



Diagenetic overprint on authigenic Nd isotope records: A case study of the Bering Slope

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ABSTRACT

Neodymium isotope ratios ($^{143}\text{Nd}/^{144}\text{Nd}$, ϵ_{Nd}) have been used as a quasi-conservative water mass tracer in the Atlantic and Southern oceans. However, boundary exchange and diagenesis can limit their use in areas where extensive interactions between seawater, pore water and bottom sediments can overwrite the original water mass signature. To avoid the misapplication of this proxy and to provide a new perspective on the application of Nd isotopes to authigenic phases, we present ϵ_{Nd} records covering the past 2.4 Myr for the authigenic ($\epsilon_{\text{Nd,auth}}$) and detrital ($\epsilon_{\text{Nd,det}}$) phases of sediments from Integrated Ocean Drilling Program site U1343 (~1950 m water depth) on the Bering Slope. The $\epsilon_{\text{Nd,det}}$ values range from -9.0 to -2.3 ($n = 70$), showing binary mixing between Alaskan and Aleutian sediments. Glacial–interglacial variations are muted in the $\epsilon_{\text{Nd,det}}$ record because sediment delivery pathways from both source areas exist regardless of climatic conditions. The $\epsilon_{\text{Nd,auth}}$ values range from -5.9 to -0.2 ($n = 153$) and show a strong correlation ($r = 0.58$, $n = 60$) with $\epsilon_{\text{Nd,det}}$ in the deeper part of the core (>160 ka), where authigenic carbonate layers are frequently observed. We attribute this covariation to diagenetic overprinting by pore water Nd during late diagenesis. The pore water Nd that is incorporated into authigenic carbonates was initially released by marine silicate weathering, therefore indicating that sediment lithology exerts a first-order control on the ϵ_{Nd} values of the pore water in deeply buried sediments. In the shallower part of the core (<160 ka), there is no correlation between $\epsilon_{\text{Nd,auth}}$ and $\epsilon_{\text{Nd,det}}$ ($r = -0.23$, $n = 10$). Here, $\epsilon_{\text{Nd,auth}}$ is typically radiogenic during interglacial periods, which may be due to the diagenetic flux from the relatively stagnant N. Pacific or the incongruent chemical weathering of freshly ground sediments on the Bering Shelf. The effect of dense water formation on $\epsilon_{\text{Nd,auth}}$ seems to be minimal when we compare the $\epsilon_{\text{Nd,auth}}$ records at three different depths of the Bering Sea. Our ϵ_{Nd} dataset provides a unique perspective on the behavior of $\epsilon_{\text{Nd,auth}}$ during late diagenesis. To apply $\epsilon_{\text{Nd,auth}}$ as a water mass tracer to long cores containing volcanogenic sediments or in productive regions, we recommend testing for deep authigenesis using pore water profiles.

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1. Introduction

Neodymium isotopes ($^{143}\text{Nd}/^{144}\text{Nd}$) have been widely used as a proxy for past ocean circulation (e.g., Horikawa et al., 2010; Jang et al., 2013; Rutberg et al., 2000) and are usually expressed in epsilon notation ($\epsilon_{\text{Nd}} = [\frac{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{sample}}}{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}} - 1] \times 10^4$, where $(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}$ is 0.512638 (Jacobsen and Wasserburg, 1980). The most recent reference value for $(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}$ is 0.512630 (Bouvier et al., 2008)). The Pacific Ocean, which is surrounded by

young volcanic belts, is more radiogenic (higher ϵ_{Nd}) than the North Atlantic, which is predominantly surrounded by old continental shields (Piepgras and Wasserburg, 1980). The residence time of dissolved Nd is shorter than the ocean mixing time (Tachikawa et al., 1999), and seawater ϵ_{Nd} values are basin-specific; therein lies the utility of ϵ_{Nd} as a water mass tracer. Past bottom water ϵ_{Nd} values can be retrieved by extracting the reducible authigenic fraction from foraminifera or bulk sediments (e.g., Roberts et al., 2010). The potential of the authigenic ϵ_{Nd} ($\epsilon_{\text{Nd,auth}}$) proxy as a water mass tracer was realized for the South Atlantic, where two water masses with contrasting ϵ_{Nd} values meet, i.e., the unradiogenic North Atlantic Deep Water (NADW; $\epsilon_{\text{Nd}} \sim -12.4$ to -13.2 from Lambelet et al., 2016) and the radiogenic Circumpolar Deep Water ($\epsilon_{\text{Nd}} \sim -8.2$ to -8.4 from Garcia-Solsona et al., 2014). The $\epsilon_{\text{Nd,auth}}$ records in the S. Atlantic have shown clear

