



Silica diagenesis and benthic fluxes in the Arctic Ocean



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ABSTRACT

Early diagenetic processes play an important role in the environmental cycling of silicic acid (H_4SiO_4), one of the most important macronutrients in the ocean. Here, we present the first compilation of silicic acid concentrations in sediment pore water sampled in various parts of the Central Arctic Ocean. Based on these data, we calculate benthic fluxes of silicic acid from the sediments to Arctic bottom waters and interpret the pore water profiles in terms of silica dissolution–precipitation reactions deeper within the sediments. Silicic acid concentrations are very low in Arctic pore waters to sediment depths of ~8 m, with maxima ranging from 30 $\mu\text{mol/L}$ on the Mendeleev Ridge to 300 $\mu\text{mol/L}$ on the Alpha Ridge. These values are 1–2 orders of magnitude lower than silicic acid pore water concentrations in Atlantic, Indian, and Pacific Ocean sediments, reflecting the low biosilica productivity in the oligotrophic, seasonally ice-covered Arctic Ocean. Low silicic acid pore water concentrations translate into equally low benthic silicic acid fluxes, ranging between 1 and 15 $\text{mmol m}^{-2} \text{a}^{-1}$ ($1 * 10^9$ to $1.5 * 10^{10} \mu\text{mol km}^{-2} \text{a}^{-1}$) at most study sites. With increasing sediment depth, asymptotic patterns typically observed in marine sediments occur at only five of twelve study sites. Deviations from this asymptotic shape at the other sites most likely reflect specific sediment intervals with elevated rates of biosilica dissolution (potentially from underlying Eocene–Cretaceous strata) in association with iron oxide dissolution, or with the consumption of silicic acid by neo-formation of authigenic clay minerals.

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1. Introduction

In global biogeochemical cycles and the Earth's climate system, solid phase silica (SiO_2) and dissolved silicic acid (H_4SiO_4 , here termed DSi) species play a crucial role. Silicic acid is the most important macronutrient for marine primary producers that form hard parts of biogenic opal (e.g., diatom frustules). Thus, DSi is an essential component of the biological carbon pump in the world oceans and a co-regulating factor of atmospheric CO_2 concentrations (De La Rocha and Passow, 2012). Biogenic synthesis of opal in the photic zone of the oceans removes DSi from the bioavailable pool, and a substantial fraction of this biogenic opal settles to the sea floor. Within the benthic boundary layer, remineralisation processes as well as the undersaturation of sea water with respect to DSi lead to the partial dissolution of biogenic opal, and remobilization of DSi to the bottom waters. Depending on water depth, upwelling and mixing conditions, the recycled DSi can be re-supplied to the photic zone as a bioavailable nutrient, closing the marine silica cycle (DeMaster, 1981; Treguer and De La Rocha, 2013). However, the benthic recycling of biogenic opal at the sea floor is incomplete, and part of it is buried into the sediments. Here it can be subjected to early diagenetic processes that lead to its dissolution, followed either by DSi

diffusion back to the bottom waters, or by re-precipitation of authigenic clay minerals that ultimately remove DSi from the bioavailable marine pool over geological timescales (recent review by Aller, 2014). The transformation of opal-A to opal-CT is another process involved in marine silica diagenesis, but it usually occurs during deeper sediment burial and/or in biosiliceous sediments (e.g., Botz and Bohrmann, 1991; Iwasaki et al., 2014). Numerous studies have investigated silica diagenesis in sediment pore water of the Atlantic, Indian, Pacific, and Southern Oceans (e.g., Gieskes, 1975; Sayles, 1981; Jahnke et al., 1982; Gehlen et al., 1995; McManus et al., 1995; Sayles et al., 1996, 2001; Hensen et al., 1998; Ragueneau et al., 2000, 2001; DeMaster, 2002; Van der Weijden and Van der Weijden, 2002). In addition, experimental studies were conducted to understand the dissolution and re-precipitation dynamics of silica in marine sediments (e.g., Mayer et al., 1991; Van Cappellen and Qiu, 1997a,b; Dixit et al., 2001; Loucaides et al., 2012). Nevertheless, while the importance of silica diagenesis has been recognised and studied in most ocean basins, only very few DSi pore water data are available from the Central Arctic Ocean (Gobeil et al., 1991; Backman et al., 2006). This lack of data is likely due to the remoteness of the Arctic Ocean, but also to the fact that biogenic opal contents in Arctic surface sediments rarely exceed 5 wt.% (Nürnberg, 1996; Lisitzin, 1996; Hoffmann et al., 2013). In addition, in the Arctic Ocean, DSi is not considered a limiting nutrient for primary productivity (Popova et al., 2012), as it is supplied by nutrient-rich North Pacific surface waters through the Bering Strait and by circum-Arctic rivers

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(Gordeev et al., 1996; Holmes et al., 2012). On the other hand, Arctic deep waters are exported to the oligotrophic Atlantic Ocean where their dissolved nutrient load could stimulate primary productivity in upwelling areas (Torres-Valdes et al., 2013).

