ARTICLE IN PRESS

Deep-Sea Research II ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at ScienceDirect

Deep-Sea Research II

journal homepage: www.elsevier.com/locate/dsr2

Aspects of the marine nitrogen cycle of the Chukchi Sea shelf and Canada Basin

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ARTICLE INFO

Keywords:
Denitrification
Nitrification
Chukchi Sea
Canada Basin
Nitrate
Ammonium

ABSTRACT

As a highly productive, seasonally ice-covered sea with an expansive shallow continental shelf, the Chukchi Sea fuels high rates of sedimentary denitrification. This contributes to its fixed nitrogen (N) deficit relative to phosphorus (P), which is among the largest in the global ocean, making the Chukchi Sea severely N-limited during the phytoplankton growth season. Here, we examine aspects of the N cycle on the Chukchi Sea shelf and the downstream Canada Basin using nutrients, dissolved oxygen (O_2), and the stable isotopes of nitrate (NO_3^-). In the northward flow path across the Chukchi shelf, bottom waters experienced strong O_2 drawdown, from which we calculated a nitrification rate of 1.3 mmol m^{-2} d^{-1}. This nitrification was likely primarily in sediments and directly fueled sedimentary denitrification, historically measured at similar rates. We observed significant accumulations of ammonium (NH_4^+) in bottom waters of the Chukchi shelf (up to $>5\ \mu M$), which were inversely correlated with $\delta^{15}N_{NO_3}$, indicating a sediment source of ^{15}N-enriched NH_4^+. This is consistent with a process of coupled partial nitrification-denitrification (CPND), which imparts significant ^{15}N enrichment and ^{18}O depletion to Pacific-origin NO_3^-. This CPND mechanism is consistent with a significant decrease in $\delta^{18}O_{NO_3}$ relative to Bering Sea source waters, indicating that at least 58% of NO_3^- populating the Pacific halocline was regenerated during its transit across the North Bering and Chukchi shelves, rather than arriving preformed from the Bering Sea slope. This Pacific-origin NO_3^- propagates into the Canada Basin and towards the North Atlantic, being significantly ^{15}N-enriched and ^{18}O-depleted relative to the underlying Atlantic waters.

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1. Introduction

The Chukchi Sea is among the most productive regions in the Arctic Ocean (Sakshaug, 2004). It sustains the longest seasonal phytoplankton blooms of any Arctic region (Arrigo and van Dijken, 2011) and attracts significant numbers of migratory seabirds and marine mammals during summer (Loeng et al., 2005). This high productivity is maintained by the primarily northward flow of nutrient-rich water through Bering Strait, which spreads over the expansive Chukchi Sea continental shelf (<200 m depth) and migrates toward the Canada Basin (Codispoti et al., 2005). However, the Chukchi Sea productivity regime is changing rapidly as sea ice is lost from the region. From 1998 to 2009, the open water season has expanded by $\sim$4.5 d yr^{-1}, and proliferation of melt ponds has stimulated massive under-ice blooms, thus allowing phytoplankton net primary production (NPP) to increase substantially (Arrigo and van Dijken, 2011; Brown and Arrigo, 2012; Arrigo et al., 2012).

On this and other shallow and highly productive shelves of the western Arctic Ocean, a large fraction of the organic matter formed in surface waters sinks to the sea floor, fueling not only a productive benthic community but also high rates of sediment denitrification. Chang and Devol (2009) estimate that Arctic shelves contribute 4–13% of the total sink of fixed nitrogen (N) in the global ocean. Due to this significant fixed N loss, NPP on the continental shelves of the western Arctic is widely considered to be N-limited. The Chukchi shelf in particular is among the regions of lowest N* (a measure of the fixed nitrogen excess relative to phosphorus; Gruber and Sarmiento, 1997) in the global ocean (Deutsch and Weber, 2012). In waters with low N*, significant phosphorus (P) remains after N has been exhausted during the growth season. This excess P flows downstream into

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http://dx.doi.org/10.1016/j.dsr2.2015.02.009
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the North Atlantic, where it may fuel high rates of N_2-fixation, thus redressing the significant fixed N loss of Arctic shelves (Yamamoto-Kawai et al., 2006).

Given that N is the limiting macronutrient over the Chukchi shelf, it is important to understand the mechanisms of N loss. Because the water column on Arctic shelves is generally well-oxygenated, denitrification, an anoxic microbial process, is restricted to the sediments. The source of nitrate (NO_3^-) that fuels sediment denitrification is a combination of direct NO_3^- flux from the overlying water column and NO_3^- production via nitrification of sediment ammonium (NH_4^+). Horak et al. (2013) recently showed that the latter mechanism of coupled nitrification-denitrification is the most important mechanism of N loss on the Bering Sea shelf. Furthermore, Granger et al. (2011) showed that unlike denitrification fueled by direct NO_3^- flux from the water column, which leaves no isotopic signature (Brandes and Devol, 1997), coupled nitrification-denitrification leads to isotopic enrichment of fixed N in the Bering Sea water column. It is important to determine whether these same N-loss dynamics apply on the Chukchi shelf, and whether this leads to N isotopic enrichment that propagates into the Arctic Ocean downstream.

In addition to the mechanism of N loss, it is equally important to understand the source of fixed N to this N-limited system. Ultimately, all fixed N of the Pacific Arctic derives from northward flow through Bering Strait, both from the Alaska Coastal Water (ACW) to the east, and especially the nutrient-rich Anadyr water emanating from western Bering Strait (Walsh et al., 1989). However, in a given season, a large fraction of the available N may be locally regenerated from the shallow organic matter-rich sediments. Large fluxes of NH_4^+ out of sediments have been observed on the Chukchi shelf (Henriksen et al., 1993; Devol et al., 1997), providing evidence of local regeneration of N. Conversely, Horak et al. (2013) measured negligible fluxes of dissolved inorganic nitrogen (DIN, nitrate+nitrite+ammonium) from Bering shelf sediments, suggesting that local regeneration is not an important source of N. Resolving the role that sediments play in the local regeneration of N is crucial for understanding the nutrient dynamics of Western Arctic shelves.