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glacial–interglacial (G–IG) variability at least since 1 Ma, which is interpreted to represent the glacial reduction of NADW formation (Dausmann et al., 2017; Piotrowski et al., 2005; Rutberg et al., 2000).

The $\epsilon_{\text{Nd,auth}}$ proxy has been critically assessed in the recent past. One reason for this is that past ocean circulation signals can be modified by boundary exchange and diagenetic flux (Abbott et al., 2016; Jeandel, 2016; Tachikawa et al., 1999). Boundary exchange is an umbrella term that refers to sediment–seawater interaction at the land–ocean interface, including continental margins (reviewed in Jeandel, 2016). Diagenetic flux refers specifically to the Nd efflux from pore water into seawater resulting from early diagenetic reactions between sediment and pore water and is not limited to these margins (Abbott et al., 2016; Du et al., 2016). Both boundary exchange and diagenetic flux occur near the sediment–bottom water interface. In the Atlantic, vigorous bottom water renewal by NADW formation seems to minimize their effect, and the variation in deep-water ϵ_{Nd} can be attributed to large-scale water mass mixing (e.g., Piotrowski et al., 2005; Rutberg et al., 2000). In the North Pacific, however, there is no deep-water formation at present, and sediments contain a considerable amount of labile volcanic material. Thus, boundary exchange and diagenetic flux can exert a stronger influence on the deep-water Nd budget in the N. Pacific (Abbott et al., 2016; Jang et al., 2017). This is demonstrated by the progressive radiogenic shift in modern deep-water ϵ_{Nd} values from the Southern Ocean to the N. Pacific (Abbott et al., 2016).

Another caveat for the $\epsilon_{\text{Nd,auth}}$ proxy concerns diagenetic overprinting by pore water during continued sediment–pore water interaction after early diagenesis (Abbott et al., 2016; Du et al., 2016). The pore water profiles of dissolved inorganic carbon (DIC), Mg and Ca indicate that in the zone of methanogenesis, the weathering of silicate minerals occurs in addition to the precipitation of authigenic carbonate minerals. Authigenic carbonate minerals have actually been observed at these depths (Pierre et al., 2016; Wehrmann et al., 2011). Marine silicate weathering can release Nd from bulk sediments into pore waters and re-incorporate it into authigenic phases and, in the process, modify the $\epsilon_{\text{Nd,auth}}$ record. There remains some uncertainty as to how large an effect late diagenesis will exert on the $\epsilon_{\text{Nd,auth}}$ record. For example, Du et al. (2016) suggested that $\epsilon_{\text{Nd,auth}}$ would be insensitive to later diagenetic alteration because of the much higher Nd contents of authigenic phases relative to pore water.

Site U1343 (57°3'N, 175°5'W; 1950 m water depth; 744 m below the sea floor (mbsf)), which is located on the Bering Slope, has one of the longest sediment cores recovered by the Integrated Ocean Drilling Program (IODP). The detrital ϵ_{Nd} ($\epsilon_{\text{Nd,det}}$) record of this core enables us to study how sediment provenance varied with G–IG climate change throughout the Pleistocene, ~2.4 Myr. In terms of the authigenic ϵ_{Nd} record, this site, which is located in the marginal sea of the North Pacific and exhibits a significant labile volcanic component, is an attractive testing ground for investigating how sediment–water interaction may modify the original water mass signal. Additionally, methanogenesis and associated deep authigenesis have been documented at this site (Pierre et al., 2016; Expedition 323 Scientists, 2011), thus offering a rare opportunity to examine how late diagenesis affects the paleoceanographic interpretation of $\epsilon_{\text{Nd,auth}}$.

