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The distribution of neodymium isotopes in Arctic Ocean basins

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Abstract

Nd concentration and isotope data have been obtained for the Canada, Amundsen, and Makarov Basins of the Arctic Ocean. A pattern of high Nd concentrations (up to 58 pM) at shallow depths is seen throughout the Arctic, and is distinct from that generally seen in other oceans where surface waters are relatively depleted. A range of isotopic variations across the Arctic and within individual depth profiles reflects the different sources of waters. The dominant source of water, and so Nd, is the Atlantic Ocean, with lesser contributions from the Pacific and Arctic Rivers. Radiogenic isotope Nd signatures (up to $\varepsilon_{Nd} = -6.5$) can be traced in Pacific water flowing into the Canada Basin. Waters from rivers draining older terrains provide very unradiogenic Nd (down to $\varepsilon_{Nd} = -14.2$) that can be traced in surface waters across much of the Eurasian Basin. A distinct feature of the Arctic is the general influence of the shelves on the Nd concentrations of waters flowing into the basins, either from the Pacific across the Chukchi Sea, or from across the extensive Siberian shelves. Water–shelf interaction results in an increase in Nd concentration without significant changes in salinity in essentially all waters in the Arctic, through processes that are not yet well understood. In estuarine regions other processes modify the Nd signal of freshwater components supplied into the Arctic Basin, and possibly also contribute to sedimentary Nd that may be subsequently involved in sediment–water interactions. Mixing relationships indicate that in estuaries, Nd is removed from major river waters to different degrees. Deep waters in the Arctic are higher in Nd than the inflowing Atlantic waters, apparently through enrichments of waters on the shelves that are involved in ventilating the deep basins. These enrichments generally have not resulted in major shifts in the isotopic compositions of the deep waters in the Makarov Basin ($\varepsilon_{Nd} \sim -10.5$), but have created distinctive Nd isotope signatures that were found near the margin of the Canada Basin (with $\varepsilon_{Nd} \sim -9.0$). The deep waters of the Amundsen Basin are also distinct from the Atlantic waters (with $\varepsilon_{Nd} = -12.3$), indicating that there has been limited inflow from the adjacent Makarov Basin through the Lomonosov Ridge.

1. INTRODUCTION

The Arctic Ocean is an important part of the global climate system. Its perennial ice cover exerts a strong control on the surface heat balance, since it reflects a large part of the incoming solar radiation during summer and acts as an insulating shield during winter. The Arctic also has an influence on the large-scale thermohaline circulation of the world ocean through the influence of the export of freshwater into the N. Atlantic (Aagaard and Carmack, 1994). Circulation within the Arctic is affected by inflows from the Pacific and Atlantic Oceans, as well as by riverine inputs

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and freshwater redistribution by sea ice. Such factors have clearly been susceptible to significant alteration due to the global climatic changes that affect ice cover, precipitation, and global circulation patterns (Broecker, 1997). In turn, changes in fluxes and geochemical processes along the vast and shallow Siberian shelf seas may also have had an impact on the marine biogeochemical cycles on a larger scale.

A number of different tracers have been used to trace the distribution and circulation within the Arctic Ocean of waters originating on the shelves, rivers, and in the adjacent basins. These include nutrients that can be used to distinguish Pacific and Atlantic waters (e.g. Wilson and Wallace, 1990; Jones et al., 1998; Ekwurzel et al., 2001), Ra isotopes that can identify shelf inputs (Smith et al., 2003), O isotopes that can distinguish between meltwater and riverine inputs (Ekwurzel et al., 2001; Macdonald et al., 2002), anthropogenic radionuclides that follow circulation patterns (e.g. Alfimov et al., 2004), U that can identify different riverine inputs (Andersen et al., 2007), ^{14}C that reflects ventilation ages (Schlosser et al., 1997), and Ba that can trace river inputs (Guay and Falkner, 1997). Nd provides an additional tracer for investigating modern ocean processes as well as past circulation patterns.

Decay of ^{147}Sm ($t_{1/2} = 1.06 \times 10^{11}$ yr) to ^{143}Nd produces variations in $^{143}Nd/^{144}Nd$ ratios (reported as ε_{Nd}, parts in 10^4 deviations from the CHUR standard; see Table 1) in continental rocks, and so in rivers supplying Nd to the oceans. In seawater, the isotopic composition of Nd varies both between and within ocean basins, since the average oceanic residence time of Nd in the oceans of ~500 yr (Tachikawa et al., 2003) is shorter than the time required for homogenising the oceanic Nd composition. In general, there is a correlation between the age of a continental terrain and the isotopic composition of Nd it supplies to the ocean (see review by Goldstein and Hemming, 2003). The resulting variations in different water masses can be used as fingerprints to study global circulation patterns.

As shown in Fig. 1, the waters entering the Arctic Ocean have a range of distinctive signatures. Pacific waters have relatively high ratios, with surface waters in the N. Pacific up to $\varepsilon_{Nd} \sim -2$, (Piepgras and Jacobsen, 1988) due to the erosion of the relatively young volcanics on the Pacific margins, although waters entering the Arctic have somewhat lower values (Dahlqvist et al., 2007). In contrast, waters from the N. Atlantic entering the Arctic through the Fram Strait and Barents Sea have values of −10.8 (Andersson et al., 2008). Waters draining from the Precambrian shields are expected to have very low values, as confirmed by values as low as −26 in the Baffin Bay (Stordal and Wasserburg, 1986). Major Arctic Rivers have values that range from −5 to −14, based on limited river water and sediment data (Guo et al., 2004; Zimmermann et al., 2009) and reflecting the more diverse age distributions elsewhere in the region.

