Pre-modern Arctic Ocean circulation from surface sediment neodymium isotopes

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[1] We present a survey of Nd isotopes measured from authigenic phases on surface sediments from the Arctic Ocean seafloor. Comparison with published dissolved water column Nd isotope distributions suggests that ε_{Nd} from sediment leachates accurately represents bottom water circulation of the Arctic Ocean, with Atlantic (~−11 ε_{Nd}) and Pacific (~−5 ε_{Nd}) water end-members clearly distinguishable. Nd isotopic data from the Chukchi margin show several apparent deviations from the expected pattern, which imply that this margin has been a region of sustained shelf-to-basin sediment redistribution and/or a region where deep water cascades downslope, such as through brine rejection during ice formation. These data also provide insights into a pre-anthropocene "baseline" (several thousand years) for investigating Arctic Ocean circulation in a changing climate; the data presented here are indicative that these changes may be impacting the deep as well as surface ocean.
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1. Introduction

[2] Arctic Ocean circulation, ice, and atmospheric conditions are changing rapidly, which is having broad impacts on climate [*Stroeve et al*., 2012], ecology [*Wassmann et al*., 2011], and socioeconomics [*Blunden*, 2012]. However, the observations that document decreasing sea ice extent and volume [*Stroeve et al*., 2012] and increasing freshwater storage [*Proshutinsky et al*., 2002; *Morison et al*., 2012] largely relate to changes in the surface ocean. Far less understood are the ongoing and potential changes in the deep Arctic Ocean [*Rudels*, 2011]. Due to observational scarcity and shifting modern conditions, proxy information from recent geological archives can help to understand the natural state of the Arctic and evaluate if and how the deep Arctic Ocean has changed in time. Here we present isotopic data from the hydrogenous component of seafloor surface sediments to make an initial pre-anthropocenic (i.e., pre-industrial revolution; *Zalasiewicz et al*. [2008]) "baseline," against which to compare recent and future observations and numerical modeling of deepwater circulation.

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

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[3] Surface circulation (summarized in Figure 1) is characterized by the Transpolar Drift to the Atlantic, and longer-lived, recirculating Beaufort Gyre in the western Arctic [e.g., *Proshutinsky et al*., 2002]. The stability of Arctic sea ice is supported by low-salinity surface waters fed by meteoric water, ice melt, and a relatively fresh (~33 practical salinity units) surface water from the Pacific [*Schlosser et al.,* 1994, 1995; *Bauch et al*., 1995; *Ekwurzel et al*., 2001; *McClelland et al*., 2012]. North Pacific water gravitationally flows into the Arctic Ocean across the shallow (40–50 m) Bering Strait due to a climate-controlled ~0.4 m difference between the surface of the Pacific and the Atlantic oceans [*Stigebrandt*, 1984]. Pacific water also ventilates the upper Arctic halocline and can extend to ~200 m depth via mixing with winter waters of the Chukchi shelf, including brines resulting from sea ice formation [*Jones et al*., 2003; *Woodgate et al*., 2005; *Pickart et al*., 2005]. However, an outstanding question is whether any of these upper ocean waters, intrinsically not sufficiently dense to subduct below the halocline, can possibly be considered a source for deep waters in the Arctic, and, if not, what is the source for the saline dense bottom waters in the deep Canada Basin [*Swift et al*., 1983; *Schlosser et al*., 1995]?

[4] Fundamental to this question is the issue that deep Arctic circulation is less well understood compared to the surface ocean. It is generally accepted that the predominant source of intermediate and deep water in the Arctic basins is from the Atlantic [e.g., *Schlosser et al*., 1995; *Rudels*, 2011]. Influences of local deep-water sources are also seen, especially as derived from brine formation and winter convection in the Barents and Kara seas, but the extent of their contribution to deep waters is not well constrained [e.g., *Bauch et al*., 2005; *Rudels*, 2011]. *Quadfasel et al*. [1988] argued that dense shelf waters can account for up to 10% of deepwater production in the polar basins, and *Schlosser et al*. [1995] observed that the dense deep waters of the Canada Basin require an additional source of salt beyond Atlantic-sourced waters, ostensibly from shelf brines. However, observational evidence is sparse at best for brine contributions to deep water in the Arctic, perhaps due to an episodic nature of formation and subduction. Compounding these uncertainties, observations of radiocarbon and other water mass tracers indicate that the Canada Basin deep water is clearly distinct from its Eurasian basin counterpart [*Macdonald et al*., 1993; *Schlosser et al*., 1990, 1995]. Neodymium (Nd) isotopes, which are thought to be "quasiconservative" tracers of ocean circulation [e.g. *Frank*, 2002], hold promise as a means to gain insight into these patterns [*Andersson et al*., 2008]. In the modern Arctic Ocean, the Nd isotope distribution is dominated by Atlantic source water, although circum-Arctic riverine and Pacific-sourced waters also have discernable impacts [*Porcelli et al*., 2009]. These non-Atlantic sources of Nd are not yet fully understood; however, the major features of the Nd

