Pathways of Siberian Freshwater and Sea Ice in the Arctic Ocean Traced with Radiogenic Neodymium Isotopes and Rare Earth Elements

Georgi Laukert[1*], Martin Frank[1], Ed C. Hathorne[1], Thomas Krumpen[1], Ben Rabe[2], Dorothea Bauch[1], Kirstin Werner[2], Ilka Peeken[2] and Heidemarie Kassens[1]

Abstract: Freshwater and sea ice in the Arctic Ocean (AO) both respond to climate forcing but changes in their distribution may in turn also significantly affect Northern Hemisphere climate. The exact mechanisms controlling their transport are, however, still subject of ongoing research. Radiogenic neodymium (Nd) isotopes (expressed as ε_{Nd}) and rare earth elements (REEs) have successfully been used to trace ocean circulation in the AO and can provide valuable information on the origin, transport and fate of Arctic freshwater and drifting sea ice. Here, we present first sea-ice data and discuss the applicability of these tracers to determine the pathways of Siberian freshwater and the source areas of Arctic sea ice within the Eurasian Basin. Freshwaters originating from various Siberian rivers are well mixed and diluted prior to their advection to the central AO and the Fram Strait, thereby inhibiting the assessment of the individual contributions of the different rivers for these regions. Recent investigations of seawater Nd isotope and REE distributions on the Siberian shelves now allow for a more detailed determination of the sources of transpolar freshwater transport, and suggest that freshwater from the Lena River can at least be qualitatively traced further away from the shelves. In addition, sea ice has the potential to preserve marine and riverine ε_{Nd} and REE signatures of the Siberian shelf source waters transported across the AO to the Fram Strait thereby avoiding the effects of mixing. Here, we test this potential for the first time by comparing ε_{Nd} and REE signatures determined from unfiltered but essentially sediment-free sea-ice samples recovered in the Eurasian Basin of the central AO in 2012 with corresponding signatures of surface seawater in the formation regions of the ice. The sampled sea ice is dominantly characterized by ε_{Nd} values near -8, in agreement with the incorporation of radiogenic freshwater (ε_{Nd} = -6) from the Yenisei and Ob rivers and its formation east of Vilkitsky Strait as identified by a backtracking approach based on satellite-derived ice drift and concentration data. The latter also indicates that sea ice with a less radiogenic ε_{Nd} signature near -11 formed north of the Laptev Sea, in agreement with mixed contributions of radiogenic waters from the Kara Sea and northward flowing highly unradiogenic Lena River water ($\varepsilon_{Nd} \leq$ -15). Compared to the REE concentrations in Siberian shelf waters, those in the sea ice are depleted, indicating rejection of REEs together with salt during sea-ice formation. Despite potential modifications during incorporation into sea ice, its REE distribution patterns indicate a predictable relationship to those of the source waters, thus in addition supporting preservation of at least some of the marine and riverine REE characteristics during sea-ice formation and transport.

Zusammenfassung: Süßwasser und Meereis im Nordpolarmeer reagieren beide einerseits auf klimatische Veränderungen, ihre Verteilung kann aber umgekehrt auch das Klima der Nordhemisphäre beeinflussen. Die genauen Mechanismen, die ihren Transport steuern, sind jedoch immer noch nicht ausreichend verstanden und umfassen daher ein zentrales Forschungsthema. Radiogene Neodymisotope (ε_{Nd}) und Seltenerdelemente (REE) wurden

bereits erfolgreich angewendet, um die Ozeanzirkulation in der arktischen Region nachzuverfolgen, und haben das Potential, wertvolle Informationen über die Herkunft, den Transport und den Verbleib des Süßwassers und des treibenden Meereises zu liefern. Hier präsentieren wir neue Meereis-Daten und diskutieren die Anwendbarkeit dieser Tracer für die Bestimmung der Transportwege sibirischen Süßwassers und der Herkunft arktischen Meereises innerhalb des Eurasischen Beckens. Das Süßwasser aus verschiedenen sibirischen Flüssen wird vor seiner Advektion in das zentrale Nordpolarmeer und die Framstraße verdünnt und mit anderen Wassermassen gemischt, so dass eine exakte Bestimmung des Eintrags einzelner Flüsse für diese Regionen nicht mehr möglich ist. Neue Ergebnisse zur Verteilung der im Meerwasser gelösten ε_{Nd}- und REE-Signaturen auf den sibirischen Schelfen erlauben nun eine genauere Bestimmung der Quellen des transpolaren Süßwassertransports und legen nahe dass abseits der Schelfregionen Flusswasser der Lena zumindest qualitativ nachverfolgt werden kann. Zusätzlich kann das Meereis die marinen und durch Flüsse eingetragenen ε_{Nd}- und REE-Signaturen der sibirischen Schelfwässer ohne Mischung über das zentrale Nordpolarmeer transportieren. Dieses Potential wird hier zum ersten Mal durch einen Vergleich von ε_{Nd}- und REE-Signaturen auf ungefiltertem, aber im Wesentlichen sedimentfreiem Meereis, das 2012 im eurasischen Teil des Nordpolarmeeres beprobt wurde, mit den marinen Signaturen im Entstehungsgebiet des Meereises getestet. Das Meereis hat hauptsächlich ε_{Nd} Werte um -8, in Übereinstimmung mit dem Einschluss der radiogenen Süßwassersignaturen (ε_{Nd} = -6) von Jenissei und Ob und der Entstehung dieses Meereises östlich der Wilkitzkistraße. Dieser Entstehungsort wurde anhand eines Backtracking-Ansatzes basierend auf durch Satellitendaten gestützte Eisdrift- und Konzentrationsdaten ermittelt. Meereis, das anhand dieses Backtracking-Ansatzes nördlich der Laptewsee gebildet wurde, hat im Gegensatz dazu eine ε_{Nd}-Signatur nahe -11 konsistent mit Zumischungen der radiogenen Wässer aus der Karasee und des nordwärts fließenden deutlich weniger radiogenen Süßwassers der Lena ($\varepsilon_{Nd} \leq$ -15). Die REE-Konzentrationen im Meereis sind insgesamt niedriger als die der Schelfwässer, was auf die Ausscheidung der REEs zusammen mit dem Salz während der Eisbildung hindeutet. Trotz Anzeichen für Modifikationen beim Einbau in das Meereis zeigen die REE-Verteilungsmuster des Meereises ein systematische Beziehung zu den Wässern im Entstehungsgebiet des Meereises an, was zusätzlich die überwiegende Erhaltung der marinen und durch Flüsse eingetragenen REE-Charakteristika andeutet.

INTRODUCTION

The surface layer of the Arctic Ocean (AO) is unique among the world's oceans due to its vast sea-ice cover and low salinity originating from riverine inputs. The AO not only receives about 11 % of the global riverine discharge (Fichot et al. 2013), but also accommodates significant amounts of Pacific-derived freshwater (e.g., WOODGATE et al. 2006, 2012), and freshwater discharged from the Greenland Ice Sheet (BAMBER et al. 2012). It is covered by the second largest amount of sea ice on Earth and strongly linked with the Earth's climate system. Due to global climate change, the content, distribution and circulation of Arctic freshwater and sea ice have changed recently, which has already had consequences for the Arctic marine ecosystems, the oceanic conditions of the AO and potentially for the global climate. In particular the transport of Arctic freshwater and sea ice to temperate latitudes

has increased (DE STEUR et al. 2017 and SMEDSRUD et al. 2017 and references therein), leading to a freshening of the North Atlantic. The additional stratification has the potential to influence deep convection and to weaken the Atlantic Meridional Overturning Circulation (cf., RAHMSTORF et al. 2015, YANG et al. 2016). Realistic predictions of future climatic changes thus need to include reliable assessments of the origin and pathways of Arctic freshwater and sea ice.

The mechanisms controlling the transport of freshwater and sea ice in the AO are still subject of ongoing research. A common conceptual model is that the distribution of Arctic freshwater and sea ice is related to oscillations in the large-scale atmospheric circulation inducing variability in the sea-ice and ocean surface circulation (cf., MAURITZEN 2012). This is most evident for the sea ice, given that its movement is clearly forced by the wind-driven anticyclonic Beaufort Gyre and the Transpolar Drift (TD). Observational studies have shown that variable amounts of liquid Siberian freshwater are in addition transported with the TD to the central AO and further to the western Fram Strait (e.g., TAYLOR et al. 2003, ANDERSON et al. 2004, KARCHER et al. 2006, ALKIRE et al. 2007, JONES et al. 2008, RABE et al. 2009, BAUCH et al. 2011, RABE et al. 2013). However, subsurface oceanic currents and regional hydrographic processes have also been suggested to affect the distribution and transport of Arctic freshwater (cf., RABE et al. 2013). For example, studies of several Arctic regions in the late 1990s showed that subsurface currents rather than atmospheric forcing mainly govern the transport of shallow waters (e.g., RUDELS et al. 1996, JONES et al. 1998, JONES 2001). In particular, for large parts of the Eurasian Basin these studies found that transport within the upper water column is controlled by the advection of Atlantic-derived waters, which enter the AO through the eastern Fram Strait and continue to flow eastward along the Siberian margin (RUDELS et al. 1996, 1999). While the inflowing branch of these waters propagates in the direction opposite to the TD, one of the returning branches (i.e., directed towards the Fram Strait) was recently confirmed to flow alongside the Lomonosov Ridge at depths between 200 and 400 m, and thus parallel to the TD (RECINOS et al. 2016). It remains to be investigated in detail how these and other currents interact with the TD and which role they play in Arctic sea-ice distribution and Siberian freshwater transport.

The reconstruction of sea-ice pathways has improved in recent years due to the availability of satellite-derived ice drift and concentration data allowing for example to determine drift trajectories and source areas of Arctic sea ice transported to the Fram Strait (KRUMPEN et al. 2016). The low resolution of the satellite sensors combined with the applied methodology to derive sea-ice motion, however, do not allow tracking of individual floes, but rather provide drift reconstructions on a larger scale. Reconstructions of the freshwater pathways are even more challenging, given that they require in-situ data from the water column. Most observational studies investigating freshwater transport to date are based on standard hydrographic tracers such as temperature, salinity, alkalinity and nutrients (e.g., ANDERSON et al. 2004, JONES et al. 2008), sometimes combined with stable oxygen isotopes (e.g., BAUCH et al. 2009, 2011). These tracers are appropriate to identify the sources (i.e., meteoric water and sea-ice melt) and quantify the corresponding amount of freshwater transported in the AO,

but they do not provide direct information on the individual contribution of the different Arctic rivers. These limitations emphasize the importance to search for alternative methods, preferably based on chemical tracers contained in both, seawater and sea ice.

