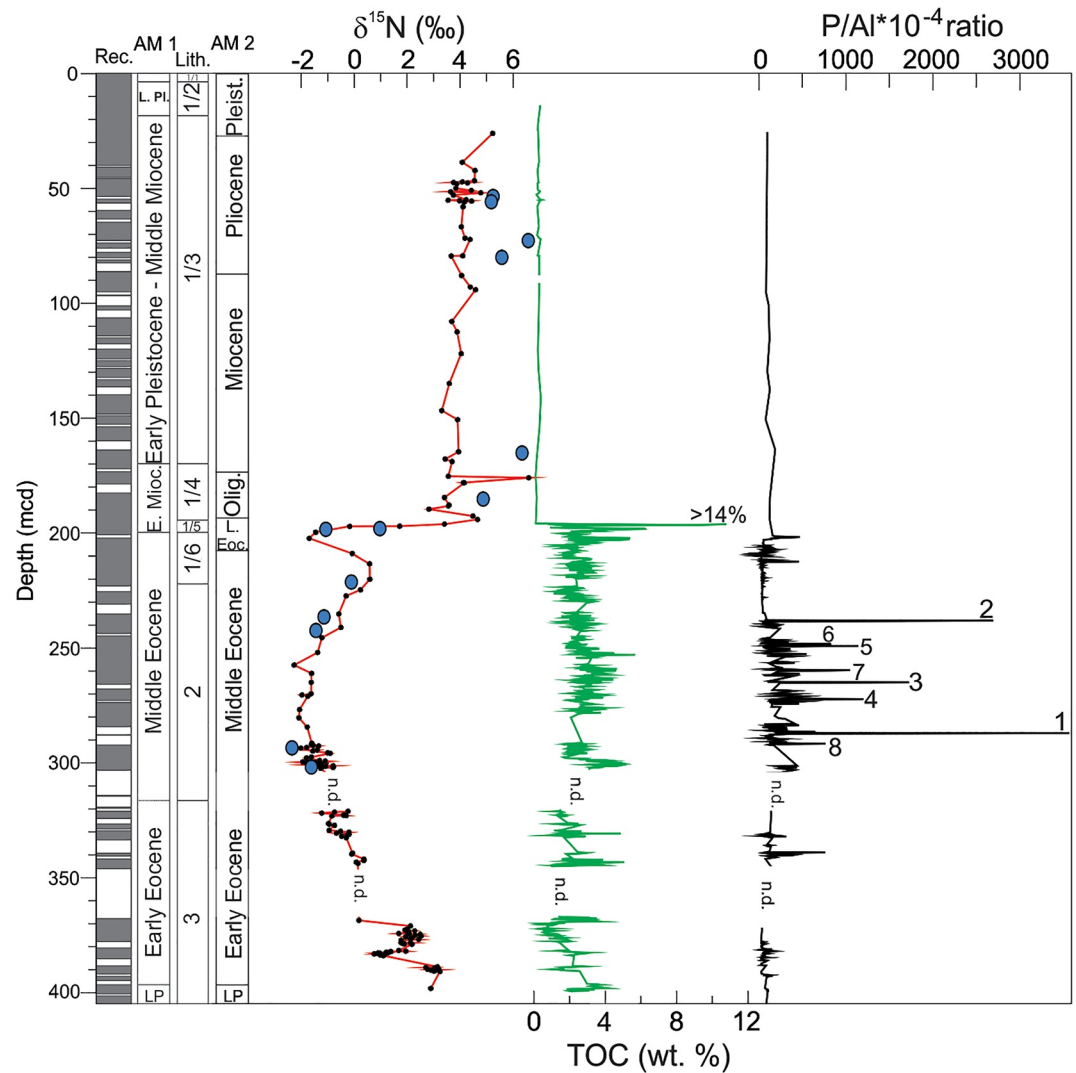


Figure 2. Borehole data from integrated ocean drilling program Expedition 302: Bulk stable nitrogen isotopes ($\delta^{15}\text{N}_{\text{tot}}$) and their organic fraction ($\delta^{15}\text{N}_{\text{org}}$) (blue dots), total organic carbon (TOC, wt. %), and phosphorus and aluminum (P/Al) ratio. Numbers in the P/Al record are identical in Figures 2 and 3 and mark horizons of excess phosphorus. For further information regarding recovery, lithological units, and age assignments see Backman et al. (2008) (AM 1) and Poirier and Hillaire Marcel (2011) (AM 2).