Here, we use the stable isotopes of oxygen (^{16}O and ^{18}O) and nitrogen (^{14}N and ^{15}N) in NO_3^-, together with nutrient and dissolved gas tracers, to examine the marine N cycle from the Bering Strait, over the Chukchi shelf, and into the Canada Basin. Our discussion draws on the important study of Granger et al. (2011), which uses similar techniques in the Bering Sea to the south. We focus on both N loss processes and the origins of fixed N in this N-limited system. We give particular attention to the process of nitrification, which has received relatively little attention compared to denitrification, but which is central to the discussion of both N losses (through coupled nitrification-denitrification) and N sources (through local regeneration).

2. Methods

2.1. Field sampling and analysis

We collected water samples for nutrients, dissolved gases, and stable isotope analyses on three cruises aboard the USCGC *Healy* to the Chukchi/Beaufort seas. HLY1001 (June 15–July 22 2010) and HLY1101 (June 25–July 29 2011) comprised the field portion of the NASA program ICESCAPE (Impacts of Climate on the EcoSystems and Chemistry of the Arctic Pacific Environment) focused on the Chukchi Sea shelf, while HLY1003 (September 5–26, 2010) was a primarily mooring service cruise on which we accessed the deeper waters of the Canada Basin (Fig. 1).

At each station, water column profiles of temperature, salinity, and oxygen were measured using a Sea-Bird conductivity, temperature, and depth (CTD) system attached to a rosette. Water was collected at multiple depths into 30 L Niskin bottles. The temperature sensors underwent laboratory calibrations before and after the cruises, and the conductivity and oxygen sensors were calibrated at sea using water sample data.

Velocity measurements were obtained using the *Healy's* hull mounted Ocean Surveyor 150 kHz acoustic Doppler current profiler (ADCP). The data were acquired using the University of Hawaii's UHDAS software and underwent further processing with the CODAS3 software package (see http://currents.soest.hawaii.edu). The velocities were then de-tided using the Oregon State University model (http://volkov.oce.orst.edu/tides; Padman and Erofeeva, 2004). The accuracy of the final velocities is estimated to be $\pm 2\ cm\ s^{-1}$.

2.1.1. Nutrients

For ICESCAPE cruises HLY1001 and HLY1101, discrete water column samples were analyzed on-board with a Seal Analytical continuous-flow AutoAnalyzer 3 (AA3) for concentrations of NO_3^-, nitrite (NO_2^-), NH_4^+, and phosphate (PO_4^{3-}) using standard methods (Armstrong et al., 1967; Bernhardt and Wilhelms, 1967; Kerouel and Aminot, 1997). Nutrients from HLY1003 were frozen and analyzed using standard methods at the University of Alaska Fairbanks. Although freezing can affect measurements of NH_4^+ (Degobbis, 1973), we do not consider this to be problematic for two reasons: first, HLY1003 focused on off-shelf waters, which generally have very low NH_4^+ concentrations; second, our in-depth analyses of NH_4^+ (distribution, changes along the northward flow path, and comparison with isotopic data) were all done on the Chukchi shelf using ICESCAPE nutrient data which were not derived from frozen samples.

2.1.2. Stable isotopes of nitrate and water

Seawater samples were filtered through Sterivex filter units (0.22 μm pore size) into duplicate acid-rinsed 60 mL HDPE bottles, then frozen until analysis. $\delta^{15}N_{NO_3}$ and $\delta^{18}O_{NO_3}$ were measured using the denitrifier method (Sigman et al., 2001; Casciotti et al., 2002). Sufficient sample to yield 20 nmol NO_3 was added to an aliquot of culture containing denitrifying bacteria that lack N_2O reductase activity. The N_2O produced was analyzed by continuous flow isotope ratio mass spectrometry on a Thermo-Finnigan Delta^{PLUS} IRMS. Isotope ratios are reported using delta notation as follows:

$$\delta^{15}N_{NO_3}(‰) = \left[\left(^{15}N/^{14}N_{sample}\right)/\left(^{15}N/^{14}N_{standard}\right) - 1\right] \times 1000,$$

and

$$\delta^{18}O_{NO_3}(‰) = \left[\left(^{18}O/^{16}O_{sample}\right)/\left(^{18}O/^{16}O_{standard}\right) - 1\right] \times 1000$$

The $^{15}N/^{14}N$ reference standard is N_2 in air, while the $^{18}O/^{16}O$ reference standard is Vienna Standard Mean Ocean Water (VSMOW). Analyses were referenced to injections from a laboratory standard N_2O tank and standardized using reference materials USGS-32, USGS-34, and USGS-35 (Böhlke et al., 2003).

$\delta^{18}O_{H_2O}$ was measured as in Cooper et al. (2013), and is reported in the standard delta notation.

2.1.3. Particulate organic nitrogen

For ICESCAPE cruises HLY1001 and HLY1101, particulate organic nitrogen (PON) samples were collected for $\delta^{15}N$ analysis by filtering water samples onto pre-combusted (450 °C for 4 h) 25 mm Whatman GF/F filters (nominal pore size 0.7 μm). Filter blanks were produced by passing ~50 mL of 0.2 μm filtered seawater through a GF/F. All filters were then immediately dried

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