There are often significant losses of riverine Nd occurring in estuaries (Elderfield et al., 1990). Exchange of Nd between seawater and the continental margins also occurs (Lacan and Jeandel, 2004a,b, 2005a,b). In some cases this leads to changes in the seawater Nd isotope composition without substantially altering the Nd concentration, so that exchange with sediments, rather than simple release of additional Nd, is clearly occurring (Arsouze et al., 2007). However, the exact processes involved are not understood. This has been observed even where continental shelves are not prominent (Lacan and Jeandel, 2004c), though it is likely to have a substantial effect on the extensive Arctic shelves. Submarine groundwater discharges may also be a significant source of Nd (Johannesson and Burdige, 2007).

Unlike many other oceanographic tracers, records of past Nd isotope compositions of seawater can be obtained from various sediments and authigenic deposits that incorporate seawater Nd, and these have been used to reconstruct past circulation patterns and weathering rates (Winter et al., 1997, Abouchami et al., 1999; Vance and

Table 1
Nd concentrations and isotopic compositions of Canada Basin waters.

Station	Depth (m)	$^{143}Nd/^{144}Nd$	$2\sigma_{mean}$	ε_{Nd} [a]	C_{Nd} [b] (pM)	Salinity (psu)	Θ[c] (°C)
Stn 3, 3850 m	5	0.5121912	±0.00002	−8.7 ± 0.4	25.3	25.92	−1.22
75°12.51′N 149°56.96 W	25	0.5121908	±0.00001	−8.7 ± 0.4	26.3	28.12	−0.98
24-Aug-2000	50	0.512308	±0.000013	−6.4 ± 0.4	26.4	31.29	−0.068
	85	0.512239	±0.000014	−7.8 ± 0.4	25.1	32.22	−1.14
	400	0.512146	±0.000024	−9.6 ± 0.4	19.3	34.79	0.50
	1000	0.512092	±0.000022	−10.7 ± 0.4	15.5	34.88	−0.027
	3000	0.512072	±0.000020	−11.0 ± 0.4	17.1	34.96	−0.51
Stn 4, 3894 m	5	0.512261	±0.000019	−7.4 ± 0.4	27.1	26.00	−1.19
73°49.50′N 152°00.73′W	25	0.512294	±0.000019	−6.7 ± 0.4	30.6	29.31	−0.69
26-Aug-2000	85	0.512308	±0.000016	−6.4 ± 0.4	28.8	32.36	−1.27
	500	0.512173	±0.000022	−9.1 ± 0.4	16.1	34.83	0.48
	2500	0.512176	±0.000015	−9.0 ± 0.4	21.6	34.95	−0.52
Stn 5, 1200 m	5	0.512229	±0.000015	−8.0 ± 0.4	32.0	26.56	−0.44
72°14.67′N 155°04.46′W	400	0.512174	±0.000016	−9.1 ± 0.4	20.5	34.82	0.75
28-Aug-2000							

[a] $\varepsilon_{Nd} = [(143Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{CHUR} - 1] \times 10^4$; where $(^{143}Nd/^{144}Nd)_{CHUR} = 0.512638$. Error includes external reproducibility.
[b] Uncertainty is estimated to be <5%.
[c] Potential temperature.

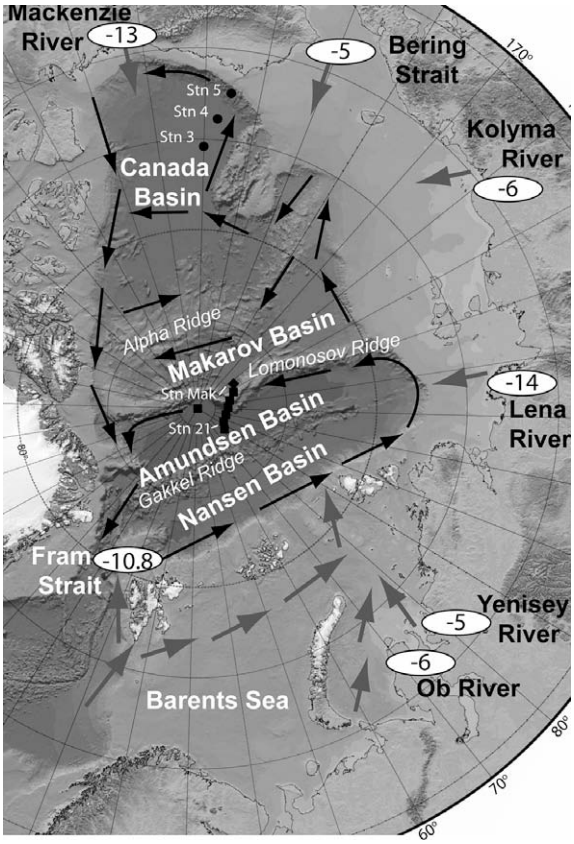

Fig. 1. Waters entering the Arctic Ocean (large arrows) have a wide range of Nd isotope compositions, with radiogenic Nd from the Pacific (Dahlqvist et al., 2007), and much less radiogenic values in rivers draining Archean shield areas. River values range from −5 to −14, based on limited river water data (Zimmermann et al., 2009). Nd with −10.8 is brought into the basin from the north Atlantic (Andersson et al., 2008). The three sampling locations in the Canada Basin are shown, along with the full profile collected in the Makarov (Stn Mak) and Amundsen (Stn 21) Basins. Other locations where shallow waters were collected in the Amundsen Basin are also shown. The black arrows indicate the general circulation pattern of the intermediate waters (Rudels et al., 2004). The bathymetric map is taken from the International Bathymetric Chart of the Arctic Ocean (IBCAO); www.ngdc.noaa.gov/bathymetry/arctic/arctic.html.

Burton, 1999; Frank, 2002; Piotrowski et al., 2005; von Blanckenburg and Nagler, 2001; Haley et al., 2008). Nd therefore can provide an important tool for understanding changes that have occurred in the Arctic, including changing interactions with the surrounding ocean basins and continents. However, understanding how the present distribution of Nd isotopes in the oceans reflects both the processes supplying Nd and the present ocean circulation patterns is an essential prerequisite for understanding past ocean conditions.