estimates (Figure 4) (Knies et al., 2008) (see Supporting Information S1, for details). Total N and $\delta^{15}\text{N}_{\text{tot}}$ were measured from homogenized subsamples via elemental analyzer isotope ratio mass spectrometry on an ANCA-GSL/20-20 system (Europa Scientific, Crewe, UK). Approximately 20% of the samples were analyzed in duplicate with a mean standard deviation of 0.16 ‰. $\delta^{15}\text{N}_{\text{tot}}$ data are reported as standard δ -values in per mil (‰ vs. air). To test whether the $\delta^{15}\text{N}_{\text{tot}}$ signal primarily records changes in oceanic nitrogen availability rather than admixture of terrestrially derived organic matter, preselected samples treated with a KOB_r-KOH solution were measured for the amount of IN and its isotopic signature ($\delta^{15}\text{N}_{\text{inorg}}$), which represent the amount of NH_4^+ fixed within the clay lattice (Schubert & Calvert, 2001) (Table S1 in Supporting Information S1). Marine organic matter-bound nitrogen (N_{org}) and its isotopic value ($\delta^{15}\text{N}_{\text{org}}$) were calculated using a simple isotope mass balance from the measured amounts of N_{tot} and N_{inorg} and the isotopic values $\delta^{15}\text{N}_{\text{tot}}$ and $\delta^{15}\text{N}_{\text{inorg}}$. The error in determining the $\delta^{15}\text{N}_{\text{org}}$ values was estimated to be better than $\pm 2.1\%$.

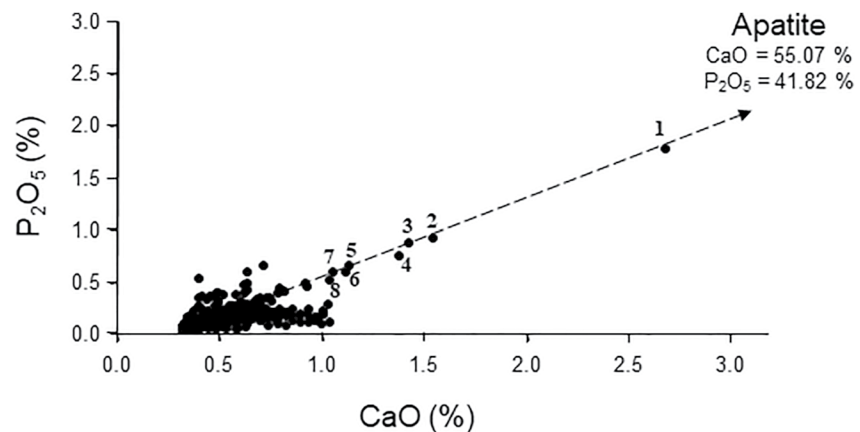


Figure 3. Crossplot of the major elements P_2O_5 and CaO measured by X-ray fluorescence. The black line points to the endmember of both elements for apatite ($P_2O_5 = 41.82\%$; $CaO = 55.07\%$).

3. Results and Discussion

3.1. Cenozoic Arctic Nutrient Utilization

The Cenozoic $\delta^{15}N_{tot}$ record in the Arctic Ocean is characterized by two distinctly different patterns (Figure 2). Sediment $\delta^{15}N_{tot}$ values down to ~ 198.7 mcd range between ~ 3.0 and 6.5 ‰. Below 198.7 mcd, $\delta^{15}N_{tot}$ values vary from ~ -2.0 to 0.6 ‰ with some higher-amplitude variations near the base of the borehole (~ 370 – 400 mcd) (Figure 2, Table S2 in Supporting Information S1). $\delta^{15}N_{org}$ values follow this trend (Figure 2), albeit being slightly enriched in ^{15}N compared to $\delta^{15}N_{tot}$, suggesting that organic matter supply from terrestrial sources has limited influence on the primarily oceanic nitrate record. To date, the overprinting of isotopic signals during particle settling and/or post-depositional isotope fractionation in the modern Arctic Ocean has not been studied, so I am unable to address possible diagenetic alteration during settling in our data set. However, following the reasoning outlined by Altabet and Francois [1,994], I assume that ranges of $\delta^{15}N$ values in the opal- and carbonate-poor sediments above 198.7 mcd (Backman et al., 2008) are similar to ranges of modern surface-generated $\delta^{15}N$ in the Arctic Ocean (Schubert & Calvert, 2001). On the other hand, moderate to good preservation of sedimentary organic matter in productive and oxygen-depleted settings is known to produce little diagenetic alteration of $\delta^{15}N$ values compared to sinking particles (Altabet, Murray, & Prell, 1999; Altabet, Pilskaln, et al., 1999). Despite having only been studied in modern settings, similar conclusions may be drawn from the “Black Sea-type,” oxygen-depleted conditions in the Paleogene Arctic Ocean below 198.7 mcd (Figure 2) (Stein et al., 2006).

