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Coupling of the distribution of silicon isotopes to the meridional overturning circulation of the North Atlantic Ocean

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ABSTRACT

The distribution of silicon isotopes within silicic acid, $\delta^{30}\text{Si}(\text{OH})_4$, was examined along a section in the North Atlantic from the Cape Verde Islands off Africa to Cape Cod, Massachusetts in North America. Surface water displayed elevated $\delta^{30}\text{Si}(\text{OH})_4$ associated with biological fractionation of Si during silica production. Below 300 m variations in $\delta^{30}\text{Si}(\text{OH})_4$ were closely tied to the distribution of water masses as diagnosed through optimum multiparameter analysis, confirming a tight relationship between $\delta^{30}\text{Si}(\text{OH})_4$ and the meridional overturning circulation in the Atlantic. A linear relationship between $\delta^{30}\text{Si}(\text{OH})_4$ and the inverse of silicic acid concentration supported control of Si isotope distribution by conservative mixing of end member water masses of different isotopic composition in the Atlantic. There was a suggestion of a weak local minimum in $\delta^{30}\text{Si}(\text{OH})_4$ in deep waters above the Trans-Atlantic Geotraverse hydrothermal zone on the mid-Atlantic Ridge consistent with the light $\delta^{30}\text{Si}(\text{OH})_4$ of hydrothermal waters. The lightest $\delta^{30}\text{Si}(\text{OH})_4$ values were observed in the deep western and deep eastern basins where Antarctic Bottom Water (AABW) dominated. The heaviest values in subsurface waters occurred in North Atlantic Deep Water due to strong ventilation and the contribution of heavy northern source waters that are influenced by the Arctic Ocean. The concept of a silicon isotope bipole is introduced to explain how the isotopic differences between the northern and southern end-member water masses arise, and how they influence Si isotope distributions. Northern end-member water masses are heavy due to the influence of the Arctic Ocean. Bottom topography prevents light deep waters from entering the Arctic and the further removal of light isotopes through local biological productivity results in extremely heavy $\delta^{30}\text{Si}(\text{OH})_4$ within the Arctic. Light AABW dominates the southern end member. The Southern Ocean silicic acid trap distills heavier isotopes of Si out of the Southern Ocean as preformed silicic acid within Antarctic mode waters, while retaining light isotopes that become incorporated into AABW. The influence of the silicon isotope bipole is predicted to be strongest in the Atlantic as the net flow of waters through the Arctic Ocean is from the Pacific to the Atlantic.

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1. Introduction

Silicon isotopes have proven to be a powerful tool for understanding diatom productivity on unprecedented spatial and temporal scales in both the modern and past ocean (e.g. [Beucher et al., 2007](#); [Brzezinski et al., 2002](#); [De La Rocha et al., 1998](#); [Fripiat et al., 2011](#); [Reynolds et al., 2008](#)). Silicon isotopes in biogenic silica, $\delta^{30}\text{Si}\text{-bSiO}_2$, and in silicic acid, $\delta^{30}\text{Si}(\text{OH})_4$, are increasingly being used to assess the roles of diatoms and of changes in silicic acid supply in past shifts in ocean productivity and their implication for climate. Application of the proxy requires several untested

assumptions. Two of the most common assumptions made when interpreting sedimentary silicon isotope records are that both the silicic acid concentration, $[\text{Si}(\text{OH})_4]$, and the silicon isotopic composition of ventilating waters masses are unique and invariant over time ([Beucher et al., 2011](#); [Fripiat et al., 2011](#); [Reynolds et al., 2006](#)). The issue of changes in water mass $[\text{Si}(\text{OH})_4]$ over time is being addressed through recent advances in the use of Si isotopes within sponge spicules as a proxy for the $[\text{Si}(\text{OH})_4]$ of deep waters ([Ellwood et al., 2010](#); [Hendry et al., 2010](#); [Wille et al., 2010](#)). Here we address the fundamental issue of whether major water masses in the modern ocean possess the unique $\delta^{30}\text{Si}(\text{OH})_4$ signatures predicted by models ([Matsumoto et al., 2012](#); [Reynolds, 2009](#); [Wischmeyer et al., 2003](#)).