In this study, seawater Nd concentrations and isotope compositions are reported for the first time for the Canada, Makarov, and Amundsen Basins of the Arctic Ocean (Fig. 1), and together with data from the Nansen Basin, Barents Sea, and Fram Strait (Andersson et al., 2008), provide the first assessment of the distribution and budget of Nd isotopes within the Arctic, the relationships between Nd signals and Arctic water masses, and the role of shelf-seawater exchange.

2. SAMPLING AND ANALYTICAL METHODS

2.1. Arctic waters

A significant feature of the Arctic Ocean (Fig. 1) is the expanse of Siberian continental shelf, which constitutes about half of the basin and comprises about 25% of the global continental shelves. The deep Arctic Ocean is divided by the prominent Lomonosov Ridge into the Canadian Basin, which in turn is subdivided by the Alpha Ridge into the Canada Basin and the Makarov Basin, and the smaller Eurasian Basin, which is subdivided by the Gakkel Ridge into the Amundsen and Nansen Basins. Water circulation in the Arctic Ocean is influenced by these sub-parallel dividing ridges (see e.g. Jones, 2001). Characterization of the distribution of Nd in the Arctic therefore requires consideration of waters within each basin as well as those flowing across the shelves.

The Arctic Ocean water column is strongly stratified (see Aagaard et al., 1985; Carmack, 1990; Jones et al., 1991), as shown in Figs. 2 and 3. The Cold Halocline Layer (CHL), a characteristic cold, low salinity surface layer, typically extends down to about 100 m depth and covers the entire Arctic Ocean (Rudels et al., 1996). It is strongly affected by inputs from Arctic Rivers and melting sea ice. The uppermost 10–20 m of the CHL is typically well mixed by ice motion and convection and is referred to as the Polar mixed layer (PML). Below the CHL is the warm Atlantic layer (AL), defined by a temperature above 0 °C and typically extending to a depth of ~750 m (Rudels et al., 2004). Below is a weak thermocline of the Upper Polar deep water (UPDW) down to ~1700 m depth, below which is the cold, more saline Polar deep water (PDW) (Jones, 2001; Rudels et al., 2004). In the Canada Basin, Pacific winter waters (PWW), with salinities around $S = 33.1$, temperatures near freezing, and high nutrient concentrations (Jones and Anderson, 1986) constitute much of the upper halocline above the Atlantic water. Summer water from the Pacific, with $S < 33$ and transported along the Alaskan coast as Alaskan current water (ACW) can be traced as a temperature maximum above the PWW (Steele et al., 2004).

2.2. Sampling

Locations and total water depths of the sampling stations are reported in Tables 1 and 2. Water samples from the Amundsen and Makarov Basins (Fig. 1) were collected on the Swedish icebreaker *I/B Oden* during the Arctic Ocean expedition AO-01 from June to August 2001 (see Björk et al., 2002); sampling is described in detail in Andersson et al. (2008). The Makarov Basin is represented by a single profile extending throughout the water column. In the Amundsen Basin, a depth profile at Station 21 was complemented by more intensive sampling of the upper 100 m at nearby Station 22, and another surface water sample at nearby Station 20. Four additional surface water samples

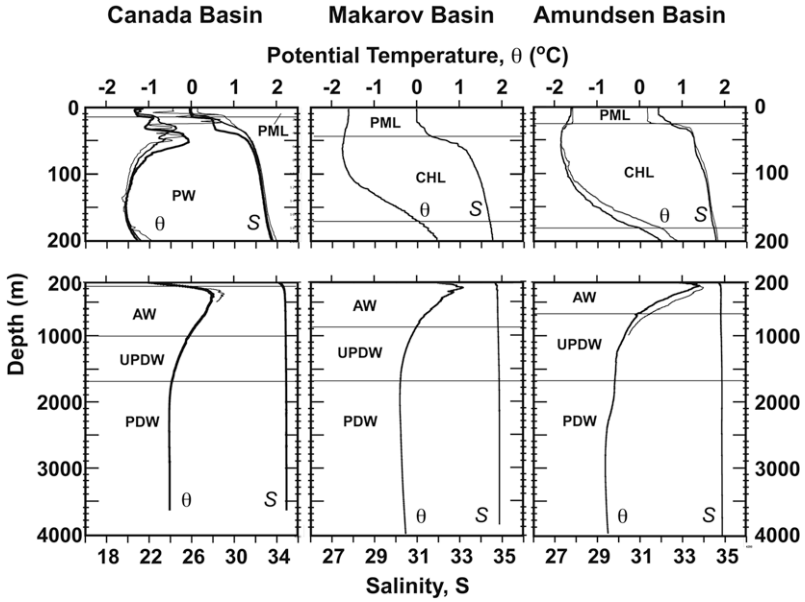

Fig. 2. The salinity and potential temperature structure for the deep profiles sampled at each basin in the upper 200 m (upper panel) and throughout the water column (lower panel). The three sampled stations are shown for the Canada Basin (Station 3, thick line, Station 4, medium line, and Station 5, fine line). In the Amundsen, both the deep profile for Station 21 (thick line), and the shallow profile for Station 22, are shown. The uppermost 10–20 m, the Polar mixed layer (PML), is well mixed and has lowered salinities. The Cold Halocline Layer (CHL) in the Eurasian Basins, with relatively constant low temperatures as salinity increases with depth. Below the CHL is the prominent temperature maximum of the Atlantic layer (AL), defined by a temperature above 0 °C (Rudels et al., 2004). This is underlain by Upper Polar Deep water to 1700 m, and the quite uniform Polar deep water (PDW) at greater depths. In the Canada Basin, the CHL is composed of Pacific waters; Pacific winter waters, with salinities around $S = 33.1$ and temperatures near freezing, underlain by summer Pacific water with $S < 33$ and represented by a temperature maximum above the PWW (Steele et al., 2004).