Radiogenic neodymium (Nd) isotopes have been widely applied to trace modern and past ocean circulation, as well as continental inputs (reviews by FRANK 2002, GOLDSTEIN & HEMMING 2003, VAN DE FLIERDT et al. 2016). Their suitability to trace ocean circulation and freshwater pathways in the AO and the Nordic Seas was demonstrated by ANDERSSON et al. (2008) and PORCELLI et al. (2009) and was recently supported by LAUKERT et al. (2017a). The seawater (i.e., dissolved) Nd isotope distribution in the open AO reflects water mass advection and mixing, while seawater-particle interactions exert important control only above the shelf regions as often also reflected by variations in Nd concentrations. However, seawater Nd isotope compositions on the vast Siberian shelves appear not to be significantly modified by such interactions (LAUKERT et al. 2017b). This allows the application of Nd isotopes combined with salinity to trace Siberian freshwater inputs and pathways. In addition to the Nd concentrations usually measured and reported along with the Nd isotope compositions, the concentrations of the other rare earth elements (REEs) and the relative distribution patterns of the complete set of REEs provide complementary information on the composition of the source material, the amount of time since the last contact of the waters with weathering inputs, as well as particle adsorption and desorption processes (e.g., GARCIA-SOLSONA et al. 2014, HALEY et al. 2014, MOLINA-KESCHER et al. 2014, HATHORNE et al. 2015, YANG & HALEY 2016, LAUKERT et al. 2017a, b).

Here we review the applicability of Nd isotopes and REEs to trace the pathways of Siberian freshwater within the Eurasian Basin, and investigate whether these tracers can also be used to provide information on the source areas of drifting Arctic sea ice. We present the first ε_{Nd} and REE signatures determined from unfiltered but essentially sediment-free sea ice and compare the results with marine and riverine signatures expected for surface seawater in the formation region of the sea ice. The findings presented here build on seawater Nd isotope and REE data recently obtained for the Laptev Sea (LAUKERT et al. 2017b).

APPLICATION OF NEODYMIUM ISOTOPES AND THEIR SYSTEMATICS IN THE ARCTIC OCEAN

The radiogenic Nd isotope composition is commonly expressed in the ε_{Nd} notation, which denotes the deviation of a measured $^{143}Nd/^{144}Nd$ ratio from the chondritic uniform reservoir (CHUR) ratio of 0.512638 in parts per 10000 (JACOBSEN & WASSERBURG 1980). The global variation of ε_{Nd} signatures in rocks covers a wide range of ε_{Nd} values from -56 to +11 (LACAN et al. 2012, VAN DE FLIERDT et al. 2016). In general, young mantle-derived rocks have more radiogenic ε_{Nd} signatures (i.e., more positive ε_{Nd} values) than old continental rocks. The range and the distribution of ε_{Nd} observed in rocks result from the fractionation between Nd and samarium (Sm) during magmatic processes, and the decay of ^{147}Sm to ^{143}Nd. The distinct signatures of the rocks are introduced through

riverine or aeolian inputs of weathered continental material to the oceans and label the water masses at the land-ocean interface. The quasi-conservative behavior of Nd in the open ocean (FRANK 2002, GOLDSTEIN & HEMMING 2003) and its intermediate average oceanic residence time of several hundred years (TACHIKAWA et al. 2003, ARSOUZE et al. 2009, REMPFER et al. 2011) then allow tracing the advection and mixing of these signatures and hence the water masses they are carried by. On a global scale, the modern seawater ε_{Nd} compositions, in particular in near surface waters, mirror the compositions of the dominating geological formations located in the basins and at their margins. The Pacific is predominantly surrounded by young mantle-derived lithologies (cf., circum-Pacific belt) and consequently exhibits relatively radiogenic seawater ε_{Nd} signatures, whereas the North Atlantic receives Nd from the old continental rocks of North America and Greenland and therefore is characterized by less radiogenic ε_{Nd} values (compilations provided by LACAN et al. 2012 and VAN DE FLIERDT et al. 2016). The water mass signatures can be further modified only through seawater-particle interactions occurring either within the water column (generally summarized under the term "reversible scavenging", e.g., NOZAKI & ALIBO 2003, SIDDALL et al. 2008) or at the land-ocean boundaries (summarized under the term "boundary exchange", LACAN & JEANDEL 2001, 2005).

The seawater Nd isotope distribution in the AO has been attributed to weathering inputs, water mass circulation and mixing, but also to seawater-particle interactions occurring on the shelves and their slopes (ANDERSSON et al. 2008, PORCELLI et al. 2009, LAUKERT et al. 2017a,b). The latter were recently shown to significantly modify the seawater Nd isotope composition on the North-East Greenland Shelf (LAUKERT et al. 2017a). There, remineralization of biogenic particles, remobilization of REEs from coagulated inorganic nanoparticles and colloids and/or REE release from detrital particles clearly occurred in bottom waters and in addition to an increase of the Nd concentration resulted in a shift of the Nd isotope composition towards less radiogenic values. Seawater-particle interactions also occur on the Siberian shelves, but they differ from those observed on the North-East Greenland Shelf. Observations from the Laptev Sea (LAUKERT et al. 2017b) and the Severnaya Dvina estuary (POKROVSKY et al. 2014) indicate that removal of riverine REEs through salt-induced coagulation of nanoparticles and colloids is the dominating process, whereas significant REE release from particulate phases with an associated change in the seawater Nd isotope composition does not occur (LAUKERT et al. 2017b). The lack of significant release of particulate REEs (except Ce) might be linked to generally low amounts of suspended particulate matter transported by the western Siberian rivers (e.g., GORDEEV et al. 2006), and perhaps also be related to the distinct composition of the riverine nanoparticles and colloids. The latter is supported by experiments simulating estuarine mixing indicating that release of REEs bound to nanoparticles and colloids likely only occurs when the riverine end-member is rich in inorganic and poor in organic material (TEPE & BAU 2016, MERSCHEL et al. 2017a), which is not the case for the major Siberian rivers including the Severnaya Dvina, Lena, Yenisei and Ob (DITTMAR & KATTNER 2003 and references therein). Hence, the inputs of dissolved riverine REEs and water mass advection and mixing likely solely account for the Nd isotope distribution observed in the open AO and on the

Siberian shelves suggesting that Nd isotopes can be applied as a water mass tracer in these regions.

All known REE sources of the AO and their Nd isotope compositions are shown in Figure 1. The Atlantic-derived waters dominate the REE budget and are characterized by ε_{Nd} signatures of -13 and -12.6 at the Iceland-Scotland Ridge and the Denmark Strait, respectively (LAUKERT et al. 2017a and references therein). In contrast, Pacific-derived waters entering the AO through the Bering Strait only represent 11 % of the REE inputs and have distinctly more radiogenic ε_{Nd} signatures around -2 to -3 (CHARETTE et al. 2016). These highly radiogenic signatures are altered through seawater-particle interactions close to the inflow resulting in ε_{Nd} signatures near -5.5 for modified Pacific-derived waters emerging from the Chukchi Sea (DAHLQVIST et al. 2007, PORCELLI et al. 2009). The Norwegian Coastal Water is another important marine source and characterized by an ε_{Nd} signature of -14.5 in the southern Barents Sea (PETROVA 2015, LAUKERT et al. 2017a). As shown (Fig. 1), the discharge of freshwater from the various Arctic rivers has a wide range of different ε_{Nd} signatures ranging from -5 to -17 (PORCELLI et al. 2009, ZIMMERMANN et al. 2009, PERSSON et al. 2011, LAUKERT et al. 2017a,b). Additional contributions of REEs via glacial meltwater and runoff do also occur but are generally limited to the shelf regions of Greenland (e.g., LACAN & JEANDEL 2004, LAUKERT et al. 2017a). The sea-ice cover prevents aeolian inputs of REEs to reach the entire AO in winter and year-round in the central AO. Instead, the particulate REEs are most likely scavenged by snow, deposited on the sea ice and only released during snow and sea-ice melting. No REE concentrations have been reported as yet for Arctic snow deposited on the sea-ice cover and the contribution of REE inputs originating from snow melting is thus unknown. However, compared to the riverine contributions, the aeolian REE inputs to the AO are expected to be comparatively small. Vertical redistribution of REEs within the water column through sea-ice formation and melting was recently suggested to play an important role for the marine REE budget of the Siberian shelves (LAUKERT et al. 2017b).

METHODS

All seawater data considered here have been published previously. Dissolved Nd isotope and REE concentration data were reported for the Laptev Sea (LAUKERT et al. 2017b) and the Fram Strait (LAUKERT et al. 2017a). Additional dissolved Nd isotope and concentration data were mainly reported for the central AO and the Canada Basin (ANDERSSON et al. 2008, PORCELLI et al. 2009, ZIMMERMANN et al. 2009).