Both laboratory ([De La Rocha et al., 1997](#); [Demarest et al., 2009](#)) and field (e.g. [Cardinal et al., 2005, 2007](#); [De La Rocha et al., 2000](#);

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Fripiat et al., 2011; Varela et al., 2004) studies indicate that the production and recycling of diatom silica are the dominant biogeochemical processes controlling silicon isotope distributions in the global ocean. Fractionation of isotopes of Si occurs during both the production of opal by diatoms (De La Rocha et al., 1997) and during opal dissolution (Demarest et al., 2009) with each process favoring the incorporation (production) or release (dissolution) of the lighter isotopes of Si. Silica production and dissolution are decoupled in space and time with silica production being confined to surface waters following the injection of nutrients from depth, while silica dissolution occurs mainly following cell death, and throughout the water column, with a vertical distribution governed by the remineralization length scale for biogenic silica. Numerical simulations suggest that the coupling of these processes to the meridional overturning circulation (MOC) creates predictable and unique $\delta^{30}\text{Si}(\text{OH})_4$ signatures within major ocean basins and water masses (Reynolds, 2009).

Reynolds (2009) used a seven box model based on Toggweiler (1999) and the ten box PANDORA model of Broecker and Peng (1986) to examine global marine silicon isotope distributions. Incomplete silicic acid use in the surface waters of the Southern Ocean was a major driver of the model results. $\delta^{30}\text{Si}(\text{OH})_4$ values within waters that subduct to form Antarctic Bottom water (AABW) are light due to high preformed $[\text{Si}(\text{OH})_4]$. Stronger biological depletion of $\text{Si}(\text{OH})_4$ occurs in the waters incorporated into Antarctic Intermediate Water (AAIW) and Subantarctic Mode Water (SAMW) giving those water masses relatively heavy $\delta^{30}\text{Si}(\text{OH})_4$ signatures. The northward flow of deep waters formed in the Southern Ocean strongly influenced the isotopic composition of modeled bottom waters in the Atlantic and Pacific basins. The modeled isotopic signature of North Atlantic Deep Water (NADW) was heavy, largely due to strong ventilation in the North Atlantic resulting in high productivity and low nutrient content. One caveat of current model results is that they do not include the contribution of the Arctic Ocean to the MOC of the North Atlantic. New data indicate that waters in the Arctic are the heaviest of all oceans with deep-water $\delta^{30}\text{Si}(\text{OH})_4$ of $+1.9\text{‰}$ (Varela, pers. com.) such that exchange between the Arctic and the Atlantic will influence the isotopic composition of northern source waters.

The focus of this study was a section across the North Atlantic conducted by the US GEOTRACES program in 2010 and 2011. The North Atlantic Ocean resides at one extreme of the MOC. More than any other basin, nutrient distributions in the North Atlantic are affected by advection. NADW transports waters to the south with northward counter flows in surface waters, AAIW, SAMW and at depth via AABW. Lateral injection is also important, most notably water from the Mediterranean outflow through the Strait of Gibraltar, and water from the Arctic Ocean entering through the Fram Strait off Greenland and through the Canadian Archipelago. The GEOTRACES cruise track from the African Coast near the Cape Verde Islands to the North American coast near Cape Cod, Massachusetts (Fig. 1) traverses the deep western and eastern basins at latitudes where there is a contrast in deep water silicic acid concentrations between basins, and it also intersects a major hydrothermal zone along the mid-Atlantic ridge. The section also spans a large range in diatom productivity from the highly productive upwelling zone off West Africa to the oligotrophic subtropical gyre. This diversity of water masses, silicic acid concentrations and gradients in productivity provide an ideal context for evaluating the coupling of Si isotope distributions and the MOC.