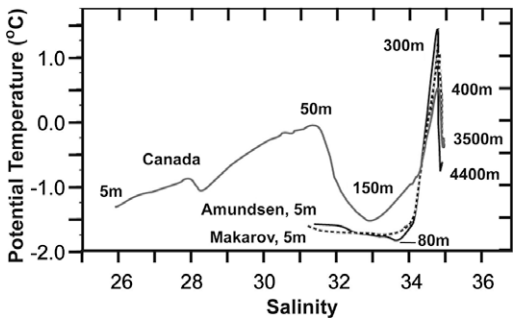

Fig. 3. The relationship between potential temperature and salinity for the deep profiles from the Canada Basin (Station 3), the Amundsen Basin (Station 21) and the Makarov Basin. The Cold Halocline Layer in the Amundsen and Makarov Basins is largely defined by mixing between surface waters of the lowest salinity, the coldest waters centred around 80 m, and Atlantic water. In the Canada Basin, warmer Pacific summer water can be seen centred at 50 m, while colder Pacific winter water peaks at 150 m. The Atlantic water component is clearly defined in all three profiles by the peak at 300–400 m, as is Polar deep water near the bottom of each profile.

were collected on a transect extending northwards to the North Pole. Water samples from the Canada Basin were collected from the USCG Polar Star during August 2000 (AWS-2000), according to the sampling procedures described by Trimble et al. (2004). Sampling locations are shown in Fig. 1. Two samples were collected from Station 5, on the Canadian slope. Station 4 was located just off the shelf, while Station 3 was further away from the slope in the deep basin.

In brief, samples during both cruises were collected using large Go–Flo samplers (20 L, 30 L, or 60 L) mounted on a rosette with a conductivity–temperature–depth (CTD) instrument. Waters from 8 m were also periodically collected in the Amundsen Basin from a purpose-built continuous inflow port beneath *I/B Oden* that fed directly into the water-processing laboratory. Upon collection, water was transferred to acid-cleaned polyethylene bottles, then filtered within a few hours after collection using a peristaltic pump through acid-cleaned 142 mm diameter 0.22 μm membrane filters. The samples were acidified to pH < 2 with ultra pure HCl (Seastar®) and stored in acid-washed 10 L polyethylene containers.

2.3. Chemical separation and mass spectrometry

The analytical procedures followed those of Andersson et al. (2008) and the details are provided therein. Briefly, filtered waters were spiked for Nd using a mixed ^{150}Nd–^{147}Sm enriched spike, and Nd was co-precipitated using an Fe carrier. The REE were separated using cation exchange resin, and Nd was separated using α-HIBA (α-hydroxyisobutyric acid) solutions in a second cation exchange column, and purified further using an EiChrom® LN-resin column. About 10 ng of Nd from each sample was loaded on Re filaments and measured on a five collector Finnigan®

Table 2
Nd concentrations and isotopic compositions of Eurasian Basin waters.

Station	Depth (m)	$^{143}Nd/^{144}Nd$	$2\sigma_{mean}$	ε_{Nd} [a]	C_{Nd} [b] (pM)	Salinity (psu)	Θ[c] (°C)
Admundsen Basin							
Leg II Station 20, 4410 m	8	0.512088	±0.000020	−10.7 ± 0.4	37.3	32.63	−1.66
88°16.70′N 82°54.31′E							
22-July-2001							
Leg II Station 21, 4400 m	8	0.512072	±0.000032	−11.0 ± 0.4	34.2	31.91	−1.59
88°24.48′N 95°22.78′E	75	0.512174	±0.000024	−9.1 ± 0.4	27.7	33.74	−1.81
22-July-2001	300	0.512074	±0.000035	−11.0 ± 0.4	17.3	34.87	1.31
	500	0.512042	±0.000016	−11.6 ± 0.4	16.0	34.87	0.53
	3900	0.512008	±0.000031	−12.3 ± 0.4	18.2	34.93	−0.95
Leg II Station 22, 4372 m	8	0.512046	±0.000013	−11.5 ± 0.4	36.7	–	–
88°26.05′N 109°50.65′E	10	0.512051	±0.000021	−11.5 ± 0.4	58.0	31.35	−1.55
23-July-2001	20	0.512157	±0.000020	−9.4 ± 0.4	24.3	31.51	−1.57
	40	0.5120368	±0.00001	−11.7 ± 0.4	53.7	33.52	−1.82
	60	0.512186	±0.000026	−8.8 ± 0.4	27.3	33.66	−1.83
	100	0.512107	±0.000018	−10.4 ± 0.4	22.5	34.18	−1.58
Leg II Station 24, 2779 m	8	0.512063	±0.000016	−11.2 ± 0.4	50.5	31.10	−1.53
88°21.85′N 126°29.76′E							
23-July-2001							
Leg II Station 26, 1472 m	8	0.512060	±0.000016	−11.3 ± 0.4	51.0	30.85	−1.53
88°08.43′N 132°33.24′E							
23-July-2001							
Leg II Station 28, 1513 m	8	0.512060	±0.000038	−11.3 ± 0.4	48.0	30.90	−1.57
87°41.93′N 148°27.86′E							
28-July-2001							
Leg II North Pole	8	0.512079	±0.000025	−10.9 ± 0.4	48.8	31.07	−1.60
Makarov Basin							
Leg II Station Makarov,	8	0.512090	±0.000069	−10.7 ± 0.4	48.7	31.23	−1.60
3985 m	10	0.512027	±0.000024	−11.9 ± 0.4	48.4	31.32	−1.64
87°54.97′N 154°22.50′E	30	0.512081	±0.000028	−10.9 ± 0.4	47.4	31.33	−1.64
26-July-2001	75	0.512101	±0.000036	−10.5 ± 0.4	30.6	33.62	−1.71
	500	0.512079	±0.000018	−10.9 ± 0.4	16.3	34.87	0.49
	1000	0.512100	±0.000023	−10.5 ± 0.4	15.3	34.89	−0.18
	2500	0.512101	±0.000026	−10.5 ± 0.4	15.9	34.95	−0.52
	3500	0.512091	±0.000018	−10.7 ± 0.4	17.0	34.96	−0.54

[a] $\varepsilon_{Nd} = [(143Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{CHUR} - 1] \times 10^4$; where $(^{143}Nd/^{144}Nd)_{CHUR} = 0.512638$. Error includes external reproducibility.
[b] Uncertainty is estimated to be <5%.
[c] Potential temperature.