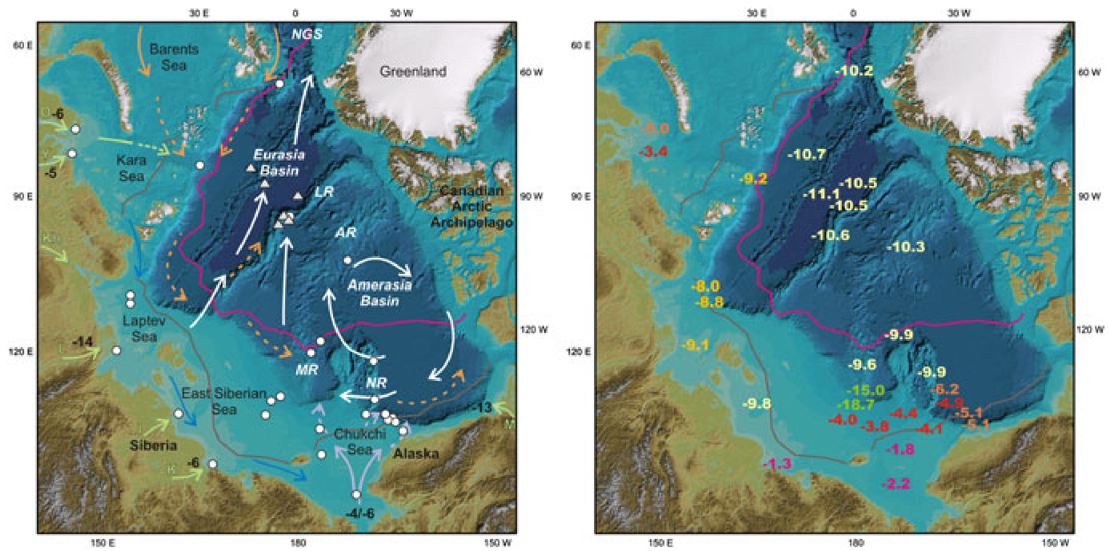

Figure 1. (left) Map of the Arctic Ocean (IBCAO) [*Jakobsson et al.*, 2012] with location of sediment samples: circles, new samples; triangles, earlier data [*Haley et al.*, 2008]. Lines show summer sea-ice margin: 2012 (magenta) and late 20th century mean (brown) (NSIDC data). White arrows, surface Arctic circulation; orange and purple, Atlantic and Pacific waters, respectively (punctured, subsurface); blue, predominant Siberian coastal currents; green, major rivers (with initial letters). NR, MR, AR, and LR, Northwind, Mendeleev, Alpha, and Lomonosov ridges, respectively; NGS, Norwegian-Greenland Sea. (right) Color-coded ε_{Nd} values in sediment samples (first acid leach): yellow, <-9.5; orange, -9.5 to -7.5; pink, -7.5 to -5; red, -5 to -2.5; purple, >-2.5; green, low outliers. Error on ε_{Nd} is 0.3 units (see Supporting Information).

isotope distribution in the modern Arctic do appear to correspond to the general circulation pattern in a predictable way [*Porcelli et al.*, 2009].

[5] Despite these insights, establishing a pre-anthropocenic baseline for Arctic deepwater circulation is becoming increasingly obscured because samples taken today are likely to reflect a changing Arctic condition [e.g., *Jackson et al.*, 2011; *Morison et al.*, 2012]. Here we attempt to reconstruct "recent" (middle/late Holocene to nearly modern) bottom water circulation in the Arctic through the use of Nd isotopes in the hydrogenous coatings of surface sediment.

2. Samples and Results

[6] Sediments were taken from core tops distributed across the Arctic Ocean (HOTRAX'05 multicorer sampling) and the Eurasian Arctic margin (mainly from grabs collected on various occasions) (Figure 1 and Supporting Information, Table 1). Depending on sedimentation at the core sites, sample age may vary between last several decades in depocenters, such as near river mouths and on the outer Chukchi margin, to a few thousands of years in the sediment-starved central Arctic Ocean and on the current-swept Chukchi shelf [e.g., *Darby et al.*, 2009; *Polyak et al.*, 2009]. These core top samples were leached using the protocols of *Haley et al.* [2008] (see Supporting Information), generating an initial "a" leach (buffered acetic acid) and a subsequent "b" leach (buffered hydroxylamine hydrochloride). These leach solutions were purified for Nd and Sr through column chemistry and run on a Nu instruments Multi Collector Inductively Coupled Plasma Mass Spectrometer at Keck Collaboratory at Oregon State University (error on all Nd data is 0.3 units; see Supporting Information for details).