2. Material and methods

2.1. Sample collection

Samples for the measurement of the silicon isotope composition of silicic acid were collected at nine stations from the

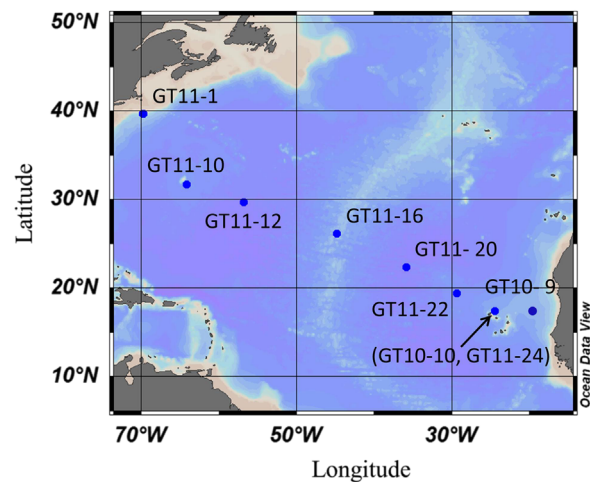


Fig. 1. Map of station locations where $\delta^{30}\text{Si}(\text{OH})_4$ was measured. Numbers denote GEOTRACES station numbers. The prefix USGT11 denotes stations occupied in 2011 with USGT10 denoting stations sampled in 2010.

Cape Verde Islands to the shelf waters off the Northeast coast of the United States (Fig. 1). Two stations were sampled in 2010 on cruise KN199-4 (USGT10-9 and USGT10-12). USGT10-12 was resampled along with six other stations during 2011 on cruise KN204-1. Comparison of data from the one station that was sampled during both years (station USGT10-12 during 2010, station USGT11-24 during 2011) allows an assessment of combined sampling, analytical and environmental variability. The 2011 section also sampled the Bermuda Atlantic Time Series (BATS) station (USGT11-10) which was the site on the earlier US GEOTRACES intercalibration exercise in the Atlantic (Cutter, 2013).

Seawater samples were obtained using a conventional CTD rosette equipped with 30 L Niskin samplers, an oxygen probe, and a seabird 911 CTD. Two liters of seawater were filtered directly from each Niskin sampler into acid-washed polypropylene or polyethylene bottles through silicone tubing using in-line AcroPak[®] filter capsules containing 0.8/0.45 μm Supor[®] membrane filters that had been washed with trace-metal-grade HCl prior to use. Filter capsules and sample bottles were rinsed extensively with sample prior to sample collection. Individual capsules were reused to filter up to 500 L and stored refrigerated between uses. The water samples were tightly capped and stored in the dark at room temperature until analysis. Samples for silicic acid concentration analysis were also drawn from all Niskin bottles.

2.2. Sample processing

Subsamples were prepared for Si isotope analysis by reacting subsamples with acid-molybdate and precipitation of the resulting silicomolybdic acid with triethylamine using a modification of the method of De La Rocha et al. (1996). The original method quantitatively precipitates the Si in samples with $>10 \mu\text{M}$ $\text{Si}(\text{OH})_4$. It was learned that quantitative precipitation of samples containing as little as $1 \mu\text{M}$ silicic acid could be accomplished by waiting a week rather than the recommended 24 h for the precipitate to form. The precipitate was collected by filtration, rinsed and combusted to form solid SiO_2 as in the original method. The SiO_2 was dissolved in HF, precipitated as Cs_2SiF_6 by the addition of CsCl, and analyzed by acid decomposition by IRMS using a modified Kiel III inlet system coupled to a Finnigan MAT 252 IRMS mass spectrometer (Brzezinski et al., 2006). The in-house standard, Big Batch (Brzezinski et al., 2006), was analyzed approximately every 8th sample to monitor system performance. Seawater samples were analyzed in duplicate or triplicate

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