MAT261 thermal ionisation mass spectrometer (TIMS) in multi-dynamic mode. During sample heating oxygen is introduced to generate a NdO^+ beam, which yields a significantly higher signal compared to that of Nd^+ ions. Possible interferences from Pr, Sm, and Ce were monitored, and no significant corrections to the data were required. After interference corrections, oxide ratios were corrected using $^{17}O/^{16}O = 0.000387$ and $^{18}O/^{16}O = 0.002110$. The results were normalised to $^{146}Nd/^{144}Nd = 0.7219$ assuming an exponential fractionation law, and were used to determine both Nd concentrations and isotope compositions.

The reproducibility and precision was determined by repeated analysis of two international standards. The Caltech Ndβ standard yielded $^{143}Nd/^{144}Nd = 0.511898 \pm 0.000018$ (2σ, n = 12), which is equivalent to $\varepsilon_{Nd} = -14.44 \pm 0.35$ using $^{143}Nd/^{144}Nd_{CHUR} = 0.512638$. The La Jolla Nd standard yielded $^{143}Nd/^{144}Nd = 0.511853 \pm 0.000023$ (2σ, n = 10), equivalent to $\varepsilon_{Nd} = -15.31 \pm 0.41$. No other corrections were applied to the measured ratios and the overall reproducibility for the samples in this study estimated from the standards is ±0.4 ε-units. The internal error was always smaller, and so this uncertainty is used for all isotope data in this study. The total Nd blank, including chemical treatment, separation, and mass spectrometry, is 40 pg. The uncertainties in the Nd concentration results of the filtered waters is estimated to be <5%.

3. RESULTS

3.1. The Canada Basin

Data for the Canada Basin are shown in Table 1, and the salinity and temperature structure of the three stations are shown in Fig. 2.The PML occupies the upper 10–15 m

of the profiles. The halocline extends to ~300 m, with relatively warm Pacific water (Steele et al., 2004) underlain by cold Pacific winter water (Jones and Anderson, 1986) centred at 150 m (Fig. 3). There are significant differences between the profiles above ~60 m. Station 3 exhibits the lowest salinity at each depth. The peak of warm Pacific water is at ~50 m in this profile (Fig. 3), while in Station 4 it is somewhat higher, at ~30 m. The highest salinities at each depth were found in Station 5 on the shelf, where the warmest waters were within the upper 30 m, with substantial variations in temperature over short depth intervals unaccompanied by comparable salinity variations. The underlying warm Atlantic water layer is clearly marked by a prominent temperature maximum, with a peak at ~400 m. Deeper waters with potential temperatures less than 0 °C are found below~1000 m.

Canada Basin Nd data are shown in Fig. 4 and Table 1. The surface waters of the Polar mixed layer contain the highest Nd concentrations, with a decrease within 5 m waters with distance from the coast from 32 pM at Station 5, to 27 pM at Station 4, and to 25 pM at Station 3. Nd isotope compositions in these waters vary from $\varepsilon_{Nd} = -8.0 \pm 0.4$ at Station 5, up somewhat to $\varepsilon_{Nd} = -7.4 \pm 0.4$ at Station 4, and down to $\varepsilon_{Nd} = -8.7 \pm 0.4$ at Station 3 in both 5 m and 25 m samples. Pacific waters in the upper 85 m have Nd concentrations that vary over a small range of about 5%, from 25 to 26 pM at Station 3 and of about 10%, from 27.1 to 30.6 pM at Station 4, with no apparent