[7] Because of the overall oceanographic consistency of the "a" leach data with respect to known seawater Nd and Sr isotopic values, we consider these data to best represent a pure authigenic record of Nd isotopic signature of bottom water (expressed as ε_{Nd}; see Supporting Information). Two samples off the Siberian margin have deviant ε_{Nd} values (Figure 1b, green color data, and Table 1). The similar location and values indicated that these data may represent a true paleoceanographic signal, possibly due to exposure of pre-Holocene sediments.

3. Discussion

3.1. Shelf Waters

[8] Around the broad Eurasian shelves, local water sources clearly play a predominant role in setting the hydrogenous ε_{Nd} (Figure 1). As such, shelf signals will reflect the higher variability due to temporal changes in circulation and Nd input from local sources, which may well include preformed signals from riverine-based metal oxide sediment coatings [*Bayon et al.*, 2004]. Nevertheless, the noteworthy consistency in Nd isotope distribution (Figure 1) implies that the riverine inputs and shelf currents are in a statistical steady state configuration over longer time scales (i.e., tens to hundreds of years: equivalent to sedimentation rates on the shelves). Radiogenic (higher) ε_{Nd} values (-5.0; -3.4) occur off the Ob' and Yenisey estuaries (Figure 1a), reflecting the influence of the Putorana Flood Basalts [*Porcelli et al.*, 2009]. Where rivers drain more "typical" continental bedrock, the ε_{Nd} values of the shelf waters are much less radiogenic (e.g., $\varepsilon_{Nd} = -8$ to -9), i.e., off the Lena ("L" in Figure 1a) delta and further east on the adjacent Siberian shelf. These values are still considerably higher than those measured in Lena River waters (around $\varepsilon_{Nd} = -14$) [*Porcelli et al.*, 2009], which is probably due to the predominant coastal currents carrying waters from the Kara Sea and western part of the Laptev Sea eastward [*Weingartner et al.*, 1999; *Dmitrenko et al.*, 2005]. Further east, off the Kolyma River (Figure 1a), ε_{Nd} gets as radiogenic

as −1.3, apparently reflecting basaltic exposures of the Okhotsk-Chukotsk volcanic belt or of the High Arctic Large Igneous Province in the easternmost Siberia [e.g., *Ledneva et al*., 2011]. This result is consistent with mineralogical provenance data from the same area [*Viscosi-Shirley et al*., 2003a, 2003b].

[9] Most distinct in this survey is the radiogenic signature of sediments on the Chukchi Sea shelf and adjacent continental slope and borderland. Measurements in the Bering Strait show that Pacific throughflow water has ε_{Nd} values of −4 to −6 [*Dahlqvist et al*., 2007; *Porcelli et al*., 2009], which is lower than the North Pacific oceanic water signature (~−2) [*Piepgras and Jacobsen*, 1988], possibly due to the influence of the Yukon and Anadyr river runoff and/or "boundary exchange" with Bering Strait sediments [*Porcelli et al*., 2009; *Lacan and Jeandel*, 2005]. On the other hand, our core-top data from the southern part of the Chukchi shelf show more radiogenic values of $\varepsilon_{Nd} = \sim -2$, similar to values for the North Pacific (Figure 1). The inner Chukchi shelf is, however, strongly affected by current and/or storm winnowing [*Darby et al*., 2009], which may mean that our core-top sediments from this area represent an earlier (middle to late Holocene) environment. This interpretation would imply that Pacific flow into the Arctic in the past was stronger than it is more recently, or was less influenced by riverine waters. A stronger Pacific throughflow in the middle Holocene (ca. 3.5–5.5 ka) has been inferred from sediment provenance in a core from the Chukchi slope [*Ortiz et al*., 2009] and further supported by oceanographic modeling [*Ortiz et al*., 2012]. This consistency suggests that hydrogenous ε_{Nd} can be used for tracking changes in Pacific inflow into the Arctic. In future studies, this proxy may elucidate patterns and fluxes of circulation change in the past that can then be compared to changes in Arctic climate [*Woodgate et al*., 2010; *Shimada et al*., 2006; *Morison et al*., 2012]. Overall, it is clear that sediments on the Eurasian shelves have distinct ε_{Nd} values compared to the deeper waters of the Eurasian or Canadian Basin (Figure 1a). The exceptions near Svalbard and at the outer Chukchi margin are shelf regions that seem to have a connection with deeper water, as discussed below.

