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## The evolution of climatically driven weathering inputs into the western Arctic Ocean since the late Miocene: Radiogenic isotope evidence

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#### ABSTRACT

We present the first continuous records of dissolved radiogenic neodymium, hafnium, and lead isotope compositions of deep waters in the western Arctic Ocean, spanning the time from the late Miocene to the present. The data were obtained from three hydrogenetic ferromanganese (Fe-Mn) crusts recovered from seamounts along the northernmost edge of the Northwind Ridge in the Canada Basin from water depths of 2200, 2400, and 3600 m. Dating the crusts using cosmogenic <sup>10</sup>Be documents undisturbed present-day growth surfaces and yields growth rates between 27 and 2.2 mm/Myr. The Nd (Hf) isotope time series of the three crusts show similar evolutions from  $\varepsilon_{Nd}$  ( $\varepsilon_{Hf}$ ) of -8.5 (+4) in the oldest parts to -11.5 (-4) at the surfaces and a pronounced trend to less radiogenic values starting at  ${\sim}4$  Ma. This coincided with a trend of the Pb isotope evolution towards more radiogenic <sup>206</sup>Pb/<sup>204</sup>Pb, <sup>207</sup>Pb/<sup>204</sup>Pb, and <sup>208</sup>Pb/<sup>204</sup>Pb. It is inferred that climatically controlled changes in weathering regime and sediment transport along the North American continent were responsible for the major change of the radiogenic isotope composition of the Arctic Deep Water (ADW) in the Canada Basin. Based on these records we conclude that weathering inputs from the North American continent linked to enhanced glacial conditions started to increase and to influence the radiogenic isotope composition of ADW  $\sim$ 4 million years ago and were further intensified at  $\sim$ 1 Ma. These new time series differ markedly from the radiogenic isotope evolution of Arctic Intermediate Water recorded on the Lomonosov Ridge and suggest that much larger isotopic differences between the water masses of the Arctic Ocean than today prevailed in the past.

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#### 1. Introduction

Unraveling the oceanographic history of the Arctic Ocean and the evolution of weathering inputs from the surrounding continents, which have responded sensitively to local and global environmental changes (e.g. Moran et al., 2006), is of major importance for understanding the mechanisms driving global climatic changes. While there is evidence for extended periods of time with a perennial sea ice cover of the Arctic Ocean since 44 Ma and continuous perennial sea ice since about 36 Ma (Darby, 2014) the onset of Northern Hemisphere Glaciation (NHG) and its intensification in the late Pliocene between 2.4 and 2.9 Ma (e.g., Shackleton et al., 1984; Raymo, 1994; Zachos et al., 2001;

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Flesche Kleiven et al., 2002) was accompanied by the build-up of major ice sheets on the continents surrounding the Arctic Ocean (Polyak et al., 2001; Moran et al., 2006). As a consequence of enhanced glacial conditions a transition of the weathering regimes on land towards increased mechanical denudation occurred (e.g. Riebe et al., 2004).

Previous studies have invoked regional tectonic forcing (Matthiessen et al., 2009; Knies et al., 2014) and changes in ocean circulation in the North Atlantic linked to the closure of the Isthmus of Panama (Driscoll and Haug, 1998) as well as moisture export from the subarctic Pacific (Haug et al., 2005) as possible triggers for the onset of Plio-Pleistocene circum-Arctic glaciation. However, the exact temporal and spatial evolution of this pronounced climatic change is not well constrained from the marine sedimentary record in the Arctic Ocean as a consequence of the paucity of long sedimentary records. The only available record covering the past 55 million years was recovered dur-

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Fig. 1. Schematic transect across the Arctic Ocean including major water masses (modified from AMAP, 1998). Approximate positions of the samples on the Northwind Ridge are indicated. Residence times of water masses have been adopted from Bönisch and Schlosser (1995).