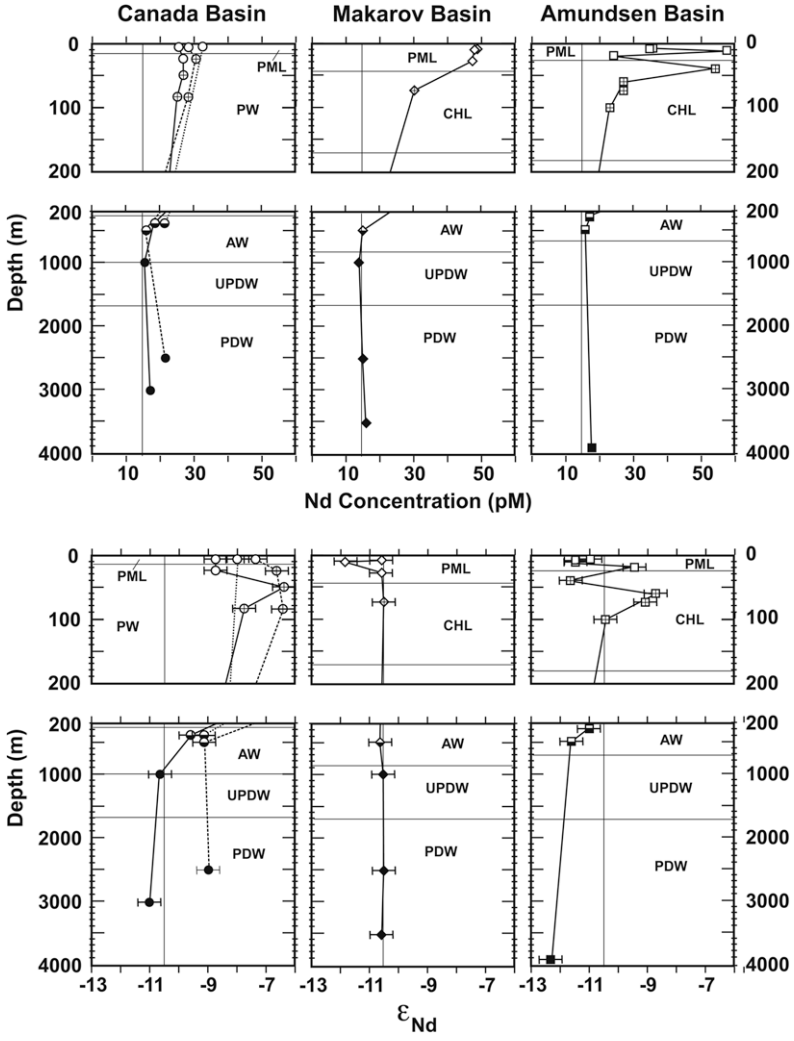

Fig. 4. Nd data for the Canada Basin (Station 3, solid lines; Station 4, dashed lines; Station 5, dotted lines), the Amundsen Basin (Stations 21 and 22), and the Makarov Basin. For comparison, the characteristics of the Atlantic inflow water (Andersson et al., 2008) are shown by vertical lines. A common pattern is evident for the three basins. Shallow waters of the PML and the CHL (open symbols) and Pacific water (crossed symbols) are enriched in Nd, with a range of isotope compositions that reflect variable inputs from rivers and shelves. Atlantic waters (half-filled symbols) generally have lower Nd concentrations, with ε_{Nd} values that vary around the Atlantic inflow values. Nd concentrations in deep waters (filled symbols) are generally lower, though above that of the Atlantic inflow. The deepest water in the Amundsen Basin has a distinctly lower isotopic ratio than the Canada and Makarov Basins, indicating at least one distinct source ventilating this basin. Deeper waters in the Canada Basin have concentrations and isotope compositions that decrease from Stations 4–5, and so with distance from the continental slope.

relationship with salinity. There are clearly resolvable isotopic variations over this interval, with values of $\varepsilon_{Nd} = -6.4 \pm 0.4$ to -7.8 ± 0.4 that do not correlate with salinity or temperature, so that there does not appear to be any difference in Nd characteristics between Pacific winter and summer waters.

Nd characteristics of Atlantic waters from 400 m depth at Stations 3 and 5 are similar, with Nd concentrations of 19 and 21 pM, while Atlantic water at 500 m in Station 4 is somewhat lower, at 16.1 pM. The isotopic compositions were similar, with $\varepsilon_{Nd} = -9.1 \pm 0.4$ to -9.6 ± 0.4 across all three stations. Deeper waters at Station 3 have 16 pM (1000 m) and 17 pM (3000 m), with similar values of $\varepsilon_{Nd} = -10.7 \pm 0.4$ and $\varepsilon_{Nd} = -11.0 \pm 0.4$, respectively, while water from 2500 m at Station 4 had a substantially higher concentration of 22 pM, and with a significantly higher value of $\varepsilon_{Nd} = -9.0 \pm 0.4$.

3.2. The Amundsen Basin

The salinity and temperature profiles for Stations 21 and 22 are shown in Fig. 2. The Polar mixed layer extends to a depth of ~25 m at Station 22, and is underlain by somewhat colder waters centred at about 80 m in the upper halocline (Fig. 3). Temperatures of >0 °C, from 170–820 m, comprise the underlying warmer Atlantic water. At Station 21, the Polar mixed layer is less well defined, and the Atlantic layer is somewhat broader.

Surface waters were collected from a depth of 8 m at seven locations (Fig. 1) on a transect across the Amundsen Basin (Fig. 5). Nd concentrations vary substantially, with values of 34–37 pM at the first three stations in the transect where salinities are 31.35–32.63, and values of 48–51 pM at the other four locations further in the central Arctic Basin where lower salinities of 30.85–31.10 were found. In contrast, the isotopic compositions (Table 2) were essentially indistinguishable from one another across the transect, with $\varepsilon_{Nd} = -10.7 \pm 0.4$ to -11.5 ± 0.4.

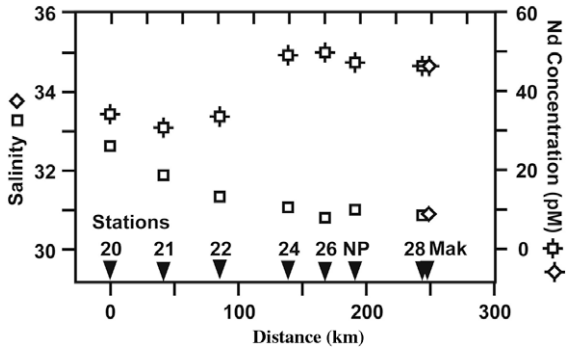

Fig. 5. Nd concentrations of surface waters collected from a depth of 8 m on a transect (Fig. 1) across the Amundsen Basin and into the Makarov Basin (plotted against distance from Station 20) vary substantially, with values of 34.2–37.3 pM at the first three stations in the transect where salinities are 31.35–32.63, and values of 48.0–51.0 pM at the other four locations further in the central Arctic Basin where lower salinities of 30.85–31.10 were found. In contrast, the isotopic compositions (Table 2) were essentially indistinguishable from one another across the transect.