New Nd isotope and REE data were determined for essentially sediment-free (based on optical control) sea-ice samples recovered in the Eurasian Basin during expedition PS80 (ARKXXVII/3, BOETIUS 2013) of the German research vessel RV "Polarstern" between 2 August and 8 October 2012 (Fig. 1). The four sea-ice cores were recovered with a Titanium corer and stored in clean plastic tubes, before they were transferred into acid-cleaned plastic barrels, wrapped up in several layers of plastic bags and melted at ~4 °C. The salinity of the sea-ice meltwater was determined by using a Salinometer (WTW Cond. 3310). For Nd isotope and REE analyses, the
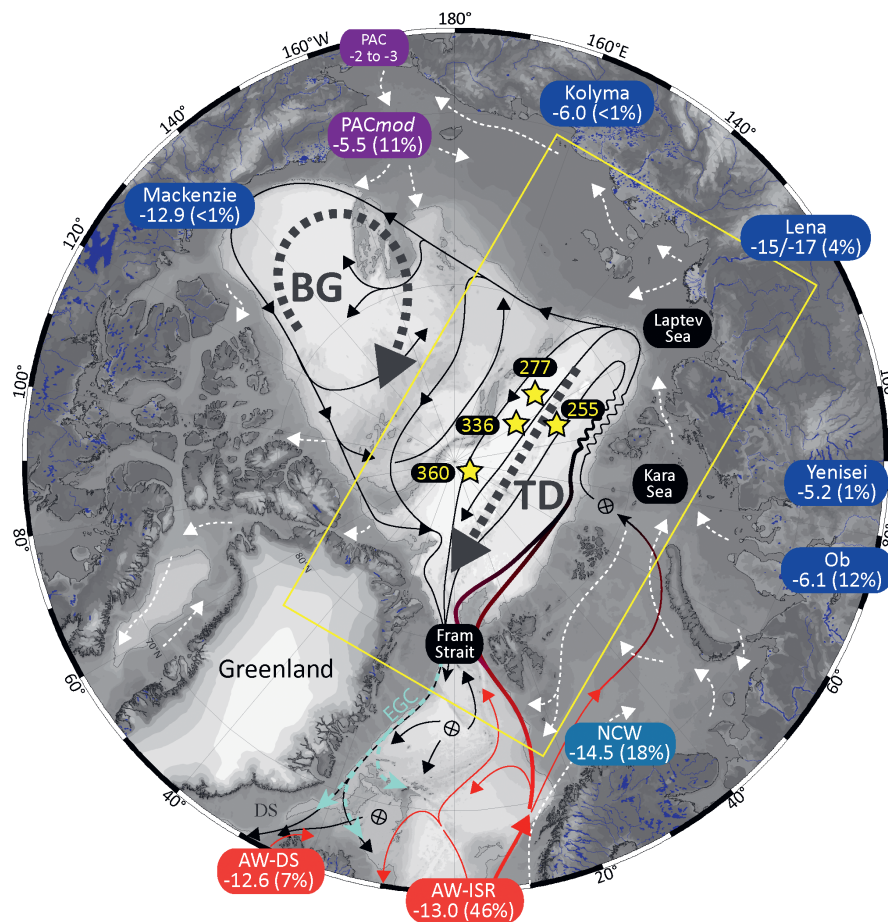
Fig. 1: Bathymetric map of the Arctic Ocean (IBCAO, Jakobsson et al. 2012) with known REE sources (Andersson et al. 2008, Charette et al. 2016, Porcelli et al. 2009, Zimmermann et al. 2009, Persson et al. 2011, Laukert et al. 2017a, b). The circulation scheme of the upper layers (large dashed grey arrows represent the Transpolar Drift (TD) and the Beaufort Gyre (BG), while dashed white lines indicate major shelf currents) and subsurface Atlantic and intermediate depth currents (solid black lines; red lines in addition indicate cooling and freshening of Atlantic-derived waters) are shown in addition (modified after Rudels et al. 2012). Dashed cyan lines indicate the East Greenland Current (EGC). REE sources with known ε_Nd values are Atlantic-derived waters (AW) entering through the Iceland-Scotland Ridge (ISR) and the Denmark Strait (DS), Norwegian Coastal Water (NCW), Pacific-derived waters (PAC) and modified Pacific-derived waters (PACmod), major Siberian Rivers (Ob, Yenisei, Lena, Kolyma), and the Mackenzie River. The estimated Nd inputs of the corresponding sources are given as percentage of the total Nd input into the AO taking into account volume transport and [Nd] of the sources (only rivers with discharges > 100 km³/yr are considered). Circled crosses indicate sites of convection or sinking from intermediate and AW layers to deeper levels. Locations of the sea-ice samples obtained during PS80 are shown together with their station numbers. The area shown in Fig. 3 is marked by the yellow rectangle. The figure was modified from Laukert et al. (2017a) after production with Ocean Data View (Schlitzer 2017) and manual modification.

Abb. 1: Karte des Nordpolarmeeres mit Bathymetrie (IBCAO, (Jakobsson et al. 2012) und den bekannten REE Quellen (Andersson et al. 2008, Charette et al. 2016, Porcelli et al. 2009, Zimmermann et al. 2009, Persson et al. 2011, Laukert et al. 2017a, b). Die Zirkulation der Oberflächenwässer (große gestrichelte graue Pfeile symbolisieren die Transpolardrift (TD) und den Beaufortwirbel (BG), wohingegen gestrichelte weiße Pfeile die Zirkulation auf den Schelfen darstellen) sowie des Atlantischen Wassers und der Zwischenwässer (schwarze Pfeile; rote Pfeile deuten zudem die Abkühlung und Abnahme des Salzgehaltes vom Atlantischen Wasser an) ist zusätzlich dargestellt (modifiziert nach Rudels et al. 2012). Der Ostgrönlandstrom (EGC) ist ebenfalls angedeutet (türkise Pfeile). Die REE Quellen mit bekannten Nd-Isotopensignaturen sind Atlantisches Wasser (AW) das den Island-Schottland-Rücken (ISR) und die Dänemarkstraße (DS) passiert, Norwegisches Küstenwasser (NCW), Pazifisches Wasser (PAC) und modifiziertes Pazifisches Wasser (PACmod), sowie die Sibirischen Flüsse (Ob, Jenissei, Lena, Kolyma) und der Mackenzie. Die geschätzten Nd-Einträge dieser Quellen sind als Prozentwerte angegeben und basieren auf deren Abflussmengen und Nd-Konzentrationen (hier wurden nur Flüsse mit Einträgen über 100 km³ pro Jahr berücksichtigt). Umrundete Kreuze markieren die Hauptgebiete der Tiefenwasserbildung. Die Positionen der während der Expedition PS80 gewonnenen Meereisproben sind zusammen mit den Stationsnummern gegeben. Das in Abb. 3 gezeigte Gebiet ist gelb umrahmt. Die Abbildung wurde nach Laukert et al. (2017a) modifiziert, nachdem sie mit Ocean Data View (Schlitzer 2017) erstellt und manuell bearbeitet wurde.

remaining meltwater was transported to the home laboratory at GEOMAR, Kiel. During a second transfer into acid-cleaned canisters, only the upper parts of the meltwater samples were transferred to avoid incorporation of any settled particles. Further sample treatment including the pre-concentration, extraction and isolation of Nd was performed following the standard procedures applied for seawater (see Laukert et al. 2017a). The Nd isotope compositions were measured on a Nu Plasma (Nu Instruments Limited) and a Neptune Plus (Thermo Scientific) MC-ICP-MS at GEOMAR and at the University of Oldenburg. For correction of instrumental mass bias an expo-

nential mass fractionation was applied using a $^{146}Nd/^{144}Nd$ ratio of 0.7219. The measured $^{143}Nd/^{144}Nd$ ratios of the four samples were normalized to the value of 0.512115 for the JNdi-1 standard (Tanaka et al. 2000). Based on repeated measurements of JNdi-1 and in-house standards with concentrations similar to those of the samples, the 2σ external reproducibility ranged between 0.2 and 0.5 ε_Nd units for the individual measurement runs.

The REE concentrations were determined on centrifuged aliquots of the sea-ice meltwater using an online pre-con-

centration (OP) ICP-MS technique at GEOMAR by directly coupling a "seaFAST" system (Elemental Scientific Inc., Nebraska, USA) to an ICP-MS (Agilent 7500ce) (HATHORNE et al. 2012). The method of HATHORNE et al. (2012) was further improved by using an 8 ml sample loop and by preparation of calibration standards with a mixed REE solution of a seawater-like composition in a natural seawater matrix (see also OSBORNE et al. 2015). Repeated measurements of GEOTRACES inter-calibration sample BATS 15 m (VAN DE FLIERDT et al. 2012) were used to monitor the external reproducibility, which at the 95 % confidence level (2σ) is below 8 % for La, Ce, Pr, Nd, Gd, Dy, Er and Yb, and below 12 % for Sm, Eu, Tb, Ho, Tm and Lu.

To determine the pathways of the sea ice and to identify the source areas, the locations of the samples were tracked backward using a combination of ice drift and concentration data obtained from low-resolution passive microwave satellites. Ice motion data used in this study are provided by different institutions and have been widely used in sea-ice studies (e.g., SPREEN et al. 2011, SUMATA et al. 2014, KRUMPEN et al. 2016). To track sea ice, two different sets of ice drift products were used: During the summer months (June to August), the Polar Pathfinder Sea Ice Motion product provided by the National Snow and Ice Data Center (NSIDC) given on a 25 km grid (TSCHUDI et al. 2016) was applied. During the rest of the year, tracking is achieved with sea-ice motion data provided by the Center for Satellite Exploitation and Research (CERSAT) at the Institut Francais de Recherche pour d'Exploitation de la Mer (IFREMER). Motion data are available with a grid size of 62.5 km, using time intervals of 3 days for the period between September and May (GIRARD-ARDHUIN & EZRATY 2012). Sea-ice concentration data used in this study have been obtained from NSIDC. Following KRUMPEN et al. (2016) the tracking procedure works as follows: Using motion and concentration data, a specific ice area is tracked backwards until: (a) the ice reaches a position next to a coastline, (b) the ice concentration at a specific location reaches a threshold value of 15 %, below which ice parcels are considered lost, or (c) the tracking time exceeds 4 years.

RESULTS AND DISCUSSION

Tracing Siberian freshwater pathways with Nd isotopes

Despite the known difficulties associated with the modifications of the seawater Nd isotope composition through seawater-particle interactions in particular Arctic regions (see section 2), a general limitation for the quantitative determination of shallow water mass compositions and thus the assessment of freshwater pathways based on Nd isotope mixing calculations is given by the fact that the source-defined water mass components can only be quantified with Nd isotopes and salinity or Nd concentrations ([Nd]) if three or less Nd sources contribute (i.e., due to the availability of only two "conservative" parameters). Most shallow waters in the AO are characterized by the contribution of four or more Nd sources due to admixture of freshwater from the various Arctic rivers, inhibiting unambiguous quantification of their source-defined components. In particular, this is likely the case for shallow Arctic-derived Polar Water that left the AO in summer 2012 through the western Fram Strait via the East Greenland Current (EGC). LAUKERT et al. (2017a) found that the average Nd isotope composition of this water mass plots close to the mixing line between Atlantic-derived waters that enter the AO through Fram Strait and modified Pacific-derived waters originating from the Chukchi Sea shelf in ε_{Nd}-salinity and ε_{Nd}-[Nd] space (Fig. 2). The offset from the mixing line towards lower salinity and higher [Nd] was attributed to admixture of meteoric waters (neglecting precipitation, this is essentially only riverine freshwater). The amount of these waters was found to be 8 % or less based on mass balance calculations involving stable oxygen isotopes and nutrients. LAUKERT et al. (2017a) noted that contributions of riverine Nd can originate from any of the major Arctic rivers and would result in varying increases in [Nd] and changes in ε_{Nd}. In fact, as discussed below, contributions from more than one Arctic river to this Polar Water are highly likely, which strictly speaking inhibits quantification of the source-defined components. Nevertheless, LAUKERT et al. (2017a) argued that the low amount of riverine freshwater contained in Polar Water in the Fram Strait implies a relatively low contribution of riverine Nd and suggested that the shift of the ε_{Nd} signature caused by addition of riverine Nd was generally small. After excluding significant shifts of the ε_{Nd} signature caused by addition of riverine Nd, LAUKERT et al. (2017a) estimated the proportion of Atlantic- and modified Pacific-derived waters and concluded that the Polar Water had been advected through the Amundsen and Makarov Basins based on a comparison with Nd isotope literature data.