Dissolved silica concentrations have frequently been applied as a “conservative” tracer for North Pacific water masses or river discharge into the Arctic Ocean (e.g., Ekwurzel et al., 2001; Roeske et al., 2012). The benthic recycling component has so far been neglected in the Arctic silica budget, but Nishino et al. (2009) reported a deeper DSi maximum in the sea ice reduction zone along the western Arctic Ocean shelf-upper slope that is interpreted as DSi diffusing out of the surface sediments. Arctic shelf areas might thus increasingly serve as DSi sources to the water column as summer sea ice disappears. As surface sediments release this DSi largely via diffusion, benthic recycling should provide DSi to bottom waters largely independent of seasons, in contrast to riverine DSi which is mainly delivered during the spring freshet. This recycled benthic DSi is eventually exported to the North Atlantic via Davies Strait and Fram Strait (Torres-Valdes et al., 2013). In terms of its isotopic composition, DSi remobilized from sediments due to biogenic opal dissolution has a distinctly light isotopic signature, i.e., a fractionation towards lower $\delta^{30}\text{Si}$ values compared to the source material (Demarest et al., 2009; Wetzel et al., 2014). Even quantitatively small inputs of benthic DSi to Arctic and North Atlantic bottom waters will influence their Si isotopic compositions, and will consequently affect calculations of global ocean $\delta^{30}\text{Si}$ budgets as well as reconstructions of deep ocean circulation (De Souza et al., 2014; Brzezinski and Jones, in press). For these reasons, an improved knowledge of the early diagenetic cycling of silica in Arctic Ocean sediments is required, and we present the first systematic study of Arctic Ocean pore water DSi concentrations, fluxes, and diagenetic processes.

2. Material and methods

Pore water samples were obtained during Expeditions ARK-XXIII/3 (2008) and ARK-XXVI/3 (2011) of the German icebreaker RV *Polarstern* to the Arctic Ocean (Jokat, 2009; Schauer, 2012). Coring stations were located in the Northwest Passage, at the Beaufort and East Siberian Sea continental margins, on the Alpha Ridge, Mendeleev Ridge, Lomonosov Ridge, in the Makarov Basin and the Nansen Basin near Gakkle Ridge (Table 1, Fig. 1). Nine multicorer tubes (MUC), eight gravity cores (GC), and four box-shaped gravity cores (“Kastenlot”, KL) were sampled for pore waters using rhizone samplers (~0.1 μm average pore size), with a vacuum being applied using 12 mL plastic syringes with wooden stoppers (Seeberg-Elverfeldt et al., 2005; März et al., 2011). Sampling resolution ranged from 1 cm (in MUCs, directly above and below the sediment–water interface) to 25 cm (in GCs and KLs). Pore water splits of 2 mL were acidified with 10 μL of concentrated HCl and stored in plastic vials at 2 °C until further analysis. Total dissolved silica, iron, potassium and magnesium concentrations of pore waters (and partly overlying bottom waters) were determined by inductively

Table 1
Sampling sites reported in this publication, with geographic locations, latitude and longitudes, full station names (acronyms used in the manuscript are fat), and water depths.

Geographic location	Longitude & latitude	Station number	Water depth (m)
Northwest Passage	90.985 W, 72.266 N	PS72/287	353
Beaufort Margin	137.180 W, 71.269 N	PS72/291	1548
East Siberian Margin	171.493 W, 77.604 N	PS72/340	2351
	179.053 W, 77.304 N	PS72/343	1226
Mendeleev Ridge	175.741 W, 80.510 N	PS72/410	1801
	178.515 W, 80.279 N	PS72/413	1263
	175.745 E, 80.543 N	PS72/422	2530
Nansen Basin	60.155 E, 86.443 N	PS78/206	1791
Lomonosov Ridge	115.189 W, 89.248 N	PS78/220	1668
	149.797 E, 84.673 N	PS78/248	1620
Alpha Ridge	137.750 E, 84.905 N	PS78/231	1618
	154.427 W, 83.748	PS78/237	2360