ing IODP Leg 302, the Arctic Coring Expedition (ACEX), from an intermediate water depth of 1200 m on the Lomonosov Ridge and is interrupted by a short and a long hiatus between 9.4 and 11.6 Ma and 18.2 and 44.4 Ma, respectively (Moran et al., 2006; Backman et al., 2006, 2008). Several studies have analyzed climatically driven changes in the provenance of the sediments supplied to the core location. Haley et al. (2008b) for example applied radiogenic isotopes to demonstrate that detrital inputs in the central Arctic Ocean have been linked to climatic forcing of weathering on the Eurasian continental margin, whereas März et al. (2010) defined geochemical units (based on major and trace elements) representing changes in detrital provenance, as well as in paleoenvironmental and diagenetic processes. Other than that only Pliocene Arctic sediments have been accessible from some very low sedimentation rate locations such as in the vicinity of the Alpha Ridge (e.g. Winter et al., 1997), for which the age models are, however, not well constrained due to the absence of carbonate microfossils. Recent evidence suggested that the true maximum ages of these records are much younger than initially published (e.g. Jakobsson et al., 2000) but there is still debate on the age models of sediments in the western Arctic Ocean (e.g. Sellén et al., 2009).

Here we present the first combined seawater-derived radiogenic isotope records of Nd, Hf, and Pb obtained from three hydrogenetic Fe–Mn crusts from the Northwind Ridge in the Canada Basin, which are condensed chemical sediments and reflect the continuous evolution of weathering regimes and erosional inputs for the period from the late Miocene (7 Ma) to the present.

## 1.1. Tracing weathering inputs and water-mass mixing with radiogenic isotopes

The main source for dissolved radiogenic Nd, Hf and Pb in the ocean is weathering of continental rocks and island arcs, the signatures of which are transferred to the ocean via dissolved and particulate phases of rivers (cf. Frank, 2002). Furthermore, hydrothermal inputs can be significant sources of Pb and Hf close to vent sites (van de Flierdt et al., 2004). Partial dissolution of and exchange with shelf sediments has recently been identified as another input source for dissolved oceanic Nd, which may even be more important than riverine inputs (Lacan and Jeandel, 2005b; Arsouze et al., 2009; Rempfer et al., 2012). Having average residence times in seawater on the order of 400–2000 years and thus shorter than or similar to the global ocean mixing time,

Nd and Hf isotopes are not distributed homogeneously and can in the case of Nd be applied as quasi-conservative tracers of present and past water masses and their mixing, at least on basinwide scales (cf. Piepgras and Wasserburg, 1987; Jeandel, 1993; Jeandel et al., 1995). Pb isotopes are suitable for the reconstruction of local inputs from land and their short distance mixing due to the shorter average residence time of Pb (50–200 years) in seawater (cf. von Blanckenburg et al., 1996).

In contrast to Nd, Hf weathers incongruently which results in more radiogenic Hf isotope compositions of rivers and seawater than expected from the corresponding Nd isotope signatures forming the so-called seawater array (Albarède et al., 1998; Chen et al., 2013; David et al., 2001; Zimmermann et al., 2009a, 2009b; Rickli et al., 2009). This observation is mainly a consequence of the "zircon effect", which is a consequence of the fact that most of the unradiogenic Hf in a rock is bound in highly weathering resistant zircons (White et al., 1986). The zircon effect is most likely not the sole reason for these signatures and differential weathering of the highly radiogenic zircon-free Hf fraction also plays a significant role in controlling seawater Hf isotope compositions (Chen et al., 2011; Bayon et al., 2009; Rickli et al., 2012).

The reconstruction of past deep-ocean circulation and weathering inputs using radiogenic isotopes on millennial time scales has been achieved by analyzing various phases of marine sediments, such as fish teeth (e.g. Martin and Haley, 2000) or Fe-Mn coatings of particles or foraminifera (e.g. Rutberg et al., 2000; Roberts et al., 2010). Hydrogenetic Fe-Mn crusts, which are chemical sediments that accrete at very low growth rates of generally 1–10 mm/Myr directly from seawater in areas of reduced or absent clastic sedimentation, enabled the investigation of the isotopic evolution on million year time scales in all major ocean basins (e.g. Ling et al., 1997; Burton et al., 1997, 1999; O'Nions et al., 1998; Reynolds et al., 1999; Frank et al., 2002). Due to the lack of long sedimentary records our new Arctic Ocean crust records offer important new insights.

#### 1.2. Hydrography of the Arctic Ocean

The Arctic Ocean is a strongly stratified and semi-enclosed basin (Fig. 1) with approximately 35% of its extent underlain by shallow continental shelves. The uppermost 50 m of the Arctic water column, termed the Polar Mixed Layer (PML), is characterized by temperatures as low as -1.7 °C as a function of highly variable freshwater supplies originating from seasonally chang-

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