Indeed, the $\delta^{15}N_{tot}$ and $\delta^{15}N_{org}$ values above 198.7 mcd concur with modern values in the AAG region (Figure S1 in Supporting Information S1), suggesting that N-containing biomass is stimulated by nutrients supplied by deep waters to the photic zone. Contemporary $\delta^{15}N_{tot}$ values in surface sediments (0–2 cm) increase with decreasing nitrate concentrations from the AAG region to the central Arctic Ocean (Table S3 and Figure S1 in Supporting Information S1) suggesting, by analogy, that $\delta^{15}N_{tot}$ values above 198.7 mcd reflect supply of Atlantic-sourced nitrate by vertical mixing and incomplete nutrient consumption. Using the stratigraphic information for the upper 198.7 m, minor deviations in the $\delta^{15}N$ record from modern $\delta^{15}N_{tot}$ ranges in the AAG region (Figure S1 in Supporting Information S1) imply that similar environmental conditions as today (i.e., low productivity, seasonal to permanent sea ice coverage) have occurred since the Early Miocene (AM 1) or Late Eocene (AM 2) times (Figure 2).

In contrast, low $\delta^{15}N_{tot}$ values (<1 ‰) from the Early to Middle Eocene (AM 1) or Late Eocene (AM 2) (Figure 2) are associated with organic-matter-rich deposits (Figure 2). They concur with higher paleo-productivity values (>50 $gCm^{-2}a^{-1}$) (Knies et al., 2008), and good preservation of organic matter in fresh to brackish, anoxic, “Black Sea-type” conditions (März et al., 2010; Stein et al., 2006). Thus, oxygen levels in the water column were low enough to have likely favored denitrification, which should have resulted in ^{15}N enrichment in the underlying sediments (Cline & Kaplan, 1975). However, if N_2 fixation was occurring in surface waters, the biological input of the diazotroph biomass (Carpenter et al., 1997) would lead to significantly reduced values of $\delta^{15}N$ for particulate nitrogen. Diazotrophs are taxonomically diverse and include a variety of cyanobacteria and bacteria. Their

N_2 -fixation is accompanied by only modest isotopic discrimination, such that diazotroph organic matter is slightly depleted relative to atmospheric N_2 [Montoya, 2008, and references therein]. Field measurements show that marine diazotrophs, including the colonial cyanobacterium *Trichodesmium* have an average $\delta^{15}N$ of -0.5‰ (Carpenter et al., 1997).

The correspondence between high organic matter supply, excess phosphate as indicated by high P/Al ratios, and the occurrence of persistently low sedimentary $\delta^{15}N_{\text{tot}}$ values below 198.7 mcd (Figure 2) suggest that atmospheric dinitrogen (N_2) fixation may be the prime nutrient N source for enhanced productivity and organic burial in a well-stratified, anoxic water mass. Other sources of nitrogen such as rivers, rain, or exchange with open-ocean water are unlikely because the isotopic value of the nitrogen inventory—assuming modern conditions—would be ^{15}N -enriched. Reduced ocean mixing due to strong thermal ocean stratification can result in nitrate limitation in the photic zone and may have given N_2 -fixing organisms a competitive ecological advantage. The consistently low sedimentary $\delta^{15}N_{\text{org}}$ may have resulted from the export of newly fixed N_2 by phytoplankton into sinking N. Alternatively, conditions in the Arctic Ocean water column may have favored complete consumption of nitrate by denitrification as has been proposed to explain the low $\delta^{15}N$ values of sinking and sedimentary organic matter in the Cariaco Basin (Thunell et al., 2004). Excess phosphate as indicated by high P/Al ratios in these laminated, organic rich sediments (Figure 2) suggests that some P that regenerated from organic phases under anoxic conditions may be retained in authigenic mineral phases during diagenesis (Ruttenberg &