The suggestion by LAUKERT et al. (2017a) regarding the relatively small shift in the ε_{Nd} signature of Polar Water exported from the AO caused by the addition of riverine freshwater is supported by the fact that most of the riverine dissolved Nd is removed in the estuaries and on the Siberian shelves (LAUKERT et al. 2017b). Another argument for a relatively small shift in the ε_{Nd} signature is that freshwater from likely more than one Arctic river was admixed to the Polar Water, as mentioned above. The average Nd isotope composition of the major Arctic rivers is near -8.5 based on a calculation that takes into account discharge (data from R-ArcticNET, LAMMERS & SHIKLOMANOV 2000) and [Nd] (data from PORCELLI et al. 2009, ZIMMERMANN et al. 2009, PERSSON et al. 2011). This signature is similar to that of the exported Polar Water (ε_{Nd} = -8.8, Fig. 2), implying that, assuming contributions from all major Arctic rivers, the marine Nd isotope composition of the Polar Water was not strongly modified through the admixture of the freshwater. Mixing between freshwater derived from the different Siberian rivers was previously suggested to occur even before the Polar Water is advected into the central AO (PORCELLI et al. 2009). The limited data available from the Siberian shelves, however, did not allow a straightforward determination of the individual contributions of the Siberian rivers (cf., PORCELLI et al. 2009).

New dissolved Nd isotope and REE data recently reported by LAUKERT et al. (2017b) for the Laptev Sea filled the data gap between the Siberian estuaries and the central AO and now allow a more detailed determination of the riverine freshwater pathways. Relatively radiogenic freshwater (ε_{Nd} = -6) is advected from the Kara Sea to the Laptev Sea via the Vilkitsky Strait Current (Fig. 3), where it mixes with northward flowing relatively unradiogenic freshwater supplied by the Lena River (ε_{Nd} = -15 to -16 in summer). The Kara Sea freshwater is likely a mixture between freshwater supplied by the Yenisei
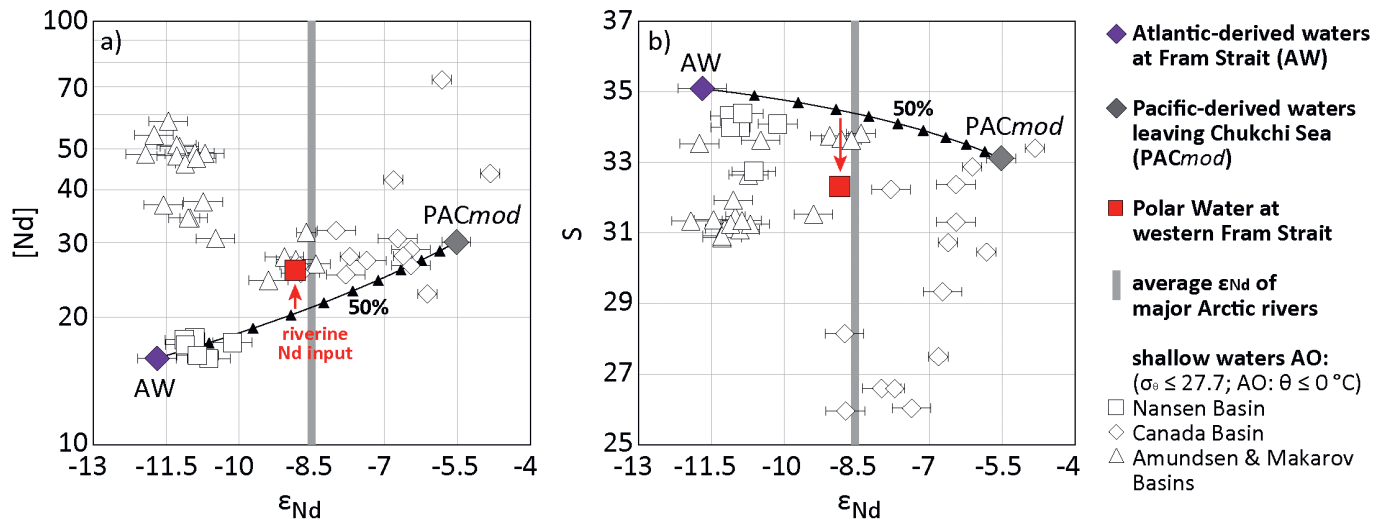
Fig. 2: Assessment of the Arctic source of Polar Water that leaves the AO through the Fram Strait (modified after Laukert et al. 2017a). The most radiogenic PW sample (red symbol) can be explained by admixture of AW and PAC*mod* and a small amount (6-8 %) of river water (red arrows). The AW to PAC*mod* ratio for this Polar Water is about 3:2. The Polar Water at Fram Strait shares characteristics with shallow samples from the Amundsen and Makarov Basins. The average Nd isotope composition of all Arctic rivers is shown as a grey line.

Abb. 2: Bestimmung der Zusammensetzung des arktischen Wassers, das das Nordpolarmeer durch die Framstraße verlässt (modifiziert nach Laukert et al. 2017a). Die radiogenste Probe dieser Wassermasse (rotes Symbol) kann durch eine Mischung zwischen AW und PAC*mod* und kleineren Zumischungen (6-8 %) von arktischem Flusswasser erklärt werden. Das Verhältnis von AW und PAC*mod* im arktischen Wasser ist etwa 3:2 und weist Ähnlichkeiten zu oberflächennahen Proben aus dem Amundsen und dem Makarov Becken auf. Die durchschnittliche Nd-Isotopenzusammensetzung aller arktischen Flüsse ist als graue Linie dargestellt.
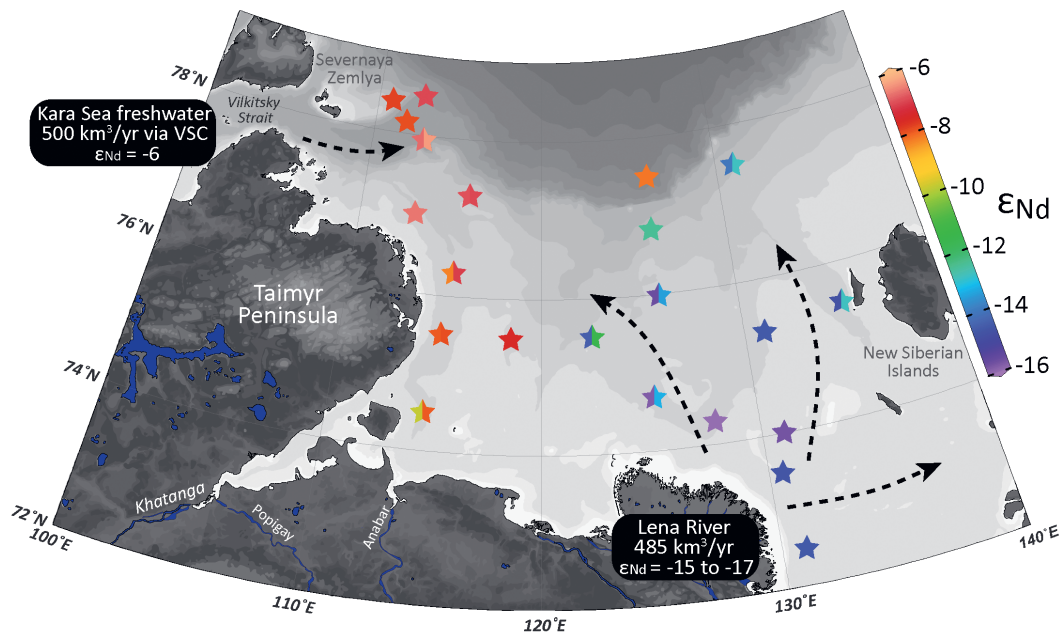

Fig. 3: Surface distribution of Nd isotope compositions (color-coded stars) of the summers of 2013 and 2014 (see Laukert et al. 2017b for the distribution during the individual years) shown together with the general circulation scheme (Janout et al., 2015). For resampled stations, the ε_{Nd} signatures of both years are shown (divided color-coded stars). Figure was produced using Ocean Data View (Schlitzer 2017) and modified manually.

Abb. 3: Darstellung der Oberflächenverteilung der Nd-Isotopensignaturen (farbkodierte Sterne) aus den Sommern der Jahre 2013 und 2014 (siehe Laukert et al. 2017b für die Verteilung der einzelnen Jahre) zusammen mit der schematischen Zirkulation (Janout et al., 2015). Für doppelt beprobte Stationen sind beide Nd-Isotopensignaturen dargestellt (unterteilte farbkodierte Sterne). Die Abbildung wurde mit Ocean Data View (Schlitzer 2017) erstellt und manuell bearbeitet.

and Ob rivers, both characterized by relatively radiogenic ε_{Nd} signatures between -5 and -6, which can be inferred from Nd isotope based mixing calculations (Laukert et al. 2017b) and results of three-dimensional coupled ice-ocean-models implying extensive mixing of freshwater supplied by these two rivers within the surface layer of the Kara Sea (Harms et al. 2000). The transport of Kara Sea freshwater through the Vilkitsky Strait was determined to be about 500 km³/yr (Janout et al. 2015), which is equivalent to about half of the freshwater volume supplied by the two major rivers draining into the Kara Sea. Most of the Kara Sea freshwater that enters the Laptev Sea is advected eastwards along the Laptev margin, and only a small proportion is advected onto the Laptev shelf or into the Eurasian Basin (Janout et al. 2015). Admixture of

northward flowing freshwater from the Lena River to the Kara Sea freshwater is documented by less radiogenic seawater ε_{Nd} signatures in the northeastern Laptev Sea (LAUKERT et al. 2017b), and most of the shelf waters that leave the Laptev Sea towards the central AO are characterized by ε_{Nd} signatures ranging between -8 and -14 and [Nd] between 30 and 100 pmol/kg (for Nd isotope signatures, see Fig. 3). These characteristics are similar to those reported for shallow seawater samples recovered from the Amundsen and Makarov Basins (Fig. 2; PORCELLI et al. 2009, ZIMMERMANN et al. 2009), indicating that mixing between Kara Sea freshwater and Lena River freshwater in variable amounts occurs before the Siberian waters are advected to the central AO. The limited data available from the central AO do not allow to constrain if pure Kara Sea shelf waters in addition advected to the central AO and transported at least to some extent within the TD (e.g., potentially advected through the passage between the Franz Josef Land and Severnaya Zemlya or through the Barents Sea). However, the observations by LAUKERT et al. (2017a) summarized above imply that mixing with freshwater derived from other rivers and modified Pacific-derived waters at the latest occurs before the Siberian waters are advected to the western Fram Strait and leave the AO via the EGC.