coupled plasma optical emission spectrometry (ICP-OES, iCAP 6000, ThermoScientific) using matrix-matched calibration solutions (Atlantic Seawater, Osil, UK) to which In or Sr (for Si) and Be (for Fe) were added as internal standards. Accuracy was validated by spiked seawater SRM (Cass-5, Nass-5, NRC Canada), and three different Si and four different Fe wavelength lines. Accuracy was within 3% for K, Mg, Si and 5% for Fe. Precision was checked by repeat analyses of reference solutions (Nass-5, Cass-5, Atlantic Seawater) and selected samples, and produced errors <9% for K, Mg, Si, and <13% for Fe.

Benthic DSi fluxes were calculated (assuming steady state conditions) according to Fick's First Law of Diffusion (Schulz, 2006):

$$J_{sed} = \Phi * D_{sed} * dC/dx,$$

where J_{sed} = diffusive H_4SiO_4 flux across the sediment–water interface, Φ = sediment porosity, D_{sed} = H_4SiO_4 diffusion coefficient, and dC/dx = H_4SiO_4 concentration gradient.

Without discrete porosity measurements, a conservative porosity of 0.8 was adopted (0.8–0.9 is typical for hemipelagic surface sediments; Schulz et al., 1994; Hensen et al., 1998). Linear DSi gradients were fitted through data points directly below (1–3 cm depth) and, where available, above the undisturbed sediment–water interface in the MUCs. Where no bottom water DSi concentrations were analysed directly, values from Middag et al. (2009) and Giesbrecht et al. (2013) were adopted for deep water masses in the Eurasian and Canada Basins, respectively. The diffusion coefficient of DSi at a temperature of 2 °C was corrected for sediment tortuosity following Archie's Law (Boudreau, 1996). An example calculation can be found in Appendix A. The data are available for download from the open access data base Pangaea (www.pangaea.de).

3. Results

3.1. Silicic acid pore water concentrations and benthic fluxes

Maximum DSi concentrations in the MUCs (core lengths 12–45 cm) ranged from ~12 $\mu\text{mol/L}$ (station 413) to ~280 $\mu\text{mol/L}$ (station 287) (Fig. 2). Bottom water DSi concentrations determined in the supernatant water of the MUC tubes ranged from ~8–16 $\mu\text{mol/L}$, in agreement with bottom water DSi concentrations reported by Middag et al. (2009) and Giesbrecht et al. (2013). At all stations (except 343), DSi concentrations generally increased with sediment depth. Deviations from the general downcore trends are likely related to bioturbation and/or non-continuous sedimentation processes (especially at station 343 which is located directly on a continental margin) (Jokat, 2009; Stein et al., 2010).

Maximum DSi concentrations in the GCs and KLs (core lengths 360–800 cm) ranged from ~35 $\mu\text{mol/L}$ (station 413) to ~300 $\mu\text{mol/L}$ (station 287) (Fig. 3). At all stations but 287, DSi concentrations generally increased with sediment depth. The shape of the profiles can be grouped into four categories (Fig. 3): The classical asymptotic DSi profiles (Boudreau, 1990; McManus et al., 1995) (category A) are clearly expressed in five of the twelve cores. Three cores exhibit overall asymptotic DSi profiles with intermittent excursions to significantly higher values (category B). Three cores do not show asymptotic decreases, but increases in DSi within sediment depth (category C). Finally, one core displays a downcore DSi decrease in the shape of a “concave-up” profile (category D). Where pore water profiles approach asymptotic shapes, the maximum DSi concentrations at depth (“saturation concentrations”) are mostly between 70 and 100 $\mu\text{mol/L}$.

Benthic DSi fluxes calculated for the nine MUC stations range across roughly two orders of magnitude (Table 2), from ~0.8 $\text{mmol m}^{-2} \text{a}^{-1}$ (~8 * 10⁸ $\mu\text{mol km}^{-2} \text{a}^{-1}$; station 413) to ~67 $\text{mmol m}^{-2} \text{a}^{-1}$ (~6.7 * 10¹⁰ $\mu\text{mol km}^{-2} \text{a}^{-1}$; station 287), with values between 1 and 15 $\text{mmol m}^{-2} \text{a}^{-1}$ (between 1 * 10⁹ and 1.5 * 10¹⁰ $\mu\text{mol km}^{-2} \text{a}^{-1}$) at most stations.

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