Water samples were collected over closely-spaced intervals in the upper 100 m of Station 22. The two samples from near the surface at 8 and 10 m, and so within the Polar mixed layer, have identical isotope compositions of $\varepsilon_{Nd} = -11.5$, but remarkably, the 10 m sample has ~60% more Nd, with 58 pM compared to 37 pM at 8 m. Water collected at 60 m, and water from 75 m at Station 21, have $\varepsilon_{Nd} = -8.8 \pm 0.4$ and -9.1 ± 0.4, and concentrations of 27 and 28 pM, respectively. Both of these waters were from around the temperature minimum. However, water from 20 m, still within the Polar mixed layer, has $\varepsilon_{Nd} = -9.4 \pm 0.4$, with 24 pM Nd, while water from 40 m, within the underlying CHL, has $\varepsilon_{Nd} = -11.7 \pm 0.4$ and 54 pM Nd. Therefore, Nd isotopic compositions and concentrations do not consistently correlate with salinity or temperature in this section of the profile. In contrast, there are only minor fluctuations in the potential temperature–salinity relationships over this depth interval.

Arctic deep water collected in the Amundsen Basin at Station 21 from a depth of 3900 m has $\varepsilon_{Nd} = -12.3 \pm 0.4$, with 18 pM Nd. Atlantic water sampled at 300 m at Station 21 has $\varepsilon_{Nd} = -11.0 \pm 0.4$, with 17 pM Nd. Water from 500 m, within the underlying thermocline, has $\varepsilon_{Nd} = -11.6 \pm 0.4$, which is a value between the Atlantic water and Arctic deep water and so is consistent with mixing between these sources (Fig. 3). This is accompanied by a concentration of 16 pM Nd, lower than the overlying and underlying waters, suggesting either persistent heterogeneities in Nd concentrations or some vertical transport of Nd.

3.3. The Makarov Basin

The salinity and temperature profile for the Makarov profile is shown in Fig. 2, and Θ–S relationships are shown in Fig. 3. The Polar mixed layer, in the top 20 m, is underlain by a less pronounced cold water layer, and an Atlantic water layer at similar depths to that seen in the Amundsen, although slightly cooler. Waters throughout the profile generally have uniform Nd isotope compositions (Fig. 4) of $\varepsilon_{Nd} = -10.5 \pm 0.4$ to -10.9 ± 0.4, with Nd concentrations that are highest at 8 m (49 pM), progressively decreasing to 15 pM at 1000 m, and slightly higher values in deeper waters, with 16 pM at 2500 m and 17 pM at 3500 m. The exception to these trends is the sample from 10 m, which has a significantly lower value of ε_{Nd} (-11.9 ± 0.4) but a similar Nd concentration (48 pM) to the 8 m sample.

3.4. Arctic Rivers

Arctic River water data are shown in Table 3. Overall, rivers entering the Arctic have a wide range of Nd concentrations, reaching to over 10^2 times that of typical seawater, and Nd isotope ratios that are both higher and lower than the most extreme values for seawater measured here. The Lena River was found to be dominated by Nd eroded from old terrains, with $\varepsilon_{Nd} = -13.6 \pm 0.4$ and 477 pM Nd. This is indistinguishable from the value of $\varepsilon_{Nd} = -14.2 \pm 0.3$ obtained by Zimmermann et al. (2009), although the

Table 3
Nd concentrations and isotopic compositions of Arctic Rivers.

River	Sampling date	$^{143}Nd/^{144}Nd$	$2\sigma_{mean}$	ε_{Nd} [a]	C_{Nd} [b] (pM)	Mean discharge ($km^3\ yr^{-1}$)
Kolyma	06-Sept-04	0.512328	±0.000027	−6.0 ± 0.4	129	74[d]
Lena	24-Aug-04	0.511941	±0.000016	−13.6 ± 0.4	477	525 [d]
Lena[c]				−14.2 ± 0.3	826	525 [d]
Yenisey[c]				−5.2 ± 0.3	154	586 [d]
Ob[c]				−6.1 ± 0.3	2152	403 [d]
McKenzie[c]				−12.9 ± 0.3	111	284[e]

All data is for filtered waters.
[a] $\varepsilon_{Nd} = [(^{143}Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{CHUR} - 1] \times 10^4$; where $(^{143}Nd/^{144}Nd)_{CHUR} = 0.512638$. Error includes external reproducibility.
[b] Uncertainty is estimated to be <5%.
[c] Data from Zimmermann et al., 2009. Analysis by MC–ICP–MS.
[d] Semiletov et al., 2000.
[e] Woo and Thorne, 2003.

concentration was almost twice as high, with 826 pM, likely due to the seasonal variations. Sediments from near the river mouth were found to also have low Nd isotope ratios, with an average of $\varepsilon_{Nd} = -12$ (Guo et al., 2004) that was also similar to sediments from the Khatanga River (to the west of the Lena) and the Indigirka River (to the east). The Kolyma River, much further to the east, had $\varepsilon_{Nd} = -6.0 \pm 0.4$ and 129 pM. Other available data include those for the relatively radiogenic Yenisey ($\varepsilon_{Nd} = -5.2 \pm 0.3$, with 154 pM) and Ob ($\varepsilon_{Nd} = -6.1 \pm 0.3$, with the highest Arctic River concentration of 2152 pM). Sediments from near the mouths of these rivers were found have similarly high ε_{Nd} values, with an average of $\varepsilon_{Nd} = -7.2$ (Guo et al., 2004). The Mackenzie River, draining into the Canada Basin, is quite unradiogenic, with $\varepsilon_{Nd} = -12.9$ and a low concentration of 111 pM.