Our observations show that the contribution of various Nd sources with similar characteristics often inhibits a quantitative assessment of the individual contributions of the different Arctic rivers based on Nd isotopes, which complicates assessments of Arctic freshwater pathways. A more reliable determination of freshwater pathways is only possible for regions where three or less Nd sources contribute, as for example shown for the Laptev Sea (LAUKERT et al. 2017b). Nevertheless, given that all major Siberian rivers have relatively radiogenic Nd isotope compositions except the Lena River (major contributions of the Mackenzie River to shallow waters in the Eurasian Basin are unlikely, cf., PORCELLI et al. 2009), one prospective application of Nd isotopes for the Eurasian Basin may be to qualitatively trace the admixture and transport of Lena River freshwater in the TD. This, however, requires more seawater Nd isotope and concentration data from the central AO.

Tracing source areas of Arctic sea ice with Nd isotopes and REEs

The new sea-ice data presented here (Tab. 1) are available from the PANGAEA database. Compared to most freshwater-influenced shallow waters in the AO, the sea ice is characterized by lower salinities and overall lower REE concentrations (in the following represented by [Nd]). The sea-ice sample from station 336 has the lowest salinity (S = 0.94), the most radiogenic Nd isotope composition with ε_{Nd} = -7.6 ±0.2 and the highest [Nd] near 85 pmol/kg, whereas the sea-ice sample from station 360 has the highest salinity (S = 2.39), the least radiogenic Nd isotope composition with ε_{Nd} = -10.8 ±0.5 and the lowest [Nd] near 4 pmol/kg (see also Figs. 4 and 5). Sea-ice samples recovered from stations 255 and 277 are characterized by intermediate values. The PAAS-normalized (MCLENNAN 2001) REE patterns of sea-ice samples recovered at stations 255, 277 and 336 are essentially flat or slightly hump-shaped (Fig 5). In contrast, the REE pattern of the sea ice sampled at station 360 is more similar to open ocean seawater patterns

Station	PS80-255	PS80-277	PS80-336	PS80-360
Date collected	20/08/2012	25/08/2012	07/09/2012	22/09/2012
Latitude	82.6707	82.8825	85.1018	88.8277
Longitude	109.5895	130.1295	122.2453	58.8635
Ice thickness (cm)	92	–	84	194
Salinity*	1.89	–	0.94	2.39
$^{143}Nd/^{144}Nd$	0.512228	0.512205	0.512250	0.512082
ε_{Nd}	-8.0	-8.4	-7.6	-10.8
2 σ ε_{Nd}	0.2	0.2	0.2	0.5
[L]	15,9	8.3	81.8	5.7
[Ce]	37.6	18.7	165.3	5.4
[Pr]	5.0	2.9	24.5	1.0
[Nd]	16.8	10.6	85.1	3.7
[Sm]	3.2	1.8	15.6	0.7
[Eu]	0.6	0.4	3.1	0.1
[Gd]	2.7	2.1	15.3	0,6
[Tb]	0.4	0.3	2.1	0.1
[Dy]	2.2	1.3	11.6	0.8
[Ho]	0.4	0.3	2.2	0.2
[Er]	1.0	0.9	6.1	0.5
[Tm]	0.1	0.1	0.7	0.1
[Yb]	1.0	0.7	4.0	0.6
[Lu]	0.2	0.1	0.6	0.1
HREE / LREE**	0.8	1.2	0.7	2.3

Tab. 1: Sampling date, location, ice thickness, salinity, Nd isotope compositions and REE concentrations for the sea-ice samples recovered during cruise PS80 (ARKXXVII/3) on RV "Polarstern". * core average, ** normalized to PAAS (MCLENNAN 2001).

Tab. 1: Zeitpunkt der Beprobung, Beprobungsort, Eismächtigkeit, Salinität, Nd-Isotopensignaturen und REE Konzentrationen von Meereisproben, die während der Expedition PS80 (ARKXXVII/3) mit FS „Polarstern" genommen wurden. * Eiskern Mittelwert; ** normiert nach PAAS (MCLENNAN 2001).

(Fig. 5), which commonly exhibit a depletion of the light (L) REEs compared to the heavy (H)REEs and a pronounced Ce anomaly (e.g., HATHORNE et al. 2015).

The sea-ice trajectories determined based on satellite-derived ice drift and concentration data suggest that the sea ice with the more radiogenic Nd isotope signature (station 336) likely formed east of Vilkitsky Strait, while the sea ice with the least radiogenic signature (station 360) presumably formed north of the Laptev Sea (Fig. 4). The sea ice with slightly less radiogenic ε_{Nd} signatures below -8 was formed north (station 255) and south (station 277) of the eastern part of the Vilkitsky Strait. The comparison with seawater Nd isotope compositions determined for surface waters in the Laptev Sea (LAUKERT et al. 2017b) reveals that the ε_{Nd} signatures of the sea ice are similar to those expected for the surface waters of its suggested source areas (Fig. 4). In detail, the most radiogenic sea-ice floes potentially derived from the area east of Vilkitsky Strait (station 336; ε_{Nd} = -7.6) and thus must have formed in
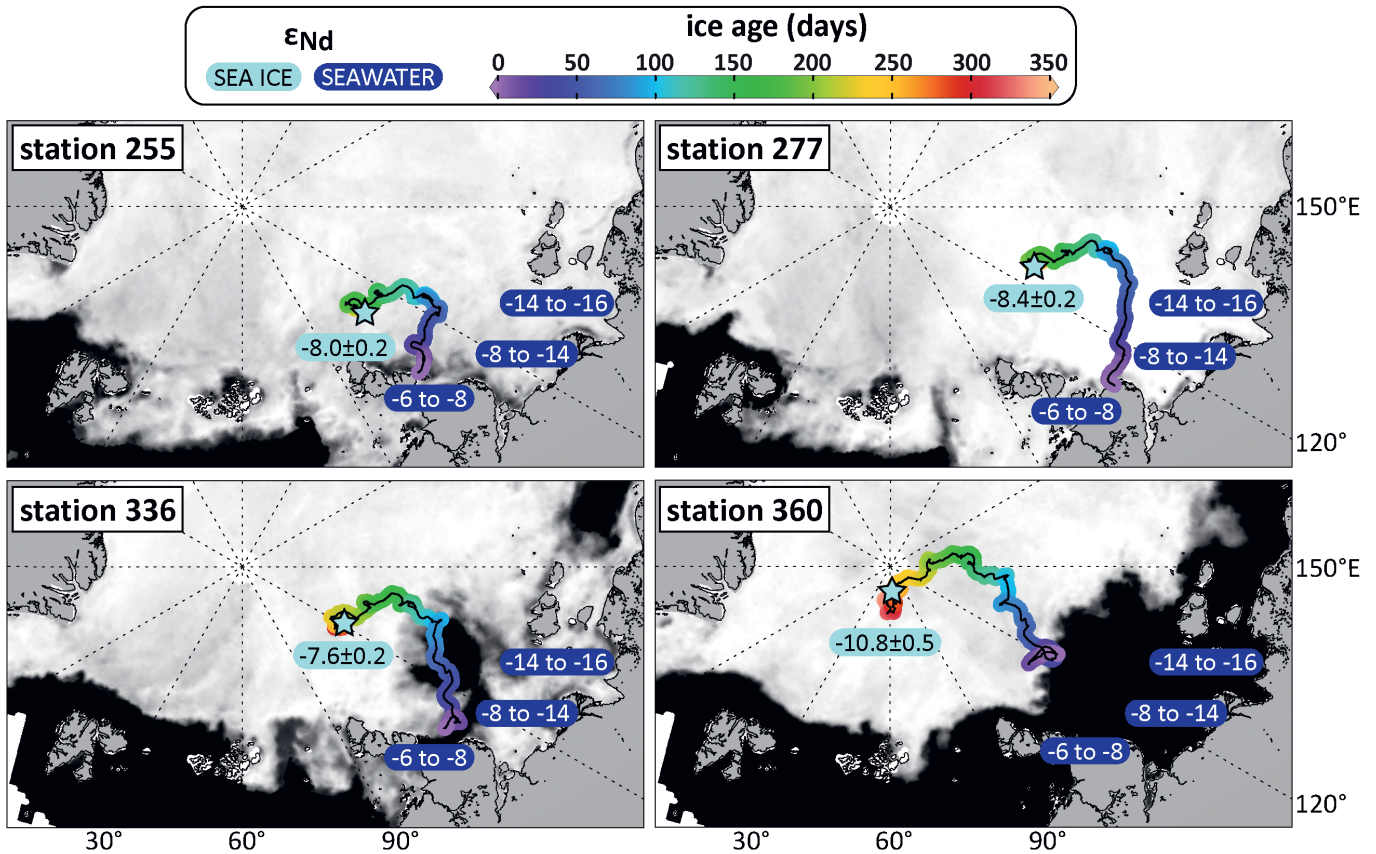
Fig. 4: Maps showing the sea ice extent during the recovery of the four sea-ice samples in 2012, along with drift trajectories based on satellite-derived data (color coded for the age of the ice) and the Nd isotope compositions of the sea-ice samples and surface waters of the Laptev Sea (see also LAUKERT et al. 2017b). The sea-ice signatures are similar to those expected in surface waters of the source areas of the drifting sea ice. Figures were produced using Ocean Data View (SCHLITZER 2017) and modified manually.