2. Materials

2.1. Site information

Sediment core samples were retrieved from site U1343 on the continental margin of the Bering Sea during IODP Expedition 323 (Fig. 1). Site U1343 is located on a topographic high of the Bering

Slope and is separated from the Bering Shelf; it is relatively unaffected by the re-transportation of shelf sediments during interglacial periods or by aeolian dust from the exposed shelf during glacial periods (Expedition 323 Scientists, 2011). There are five giant submarine canyons in the Bering Sea, and site U1343 is located on the interfluvial of the Zhemchug Canyon, which is the world's largest canyon in terms of depth (~2550 m) and volume (5800 km³) (Expedition 323 Scientists, 2011) (Fig. 1).

Biological productivity is high on the Bering Slope; it is sustained by a continuous supply of nutrients to the euphotic zone by the Bering Slope Current (BSC) and tidal mixing (Springer et al., 1996). High organic carbon export results in low dissolved oxygen concentrations (~65.9 $\mu\text{mol/L}$) at site U1343, which is below the modern oxygen minimum zone (Expedition 323 Scientists, 2011). The site is close to the maximum extent of present-day sea ice cover (Stabeno et al., 2012) (Fig. 1). During glacial periods, seasonal sea ice coverage probably extended to the central Aleutian Basin (Katsuki and Takahashi, 2005 and references therein). With the drop in sea level, the shallow continental shelf was aerially exposed (Fig. 1) (Katsuki and Takahashi, 2005), and the closing of the Bering Strait changed the water circulation pattern in the Bering Sea by preventing northward outflow to the Arctic (Hu et al., 2012).

Considering the present anti-clockwise water circulation in the Bering Sea, the ultimate Nd sources to site U1343 are the Aleutian Arc and Alaska (Jang et al., 2017) (Fig. 1). The Aleutian Arc supplies radiogenic Nd ($\epsilon_{\text{Nd}} \sim +6$ to $+10$) (data from GEOROC, 2003), while Alaska supplies unradiogenic Nd ($\epsilon_{\text{Nd}} \sim -10.1$ to -8.4) (data from VanLaningham et al., 2009). When the Bering Strait was closed during glacial sea level lowstands (Hu et al., 2012; Jang et al., 2017), the Anadyr River in Siberia may also have supplied radiogenic ϵ_{Nd} ($\epsilon_{\text{Nd}} \sim -2$ to $+3$) (data from GEOROC, 2003) to site U1343.

2.2. Core description

We mainly used Hole U1343E (744 mbsf) to reconstruct the dissolved and particulate Nd isotopic compositions of the Bering Slope. The sediment in this area alternates between siliciclastic and mixed siliciclastic–biogenic sediment (Expedition 323 Scientists, 2011). The siliciclastic sediments (<40% biogenic components) are mainly composed of diatom-rich silt and clay, and the mixed siliciclastic–biogenic sediments (>40% biogenic components) further include diatom ooze and minor foraminifera, calcareous nannofossils and sponge spicules. The carbonate and TOC contents are typically less than 4% and 1%, respectively. Authigenic carbonate-rich layers and nodules appear deeper than ~37 mbsf at irregular intervals, and their occurrences significantly increase at 250–290 mbsf and below 570 mbsf (Pierre et al., 2016; Expedition 323 Scientists, 2011) (Fig. 2). Authigenic carbonates are also present as small crystals (generally <10 μm) within the sediment. Shipboard visual description reports the presence of several thin ash layers with ash-filled mottles (Expedition 323 Scientists, 2011).

The age of the core extends to ~2.4 Ma, according to the age model of Asahi et al. (2016), which is based on the correlation of benthic foraminiferal $\delta^{18}\text{O}$ ($\delta^{18}\text{O}_{\text{bf}}$) values between site U1343 and the global LR04 stack. Ninety-three boundaries of marine isotope stages (MIS) and 344 tie points were assigned. The average sedimentation rate was ~32.8 cm kyr⁻¹, and it exhibits temporal fluctuations (11.6–72.0 cm kyr⁻¹) that do not correspond to G–IG cycles.

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