Berner, 1993). As known from modern phosphorite environments, decreasing pore-water concentrations of phosphate and fluoride in near-surface sediments reflect the removal of both ions from pore water as they are incorporated into authigenic mineral phases. Precipitation of carbonate fluorapatite concomitant with the highest P concentration (see numbers in Figure 2) is inferred by the positive correlation between CaO and P_2O_5 (Figure 3). This supports the idea that phosphogenesis and water-column denitrification are closely coupled processes in oxygen-depleted environments (Ganeshram et al., 2002). However, wintertime mixing in the “Black Sea-type” Paleogene Arctic Ocean occasionally replenished surface nutrients with low N relative to P (low N:P ratio), which if followed by Redfield-type nutrient drawdown would create an environment favoring the growth of N_2 fixers until P became depleted (Tyrrell, 1999).

3.2. Tectonic Control on Arctic Ocean Nutrient Regimes

The termination of the coupling between denitrification and N_2 fixation is documented by a positive shift ($\sim 5\text{‰}$) in the $\delta^{15}N_{\text{tot}}$ record (Figure 4) suggesting a rapid adjustment in nutrient availability in response to a changing environment. Following modern observations of nutrient utilization in the Arctic Ocean (Schubert & Calvert, 2001), the progressive rise of N-isotopes from -1 to $\sim 4\text{‰}$ suggests increased availability of Atlantic-sourced nitrate in the photic zone and incomplete nitrate consumption due to weaker ocean stratification. It marked the end of ocean stagnation in an isolated, well-stratified Arctic Ocean with high nutrient inventories. The data of gradual recovery from ocean stagnation corroborate evidence for alternating fresh to marine conditions, improved ocean ventilation, and cooler climate (März et al., 2010; Moran et al., 2006). With the upwelling of $\delta^{15}N$ -enriched water masses into the photic zone, residual nitrate gradually replaced N_2 -fixing cyanobacteria as the primary N-source utilized by the marine phytoplankton that fueled the biological activity, as indicated by high paleo-productivity ($>200\text{ g/cm}^3/\text{yr}$) (Figure 4). With the evolution of a permanent sea ice cover thereafter (Darby, 2014; Krylov et al., 2008) paleo-productivity declined significantly ($<20\text{ gCm}^{-2}\text{a}^{-1}$) (Figure 4).

The timing of the prominent environmental change, $\sim 36\text{ Ma}$ versus $\sim 18\text{ Ma}$, is debatable and depends on the age model that is used (Figure 4). Clearly, the enhanced availability of Atlantic-sourced nitrate in the central Arctic Ocean