Abb. 4: Meereisbedeckung während der Beprobung der Eisproben im Jahr 2012 zusammen mit den auf Satellitendaten basierenden Drift-Trajektorien und den Nd-Isotopenzusammensetzungen der Meereisproben und der Oberflächenwässer der Laptewsee (siehe auch LAUKERT et al. 2017b). Die Nd-Isotopensignaturen der Eisproben sind konsistent mit den Oberflächenwassersignaturen, die in den Entstehungsgebieten des Meereises erwartet werden. Die Abbildungen wurden mit Ocean Data View (SCHLITZER 2017) erstellt und manuell bearbeitet.
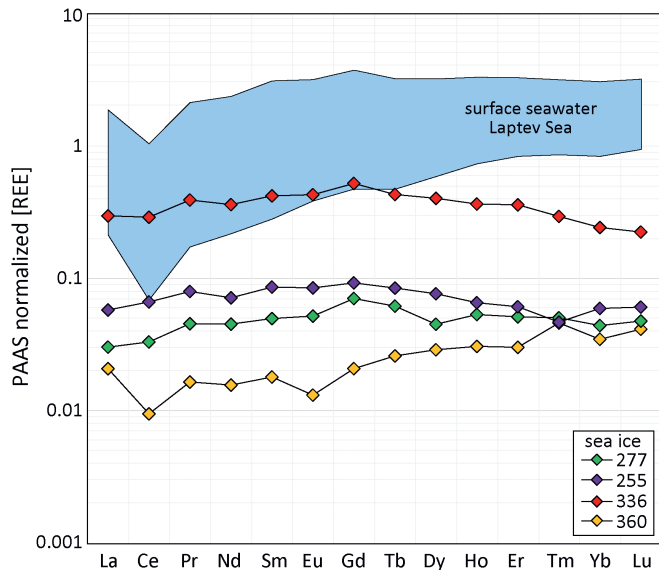
Fig. 5: PAAS normalized (MCLENNAN 2001) REE distribution patterns of the four sea-ice samples shown together with surface waters from the Laptev Sea (data from LAUKERT et al. 2017b).

Abb. 5: PAAS-normierte (MCLENNAN 2001) REE-Verteilungsmuster der vier Meereisproben zusammen mit den Konzentrationen der Oberflächenwässer aus der Laptewsee (Daten von LAUKERT et al. 2017b).

shelf waters with relatively radiogenic ε_{Nd} signatures reflecting high contributions of Kara Sea freshwater (ε_{Nd} = -6). The latter is advected to this area via the Vilkitsky Strait Current, which flows eastward along the Laptev shelf margin (JANOUT et al. 2015). Sea-ice floes derived from the areas north and south of the eastern Vilkitsky Strait (stations 255 and 277) likely formed in waters that contain less Kara Sea freshwater and therefore have slightly less radiogenic seawater Nd isotope compositions (ε_{Nd} = -8.0 to -8.4). Even the less radiogenic signature (ε_{Nd} = -10.8) of the sea ice that originated north of the Laptev Sea (station 360) is in agreement with formation in surface waters presumably resulting from a mixture of Kara Sea freshwater and Lena River freshwater ($\varepsilon_{Nd} \le$ -15).

The geochemical sea-ice characteristics described above, together with the information obtained from our backtracking approach based on satellite data support recent considerations that the marine and riverine REEs are only to a small extent incorporated into the sea ice (LAUKERT et al. 2017b). However, the data also suggest that Nd isotope and REE signatures are preserved during sea-ice transport. It has been shown earlier that small fractions of seawater-derived salt and other seawater constituents (e.g., dissolved organic matter and inorganic nutrients) remain trapped in the interstitial brine after the sea ice has been formed (THOMAS et al. 2010 and refer-

ences therein). Based on our observations, we suggest that the incorporation of the marine or riverine REEs is proportional to this salt entrapment. This is best evidenced by a negative correlation between [Nd] and salinity and a positive correlation between the HREE/LREE ratio and salinity in our sea-ice samples, which agrees with an inverse relationship between the REE concentrations and salinity observed in seawater of the Laptev Sea (LAUKERT et al. 2017b).

The LREE concentrations in the sea ice recovered from station 336 are highest (e.g., [Nd] = 85 pmol/kg), and thus even higher than those of the surface waters in its suggested source area (~50 pmol/kg, LAUKERT et al. 2017b). These elevated LREE concentrations may either reflect incorporation of younger Kara Sea freshwater characterized by less riverine REE removal or may result from contributions of particulate REEs (i.e., >0.2 µm) that may have been present in the unfiltered molten sea-ice. In addition, the HREE concentrations of all sea-ice samples are slightly more depleted than the HREE concentrations of surface waters from the Laptev Sea (Fig. 5), resulting in generally lower HREE/LREE ratios in the sea-ice samples than in the corresponding Siberian shelf waters. The difference in REE patterns between the sea ice and the source waters may also be related to contribution of particulate REEs (e.g., contribution of particles with elevated LREEs and MREEs), or alternatively reflect differences in the incorporation of the distinct dissolved size pools characterized by dissimilar REE distribution patterns. Such different incorporation of the truly dissolved (e.g., <1 KDa) and colloidal (e.g., >1 KDa and <0.2 µm) REEs during sea-ice formation is in line with the general observation that particle size plays a key role for the incorporation of seawater constituents into the sea ice (JANSSENS et al. 2016 and references therein). No truly dissolved REE data have been reported for open Arctic Ocean seawater to date, but marked differences between the REE patterns of the truly dissolved and the colloidal fractions have recently been observed for riverine freshwater in the Amazon Basin (e.g., MERSCHEL et al. 2017b). We therefore speculate that such differences also exist for the different size fractions of the freshwater-rich surface layer of the Laptev Sea, and that combined with size-dependent REE incorporation during sea-ice growth they may account for the depletion of HREEs observed in our sea-ice samples. However, here we refrain from further discussions of this observation given that we cannot entirely exclude contributions from partially dissolving particulate phases in our samples. Apart from the differences between the sea ice and the seawater data, the high correlation between the REE concentrations and the HREE/LREE ratios with salinity argue for at least partial preservation of the seawater REE patterns of the source waters. This is supported by the more seawater-like REE pattern of sea ice from station 360 that mirrors REE characteristics of higher salinity waters the ice presumably formed from.

CONCLUDING REMARKS

Tracing freshwater pathways in the Arctic Ocean with Nd isotopes is challenging due to contributions of various riverine REE sources with similar characteristics. For shallow waters circulating on the Siberian shelves (i.e., the main source regions of the TD), new observations indicate that Nd isotopes can successfully be applied to quantify the individual contri-

butions of the different Siberian rivers. Further downstream the TD, admixture of freshwater from other Arctic rivers and Pacific-derived waters prevents such a quantitative assessment of the individual freshwater contributions. Nevertheless, the distinctly unradiogenic Nd isotope composition of the Lena River may still allow at least for a qualitative assessment of the transport of freshwater it supplies, thereby also providing important information on freshwater transport and mixing in the TD. However, more seawater Nd isotope and concentration data are needed to reliably trace the transport of Lena River freshwater in the Eurasian Basin of the Arctic Ocean.

Apart from studies discussing the provenance of sediment entrained and transported within drifting Arctic sea ice in the context of paleoceanographic reconstructions (e.g., NÜRN-BERG et al. 1994, TÜTKEN et al. 2002, WERNER et al. 2014), there are no previously published works using Nd isotopes or REEs to determine the source regions of drifting Arctic sea ice. We show for the first time that these tracers can be used to identify the source areas of sea ice and their mixtures provided that the marine and riverine Nd isotope compositions and the corresponding REE distribution patterns of all source areas are known. Our most striking observation is that marine and riverine Nd isotope compositions are preserved during sea-ice formation and long-range transport. For sea ice formed close to distinct REE sources, this approach has the potential to avoid the problem of mixing of multiple sources that affects freshwater tracing in the AO based on dissolved surface water signatures. We further observe that the incorporation of the REEs into the sea ice is proportional to salinity variations and that the REE characteristics are at least partly preserved. However, contributions from particulate phases cannot be entirely excluded for our unfiltered sea-ice samples, which clearly requires dedicated future studies that investigate REE characteristics of the different size fractions in sea ice and their differential incorporation during sea-ice formation.

References

Alkire, M.B., Falkner, K.K., Rigor, I., Steele, M. & Morison, J. (2007): The return of Pacific waters to the upper layers of the central Arctic Ocean.- Deep Sea Res. PT I 54: 1509-1529, doi.org/10.1016/j.dsr.2007.06.004

Anderson, L.G., Jutterstrom, S., Kaltin, S., Jones, E.P. & Bjork, G.R. (2004): Variability in river runoff distribution in the Eurasian Basin of the Arctic Ocean.- J. Geophys. Res. Oceans 109, doi.org/10.1029/2003jc001773

Andersson, P.S., Porcelli, D., Frank, M., Bjork, G., Dahlqvist, R. &

Gustafsson, O. (2008): Neodymium isotopes in seawater from the Barents Sea and Fram Strait Arctic-Atlantic gateways.- Geochim. Cosmochim. Acta 72: 2854-2867, http://dx.doi.org/10.1016/j.gca.2008.04.008

Arsouze, T., Dutay, J.C., Lacan, F. & Jeandel, C. (2009): Reconstructing the Nd oceanic cycle using a coupled dynamical – biogeochemical model.- Biogeosci. 6: 2829-2846, doi.org/10.5194/bg-6-2829-2009

Bamber, J., van den Broeke, M., Ettema, J., Lenaerts, J. & Rignot, E. (2012): Recent large increases in freshwater fluxes from Greenland into the North Atlantic.- Geophys. Res. Lett. 39: L19501, doi.org/10.1029/2012gl052552

Bauch, D., Dmitrenko, I.A., Wegner, C., Hölemann, J., Kirillov, S.A., Timokhov, L.A. & Kassens, H. (2009): Exchange of Laptev Sea and Arctic Ocean halocline waters in response to atmospheric forcing.- J. Geophys. Res. 114, C05008, doi.org/10.1029/2008jc005062

Bauch, D., Rutgers van der Loeff, M., Andersen, N., Torres-Valdes, S., Bakker, K. & Abrahamsen, E. P. (2011): Origin of freshwater and polynya water in the Arctic Ocean halocline in summer 2007.- Prog. Oceanogr. 91: 482-95, doi.org/10.1016/j.pocean.2011.07.017

Boetius, A. (2013): The expedition of the research vessel "Polarstern" to the Arctic in 2012 (ARK-XXVII/3).- Rep. Polar Mar. Res. 663: 1-166.