3.2. Deeper Water

[10] The pattern of ε_{Nd} distribution from our core top survey (Figure 1) is consistent with the notion that deep water in most of the Arctic Ocean basins is primarily sourced from the North Atlantic inflow with $\varepsilon_{Nd} \sim -10.8$ [*Andersson et al*., 2008; *Porcelli et al*., 2009]. Closer to the northern margin of the Chukchi shelf, where environments are favorable for sediment accumulation [*Darby et al*., 2009], our core-top data show ε_{Nd} values of −4 to −5 (Figure 1), similar to modern water column measurements in the Bering Strait. Interestingly, similarly radiogenic values $\varepsilon_{Nd} = \sim -5$) characterize even deep (to 1200 m) sites on the continental slope off the Chukchi shelf, with gradually decreasing ε_{Nd} values (−9.9) northward along the Northwind Ridge and to the Mendeleev Ridge (Figure 1). This pattern is consistent with measurements of elevated ε_{Nd} values (−9) along with elevated Nd concentrations in bottom waters of the Canada Basin near the Chukchi margin [*Porcelli et al*., 2009]. As yet, there is no observational evidence for downwelling of winter Chukchi waters to penetrate below the halocline at ~200 m depth [*Woodgate et al*., 2005; *Pickart et al*., 2005]. Despite this lack of observational evidence, the input of salt from a shelf brine source seems a possible explanation for the high salinities measured in the deep Canada Basin [*Schlosser et al*., 1995; *Ivanov et al.,* 2004]. Similarly, *Porcelli et al*. [2009] observe that dissolved Nd concentrations are also highest in the Canada Basin deep water, higher than Atlantic-sourced waters, implying the need for an additional source of Nd. These authors particularly note that the highest deep water Nd concentrations are found "near the slope in the Canada Basin," consistent with the idea of a channelized brine flow from the Chukchi shelf.

[11] Our data may represent a time-averaged record of subducting brines that overcomes the potential observational shortcomings of missing episodic events. Moreover, considering the arguments for a contemporary changing Arctic oceanography, it is possible that deep brine subduction was more common in the past and our records reflect this older condition. An observation that may support this idea is that presently the Canada Basin has an increasing freshwater and heat buildup [*Jackson et al*., 2011; *Morison et al*., 2012*; Timmermans et al.,* 2011], which would serve to increasingly suppress brine formation. A second observation in support of brine subduction is that deep Canada Basin water is known to be ~450 years old [*Schlosser et al*., 1997], providing the potential for increasing its salinity through brine input over time. Such an explanation is consistent with the predictions of *Porcelli et al*. [2009], who suggest that an Nd source with $\varepsilon_{Nd} = -3.8$ is the most straightforward explanation of their observations from deep Canada Basin.

[12] Alternatively, these radiogenic deepwater ε_{Nd} values may represent redistributed sediment from the shelf [*Gutjahr et al*., 2008]. In this way, the deepwater radiogenic signals would represent a mixture of locally formed and preformed (i.e., shelf-derived) sediment coatings. Storm events can cause cascading of suspended sediment to large depths via slope canyons [e.g., *Puig et al*., 2003]. Although not yet observed directly on the canyonized Chukchi margin, this mechanism has been inferred for sediment accumulation on the mid-to-lower slope (~400–1300 m) [*Darby et al*., 2009]. We note that while such density flows could carry brine water with them, they would not necessarily help explain the high salinity found in the deep Canada Basin water.

4. Conclusion

[13] The surveyed distribution of ε_{Nd} values in surface sediment leachates across the Arctic Ocean, representing the averaged late Holocene to pre-modern conditions, shows an overall similar pattern with the isotopic signature carried by bottom water and thus reflects deepwater circulation in the Arctic. Local deviations are explained by exposure of older Holocene sediments. Our data confirm that most of the deep Arctic Ocean water is sourced from Atlantic waters. Pacific water input over the Chukchi Shelf is also clearly delimited by elevated (radiogenic) ε_{Nd} isotopic signals. A distinct radiogenic ε_{Nd} signal is also observed on the slopes of the Chukchi margin and adjacent borderland, indicative of Pacific water subduction (e.g., via brine rejection) and/or persistent sediment redistribution from the shelf. Such a source of local ventilation would represent a significant difference in the subsurface circulation compared to the modern hydrographic observations and is, therefore, important for understanding of past changes in Arctic oceanography

with respect to climate change. This work provides a basis for comparing down-core ε_{Nd} data from this region to elucidate the history of water-mass exchange, sea ice conditions, and shelf-to-basin sediment transport, which has important implications for the dramatic ongoing change in the western Arctic [e.g., *Morison et al*., 2012; *Proshutinsky et al*., 2002; *Shimada et al*., 2006; *Woodgate et al*., 2010].

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