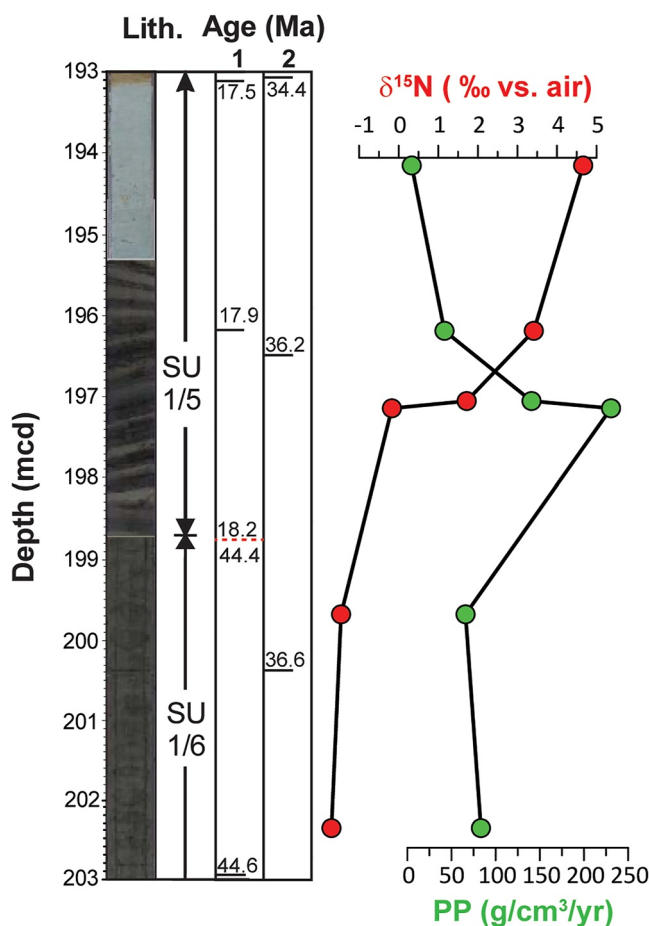


Figure 4. Nitrogen isotopes and paleo-productivity estimates during Mid-Cenozoic times. Borehole images, lithological units, and age fix points according to Backman et al. (2008) (AM 1) and Poirier and Hillaire Marcel (2011) (AM 2).

requires northward transport of Atlantic-derived water masses through the AAG. One argument for the change in Arctic Ocean ventilation during the late Eocene (~36.3–36.4 Ma) is the shift from a transpressional to a transtensional tectonic regime of the strike-slip motions between Greenland and Svalbard after ~34 Ma (Piepjohn et al., 2016), allowing the first narrow connection and thus water mass exchange between the Arctic and the Atlantic oceans. The time delay (~2 Ma) between the change in Arctic Ocean ventilation (~36 Ma) and inferred AAG opening (<34 Ma) may be explained by the uncertainties in the ACEX chronology for AM 2. However, recent coupled atmosphere-ocean model results for the late Eocene through the Eocene/Oligocene transition suggest a lagoonal to estuarine circulation system in the Arctic Ocean and northern North Atlantic (Stärz et al., 2017), implying that the Atlantic water supply to the Arctic Ocean was restricted during the inferred $\delta^{15}\text{N}$ shift (~36 Ma). Furthermore, improved Arctic Ocean ventilation may be established only when the AAG was 100 km wide and 1,000 m deep (Thompson et al., 2012), a scenario that is not very likely before the early Neogene. Indeed, new interpretations from AAG aeromagnetic data (Dumais et al., 2021) suggests a major spreading and reorganization of the Fram Strait at approximately 20 Ma, allowing for break-up at several locations in the wider AAG region to initiate simultaneously. An early Neogene age for northward transport of Atlantic-sourced water masses through the AAG is also supported by well-dated contourite drift deposits underlying the North Atlantic Current south of the gateway (Rydningen et al., 2020). The onset of drift growth during the early Neogene marks the reorganization of the ocean circulation south of the AAG and is likely the consequence of large volumes of Atlantic derived water masses entering the Arctic Ocean. The latter explains the positive shift (~5 ‰) in the ACEX $\delta^{15}\text{N}_{\text{tot}}$ record and thus change toward Atlantic-sourced nitrate supplied to the photic zone on Lomonosov Ridge during the early Neogene. Paleogeographic and paleotectonic reconstructions by Blakey (2021) confirm the widening of the AAG during the early Miocene (~20 Ma). More precisely, geophysical evidence from the first oceanic crust formation within the AAG between chrons 7n.1 and 6AA (24.16–21.08 Ma) (Jokat et al., 2016) and findings of modern-like three-layer stratification in the Arctic Ocean since the Early Miocene (Hossain et al., 2021) substantiate my inferences that changes in nutrient sources and thus an open-ocean ventilation regime in the Arctic have prevailed since the early Neogene.

4. Conclusions

From the study of the nitrogen isotopic composition of organic matter in Cenozoic Arctic Ocean sediments, I conclude that the opening of the AAG had a major impact on the marine nitrogen cycle in the Arctic Ocean. In a tectonically closed and well-stratified oceanic basin, the coupling of N_2 fixation and denitrification under oxygen-depleted conditions stabilized the N-inventory over millions of years during the Paleogene. A positive shift (~5 ‰) in the $\delta^{15}\text{N}$ record indicates a switch to Atlantic-sourced nitrate as the principal source of new nitrogen to the surface ocean during the early Neogene. With the connection to the Atlantic Ocean established, the development of modern ice-covered conditions, and low productivity, the variable inflow of nutrient-enriched Atlantic-derived water masses may have caused different relative nitrate utilization rates of phytoplankton in a fully ventilated Arctic Ocean.

Data Availability Statement

All data available in supporting information at <https://doi.org/10.6084/m9.figshare.19746355.v1>.

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