Charette, M.A., Lam, P.J., Lohan, M.C., Kwon, E.Y., Hatje, V., Jeandel, C., Shiller, A.M., Cutter, G.A., Thomas, A., Boyd, P.W., Homoky, W.B., Milne, A., Thomas, H., Andersson, P.S., Porcelli, D., Tanaka, T., Geibert, W., Dehairs, F. & Garcia-Orellana, J. (2016): Coastal ocean and shelf-sea biogeochemical cycling of trace elements and isotopes: lessons learned from GEOTRACES.- Philos. Trans. A Math. Phys. Eng. Sci. I 374, doi.org/10.1098/rsta.2016.0076

Dahlqvist, R., Andersson, P.S. & Porcelli, D. (2007): Nd isotopes in Bering Strait and Chukchi Sea Water.- Geochim. Cosmochim. Acta 71(15): A196.

de Steur, L., Pickart, R.S., Macrander, A., Våge, K., Harden, B., Jónsson, S., Østerhus, S. & Valdimarsson, H. (2017): Liquid freshwater transport estimates from the East Greenland Current based on continuous measurements north of Denmark Strait.- J. Geophys. Res. Oceans 122: 93-109, doi.org/10.1002/2016jc012106

Dittmar, T. & Kattner, G. (2003): The biogeochemistry of the river and shelf ecosystem of the Arctic Ocean: a review.- Mar. Chem. 83: 103-20, doi.org/10.1016/s0304-4203(03)00105-1

Fichot, C.G., Kaiser, K., Hooker, S.B., Amon, R.M., Babin, M., Belanger, S., Walker, S.A. & Benner, R. (2013): Pan-Arctic distributions of continental runoff in the Arctic Ocean.- Sci. Rep. 3: 1053, doi.org/10.1038/srep01053

Frank, M. (2002): Radiogenic isotopes: Tracers of past ocean circulation and erosional input.- Rev. Geophys. 40, doi.org/10.1029/2000rg000094

Garcia-Solsona, E., Jeandel, C., Labatut, M., Lacan, F., Vance, D., Chavagnac, V. & Pradoux, C. (2014): Rare earth elements and Nd isotopes tracing water mass mixing and particle-seawater interactions in the SE Atlantic.- Geochim. Cosmochim. Acta 125: 351-372, doi.org/10.1016/j.gca.2013.10.009

Girard-Ardhuin, F. & Ezraty, R. (2012): Enhanced Arctic Sea Ice Drift Estimation Merging Radiometer and Scatterometer Data.- IEEE Trans. Geosci. Remote Sens. 50: 2639-2648, doi.org/10.1109/tgrs.2012.2184124

Goldstein, S.L. & Hemming, S.R. (2003): Long-lived Isotopic Tracers in Oceanography, Paleoceanography, and Ice-sheet Dynamics.- Treatise on Geochemistry 6: 453-489, doi.org/10.1016/B0-08-043751-6/06179-X

Gordeev, V.V. (2006): Fluvial sediment flux to the Arctic Ocean.- Geomorphology 80: 94-104, doi.org/10.1016/j.geomorph.2005.09.008

Haley, B.A., Frank, M., Hathorne, E.C. & Pisias, N. (2014): Biogeochemical implications from dissolved rare earth element and Nd isotope distributions in the Gulf of Alaska.- Geochim. Cosmochim. Acta 126: 455-474, doi.org/10.1016/j.gca.2013.11.012

Harms, I.H., Karcher, M.J. & Dethleff, D. (2000): Modelling Siberian river runoff — implications for contaminant transport in the Arctic Ocean.- J. Mar. Syst. 27: 95-115, doi.org/10.1016/s0924-7963(00)00062-2

Hathorne, E.C., Haley, B.A., Stichel, T., Grasse, P., Zieringer, M. & Frank, M. (2012): Online preconcentration ICP-MS analysis of rare earth elements in seawater.- Geochem. Geophys. Geosyst. 13: Q01020, doi.org/10.1029/2011gc003907

Hathorne, E.C., Stichel, T., Bruck, B. & Frank, M. (2015): Rare earth element distribution in the Atlantic sector of the Southern Ocean: The balance between particle scavenging and vertical supply.- Mar. Chem. 177: 157-171, doi.org/10.1016/j.marchem.2015.03.011

Jacobsen, S. B. & Wasserburg, G. J. (1980): Sm-Nd Isotopic Evolution of Chondrites.- Earth Planet. Sci. Lett. 50: 139-155. http://dx.doi.org/10.1016/0012-821x(80)90125-9.

Jakobsson, M., Mayer, L., Coakley, B., Dowdeswell, J.A., Forbes, S., Fridman, B., Hodnesdal, H., Noormets, R., Pedersen, R., Rebesco, M., Schenke, H.W., Zarayskaya, Y., Accettella, D., Armstrong, A., Anderson, R.M., Bienhoff, P., Camerlenghi, A., Church, I., Edwards, M., Gardner, J.V., Hall, J.K., Hell, B., Hestvik, O., Kristoffersen, Y., Marcussen, C., Mohammad, R., Mosher, D., Nghiem, S.V., Pedrosa, M.T., Travaglini, P. G. & Weatherall, P. (2012): The International Bathymetric Chart of the Arctic Ocean (IBCAO) Version 3.0.- Geophys. Res. Lett. 39: L12609, doi.org/10.1029/2012gl052219

Janout, M.A., Aksenov, Y., Hölemann, J.A., Rabe, B., Schauer, U., Polyakov, I.V., Bacon, S., Coward, A.C., Karcher, M., Lenn, Y.D., Kassens, H. & Timokhov, L. (2015): Kara Sea freshwater transport through Vilkitsky Strait: Variability, forcing, and further pathways toward the western Arctic Ocean from a model and observations.- J. Geophys. Res. Oceans 120: 4925-4944, doi.org/10.1002/2014jc010635

Janssens, J., Meiners, K.M., Tison, J.-L., Dieckmann, G., Delille, B. & Lannuzel, D. (2016): Incorporation of iron and organic matter into young Antarctic sea ice during its initial growth stages.- Elementa: Sci. Anthrop., 4, doi.org/10.12952/journal.elementa.000123

Jones, E.P., Anderson, L.G., Jutterstrom, S., Mintrop, L. & Swift, J.H. (2008): Pacific freshwater, river water and sea ice meltwater across Arctic Ocean basins: Results from the 2005 Beringia Expedition.- J. Geophys. Res. Oceans 113: C08012, doi.org/10.1029/2007jc004124

Jones, E.P., Anderson, L.G. & Swift, J.H. (1998): Distribution of Atlantic and Pacific waters in the upper Arctic Ocean: Implications for circulation.- Geophys. Res. Lett. 25: 765-768, doi.org/10.1029/98gl00464

Jones, E.P. (2001): Circulation in the Arctic Ocean.- Polar Res. 20: 139-146, doi.org/10.1111/j.1751-8369.2001.tb00049.x

Karcher, M., Gerdes, R. & Kauker, F. (2006): Modeling of $\delta^{18}O$ and 99Tc dispersion in Arctic and subarctic seas.- ASOF Newsletter, Issue No. 5.

Krumpen, T., Gerdes, R., Haas, C., Hendricks, S., Herber, A., Selyuzhenok, V., Smedsrud, L. & Spreen, G. (2016): Recent summer sea ice thickness surveys in Fram Strait and associated ice volume fluxes.- The Cryosphere 10: 523-534, doi.org/10.5194/tc-10-523-2016

Lacan, F. & Jeandel, C. (2001): Tracing Papua New Guinea imprint on the central Equatorial Pacific Ocean using neodymium isotopic compositions and Rare Earth Element patterns.- Earth Planet. Sci. Lett. 186: 497-512, doi.org/10.1016/S0012-821x(01)00263-1

Lacan, F. & Jeandel, C. (2004): Denmark Strait water circulation traced by heterogeneity in neodymium isotopic compositions.- Deep Sea Res. PT I 51: 71-82, doi.org/10.1016/j.dsr.2003.09.006

Lacan, F. & Jeandel, C. (2005): Neodymium isotopes as a new tool for quantifying exchange fluxes at the continent–ocean interface.- Earth Planet. Sci. Lett. 232: 245-257, doi.org/10.1016/j.epsl.2005.01.004

Lacan, F., Tachikawa, K. & Jeandel, C. (2012): Neodymium isotopic composition of the oceans: A compilation of seawater data.- Chem. Geol. 300-301: 177-184, doi.org/10.1016/j.chemgeo.2012.01.019

Lammers, R.B. & Shiklomanov, A.I. (2000): R-ArcticNet, a regional hydrographic data network for the Pan-Arctic Region. Water Systems Analysis Group, University of New Hampshire. Distributed by the National Snow and Ice Data Center, Durham. Data available at http://www.r-arcticnet.sr.unh.edu/v4.0/index.html (accessed 01.06.2017)

Laukert, G., Frank, M., Bauch, D., Hathorne, E.C., Rabe, B., von Appen, W.-J., Wegner, C., Zieringer, M. & Kassens, H. (2017a): Ocean circulation and freshwater pathways in the Arctic Mediterranean based on a combined Nd isotope, REE and oxygen isotope section across Fram Strait.- Geochim. Cosmochim. Acta 202: 285-309, doi.org/10.1016/j.gca.2016.12.028

Laukert, G., Frank, M., Bauch, D., Hathorne, E.C., Gutjahr, M., Janout, M. & Hölemann, J. (2017b): Transport and transformation of riverine Nd isotope and rare earth element signatures in high latitude estuaries: A case study from the Laptev Sea.- Earth Planet. Sci. Lett. 477: 205-217. doi.org/10.1016/j.epsl.2017.08.010

Mauritzen, C. (2012): Oceanography: Arctic freshwater.- Nat. Geosci. 5: 162-164, doi.org/10.1038/ngeo1409

McLennan, S.M. (2001): Relationships between the trace element composition of sedimentary rocks and upper continental crust.- Geochem. Geophys. Geosyst. 2: 1021, doi.org/10.1029/2000gc000109

Merschel, G., Bau, M. & Dantas, E.L. (2017a): Contrasting impact of organic and inorganic nanoparticles and colloids on the behavior of particle-reactive elements in tropical estuaries: An experimental study.- Geochim. Cosmochim. Acta 197: 1-13, doi.org/10.1016/j.gca.2016.09.041

Merschel, G., Bau, M., Schmidt, K., Münker, C. & Dantas, E.L. (2017b): Hafnium and neodymium isotopes and REY distribution in the truly dissolved, nanoparticulate/colloidal and suspended loads of rivers in the Amazon Basin, Brazil.- Geochim. Cosmochim. Acta 213: 383-399, doi.org/10.1016/j.gca.2017.07.006

Molina-Kescher, M., Frank, M. & Hathorne, E.C. (2014): South Pacific dissolved Nd isotope compositions and rare earth element distributions: Water mass mixing versus biogeochemical cycling.- Geochim. Cosmochim. Acta 127: 171-189, doi.org/10.1016/j.gca.2013.11.038

Nozaki, Y. & Alibo, D.S. (2003): Dissolved rare earth elements in the Southern Ocean, southwest of Australia: Unique patterns compared to the South Atlantic data.- Geochem. J. 37: 47-62, doi.org/10.2343/geochemj.37.47

Nürnberg, D., Wollenburg, I., Dethleff, D., Eicken, H., Kassens, H., Letzig, T., Reimnitz, E. & Thiede, J. (1994): Sediments in Arctic sea ice: Implications for entrainment, transport and release.- Mar. Geol. 119: 185-214, doi.org/10.1016/0025-3227(94)90181-3

Osborne, A.H., Haley, B.A., Hathorne, E.C., Plancherel, Y. & Frank, M. (2015): Rare earth element distribution in Caribbean seawater: Continental inputs versus lateral transport of distinct REE compositions

in subsurface water masses.- Mar. Chem. 177: 172-183 doi.org/10.1016/j.marchem.2015.03.013

Persson, P.O., Andersson, P.S., Porcelli, D. & Semiletov, I. (2011): The influence of Lena River water inflow and shelf sediment-sea water exchange for the Nd isotopic composition in the Laptev Sea and Arctic Ocean.- EGU General Assembly Conference Abstracts 13: 1-672.

Petrova, M. (2015): Neodymium isotopes and rare earth element distribution in the Barents Sea, Arctic Ocean.- Master thesis, Saint Petersburg State University / University of Hamburg, Saint Petersburg / Hamburg. 1-60.

Pokrovsky, O.S., Shirokova, L.S., Viers, J., Gordeev, V.V., Shevchenko, V.P., Chupakov, A.V., Vorobieva, T.Y., Candaudap, F., Causserand, C., Lanzanova, A. & Zouiten, C. (2014): Fate of colloids during estuarine mixing in the Arctic.- Ocean Sci. 10: 107-125, doi.org/10.5194/os-10-107-2014

Porcelli, D., Andersson, P.S., Baskaran, M., Frank, M., Bjork, G. & Semiletov, I. (2009): The distribution of neodymium isotopes in Arctic Ocean basins.- Geochim. Cosmochim. Acta 73: 2645-2659, doi.org/10.1016/j.gca.2008.11.046

Rabe, B., Dodd, P.A., Hansen, E., Falck, E., Schauer, U., Mackensen, A., Beszczynska-Möller, A., Kattner, G., Rohling, E.J. & Cox, K. (2013): Liquid export of Arctic freshwater components through the Fram Strait 1998-2011.- Ocean Sci. 9: 91-109, doi.org/10.5194/os-9-91-2013

Rabe, B., Schauer, U., Mackensen, A., Karcher, M., Hansen, E. & Beszczynska-Möller, A. (2009): Freshwater components and transports in the Fram Strait – recent observations and changes since the late 1990s.- Ocean Sci. 5: 219-233, doi.org/10.5194/os-5-219-2009

Rahmstorf, S., Box, J.E., Feulner, G., Mann, M. E., Robinson, A., Rutherford, S. & Schaffernicht, E.J. (2015): Exceptional twentieth-century slowdown in Atlantic Ocean overturning circulation.- Nat. Clim. Chang. 5: 475-480, doi.org/10.1038/nclimate2554

Recinos, B., Rabe, B. & Schauer, U. (2016): Upper Arctic Ocean velocity structure from in-situ observations.- EGU General Assembly Conference Abstracts 18, 6809.

Rempfer, J., Stocker, T.F., Joos, F., Dutay, J.C. & Siddall, M. (2011): Modelling Nd-isotopes with a coarse resolution ocean circulation model: Sensitivities to model parameters and source/sink distributions.- Geochim. Cosmochim. Acta 75: 5927-5950, doi.org/10.1016/j.gca.2011.07.044

Rudels, B., Anderson, L., Eriksson, P., Fahrbach, E., Jakobsson, M., Jones, E.P., Melling, H., Prinsenberg, S., Schauer, U. & Yao, T. (2012): Observations in the ocean.- Arctic Climate Change, 43: 117-198, doi.org/10.1007/978-94-007-2027-5_4

Rudels, B., Anderson, L. G. & Jones, E. P. (1996): Formation and evolution of the surface mixed layer and halocline of the Arctic Ocean.- J. Geophys. Res. Oceans 101: 8807-8821, doi.org/10.1029/96jc00143

Rudels, B., Friedrich, H. J. & Quadfasel, D. (1999): The arctic circumpolar boundary current.- Deep Sea Res. PT II 46: 1023-1062, doi.org/10.1016/S0967-0645(99)00015-6

Schlitzer, R. (2017): Ocean Data View. http://odv.awi.de

Siddall, M., Khatiwala, S., van de Flierdt, T., Jones, K., Goldstein, S.L., Hemming, S. & Anderson, R.F. (2008): Towards explaining the Nd paradox using reversible scavenging in an ocean general circulation model.- Earth Planet. Sci. Lett. 274: 448-461, doi.org/10.1016/j.epsl.2008.07.044

Smedsrud, L.H., Halvorsen, M.H., Stroeve, J.C., Zhang, R. & Kloster, K. (2017): Fram Strait sea ice export variability and September Arctic sea ice extent over the last 80 years.- The Cryosphere 11: 65-79, doi.org/10.5194/tc-11-65-2017

Spreen, G., Kwok, R. & Menemenlis, D. (2011): Trends in Arctic sea ice drift and role of wind forcing: 1992-2009.- Geophys. Res. Lett. 38: L19501. doi.org/10.1029/2011gl048970

Sumata, H., Lavergne, T., Girard-Ardhuin, F., Kimura, N., Tschudi, M.A., Kauker, F., Karcher, M. & Gerdes, R. (2014): An intercomparison of Arctic ice drift products to deduce uncertainty estimates.- J. Geophys. Res. Oceans 119: 4887-4921, doi.org/10.1002/2013jc009724

Tachikawa, K, Athias, V & Jeandel, C. (2003): Neodymium budget in the modern ocean and paleo-oceanographic implications.- J. Geophys. Res. Oceans 108, doi.org/10.1029/1999jc000285

Tanaka, T., Togashi, S., Kamioka, H., Amakawa, H., Kagami, H., Hamamoto, T., Yuhara, M., Orihashi, Y., Yoneda, S., Shimizu, H., Kunimaru, T., Takahashi, K., Yanagi, T., Nakano, T., Fujimaki, H., Shinjo, R., Asahara, Y., Tanimizu, M. & Dragusanu, C. (2000): JNdi-1: a neodymium isotopic reference in consistency with LaJolla neodymium.- Chem. Geol. 168: 279-281, doi.org/10.1016/S0009-2541(00)00198-4

Taylor, J.R., Falkner, K.K., Schauer, U. & Meredith, M. (2003): Quantitative considerations of dissolved barium as a tracer in the Arctic Ocean.- J. Geophys. Res. Oceans 108, doi.org/10.1029/2002jc001635

Tepe, N. & Bau, M. (2016): Behavior of rare earth elements and yttrium during simulation of arctic estuarine mixing between glacial-fed river waters and seawater and the impact of inorganic (nano-)particles.- Chem. Geol. 438: 134-145, doi.org/10.1016/j.chemgeo.2016.06.001

Thomas, D.N., Papadimitriou, S. & Michel, C. (2010): Biogeochemistry of Sea Ice.- In: D.N. Thomas & G. Dieckmann (eds), Sea Ice, Wiley-Blackwell, 425-467, doi.org/10.1002/9781444317145.ch12

Tschudi, M., Fowler, C., Maslanik, J., Stewart, J.S. & Meier, W. (2016): Polar Pathfinder Daily 25 km EASE-Grid Sea Ice Motion Vectors, Version 3.- NSIDC National Snow and Ice Data Center, Boulder, Colorado USA, doi.org/10.5067/O57VAIT2AYYY

Tütken, T., Eisenhauer, A., Wiegand, B. & Hansen, B.T. (2002): Glacial–interglacial cycles in Sr and Nd isotopic composition of Arctic marine sediments triggered by the Svalbard/Barents Sea ice sheet.- Mar. Geol. 182: 351-372, doi.org/10.1016/s0025-3227(01)00248-1

van de Flierdt, T., Griffiths, A.M., Lambelet, M., Little, S.H., Stichel, T. & Wilson, D.J. (2016): Neodymium in the oceans: a global database, a regional comparison and implications for palaeoceanographic research.- Philos. Trans. R. Soc. A 374: 20150293, doi.org/10.1098/rsta.2015.0293

van de Flierdt, T., Pahnke, K., Amakawa, H., Andersson, P.S., Basak, C., Coles, B., Colin, C., Crocket, K., Frank, M., Frank, N., Goldstein, S.L., Goswami, V., Haley, B.A., Hathorne, E.C., Hemming, S.R., Henderson, G.M., Jeandel, C., Jones, K., Kreissig, K., Lacan, F., Lambelet, M., Martin, E.E., Newkirk, D.R., Obata, H., Pena, L., Piotrowski, A.M., Pradoux, C., Scher, H.D., Schöberg, H., Singh, S.K., Stichel, T., Tazoe, H., Vance, D. & Yang, J. (2012): GEOTRACES intercalibration of neodymium isotopes and rare earth element concentrations in seawater and suspended particles. Part 1: reproducibility of results for the international intercomparison.- Limnol. Oceanogr. Methods 10: 234-251, doi.org/10.4319/lom.2012.10.234

Werner, K., Frank, M., Teschner, C., Muller, J. & Spielhagen, R.F. (2014): Neoglacial change in deep water exchange and increase of sea-ice transport through eastern Fram Strait: evidence from radiogenic isotopes.- Quat. Sci. Rev. 92: 190-207, doi.org/10.1016/j.quascirev.2013.06.015

Woodgate, R.A., Aagaard, K. & Weingartner, T.J. (2006): Interannual changes in the Bering Strait fluxes of volume, heat and freshwater between 1991 and 2004.- Geophys. Res. Lett. 33, doi.org/10.1029/2006gl026931

Woodgate, R.A., Weingartner, T.J. & Lindsay, R. (2012): Observed increases in Bering Strait oceanic fluxes from the Pacific to the Arctic from 2001 to 2011 and their impacts on the Arctic Ocean water column.- Geophys. Res. Lett. 39: L24603, doi.org/10.1029/2012gl054092

Yang, J. & Haley, B. (2016): The profile of the rare earth elements in the Canada Basin, Arctic Ocean.- Geochem. Geophys. Geosyst. 17: 3241-3253, doi.org/10.1002/2016gc006412

Yang, Q., Dixon, T.H., Myers, P.G., Bonin, J., Chambers, D. & van den Broeke, M.R. (2016): Recent increases in Arctic freshwater flux affects Labrador Sea convection and Atlantic overturning circulation.- Nat. Commun. 7: 10525, doi.org/10.1038/ncomms10525

Zimmermann, B., Porcelli, D., Frank, M., Andersson, P.S., Baskaran, M., Lee, D.C. & Halliday, A.N. (2009): Hafnium isotopes in Arctic Ocean water.- Geochim. Cosmochim. Acta 73: 3218-3233, doi.org/10.1016/j.gca.2009.02.028