4. DISCUSSION

4.1. Nd and Arctic water masses

Overall, there are four main contributors of Nd to the Arctic to consider, and the Nd concentrations and isotopic compositions reflect the complexities of different source waters at different depths in the profiles, as discussed below. North Atlantic water is the main source of water, and so is expected to dominate the Nd budget of the deep waters, as well as of the warm Atlantic layer. The characteristics of Atlantic water flowing into the Arctic, with ~16 pM and $\varepsilon_{Nd} = -10.8$ (Andersson et al., 2008) in the main inflow, therefore provide a benchmark for evaluating the influence of other components. It has been found (Andersson et al., 2008) that the Nd isotopic compositions throughout the water column of the Nansen Basin (with an average of $\varepsilon_{Nd} = -11.0$) are indistinguishable from Atlantic inflow waters, although Nd concentrations (of 16–18 pM) are somewhat higher, indicating another source of Nd. The isotopic compositions of the waters from the Makarov Basin are also largely indistinguishable from Atlantic waters, and Nd concentrations are also greater. Pacific water is seen in the Canada Basin and provides more radiogenic Nd to the Arctic. Rivers provide Nd into shallow, low salinity waters, with isotopic compositions that may be higher or lower than that of the Atlantic waters and so imparting distinct signatures to surface waters, as seen in some samples from the Amundsen and Makarov Basins. A fourth source of Nd is the continental shelves. Since the source of the Nd is likely to be the sediments from the regions drained by nearby rivers, the isotopic compositions might be expected to be related to riverine Nd (mixed with seawater Nd), although the Nd concentration imparted onto waters is more difficult to constrain. As discussed below, this source has an influence on Nd in deeper waters.

A distinctive feature of all of the sampling profiles is that surface waters do not have lower concentrations of Nd than deeper waters, in contrast to the general relative surface depletion of Nd in other major ocean basins (Fig. 6; see review by Goldstein and Hemming, 2003). This has been reported previously by Westerlund and Ohman (1992) for a profile in the Nansen Basin (see also Andersson et al., 2008), and now appears to be a common feature of the Arctic.

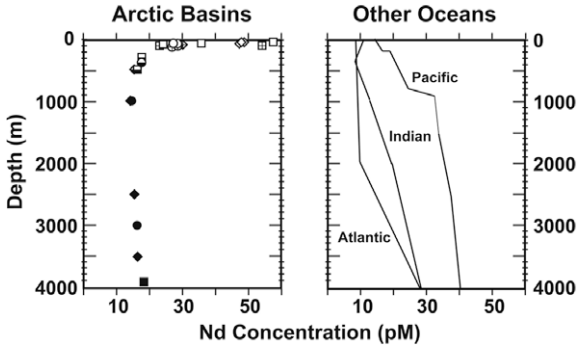

Fig. 6. Nd concentration profiles for the Canada, Makarov, and Amundsen Basins (symbols as in Fig. 4) indicate that a common feature of Nd concentration profiles across the Arctic is elevated Nd concentrations in near-surface waters. This is in contrast to the pattern typically seen elsewhere, as shown by typical profiles from the Pacific (Amakawa et al., 2004), Indian (German and Elderfield, 1990), and Atlantic (Piepgras and Wasserburg, 1988) oceans. This largely reflects the inputs of Nd to near-surface waters within the Arctic Basins.

4.2. Pacific waters and radiogenic Nd

Waters from around the Pacific layer in the Canada Basin have higher ε_{Nd} values and substantially higher concentrations than deep waters or Atlantic inflow water. Pacific waters have been shown to have radiogenic Nd due to the inputs from young circum-Pacific volcanics (Piepgras and Jacobsen, 1988; Amakawa et al., 2004), although data for the northern Pacific is very limited. Shallow waters (<400 m) at a site south of Kamchatka Peninsula (47°N, 161°W) had ε_{Nd} values of −0.1 to −2.1, with concentrations of 16–23 pM (Piepgras and Jacobsen, 1988). Further east, and south of the Bering Sea (45°N, 177°W), a composite water from the upper 500 m had $\varepsilon_{Nd} = -7.7$, with concentrations of ~12 pM (Shimizu et al., 1994). However, Pacific waters emerging from the Chukchi Sea have concentrations of up to ~30 pM and ε_{Nd} values of ~ −5.5 (Dahlqvist et al., 2007), and were thus apparently modified by interactions with the underlying sediments on the shelf. The Pacific layer waters fall in the region of this composition (Fig. 7), with values of $\varepsilon_{Nd} = -6.4$ (with 26 pM) and −6.7 (with 31 pM) at Station 3 (50 m) and Station 4 (25 m), respectively. However, these waters do have slightly lower salinities than the inflow salinities reported by Dahlqvist et al. (2007), due to the seasonal variations in Pacific water inflows. Unfortunately, no data is available for Pacific winter water. Water from Station 3, 85 m, with a temperature midway between the summer and the winter temperature extremes in the profile (Fig. 3), has a somewhat less radiogenic value of $\varepsilon_{Nd} = -7.8 \pm 0.4$ and a somewhat lower concentration of 25.1, suggesting significantly more radiogenic Nd at lower concentration in the winter water, though water from Station 4, 85 m, with a similar mixture of the two water components based on temperature and salinity (Fig. 2) has a value of $\varepsilon_{Nd} = -6.4 \pm 0.4$, suggesting much less difference in the ε_{Nd} value of the two components.

The relationship between Nd concentrations, ε_{Nd} values, and salinities are shown in Fig. 7. Pacific waters within the Canada Basin profiles generally fall on a mixing line for ε_{Nd} and Nd concentrations between Atlantic inflow waters and the composition of Pacific water flowing into the Arctic (Dahlqvist et al., 2007), although some contribution from waters with MacKenzie River characteristics of relatively high Nd concentration and unradiogenic Nd can be accommodated, especially considering that there may be considerable variation in the composition of the Pacific inflow. Such river inputs are clearer in the relationships between Nd concentrations and salinities (Fig. 7), where Pacific waters in the Canada Basin profiles are less saline than mixtures of Pacific and Atlantic inflow waters.

Further data are necessary to determine whether there are significant variations in Pacific inflow Nd characteristics. Since the Pacific water inflow to the Arctic is very clearly reflected in the Nd isotope distribution, this signal might prove useful for documenting changes in Pacific inflow and so contributions to the halocline across the Arctic (Jones and Anderson, 1986) during climate and sea level changes if suitable archives of shallow depths are available.

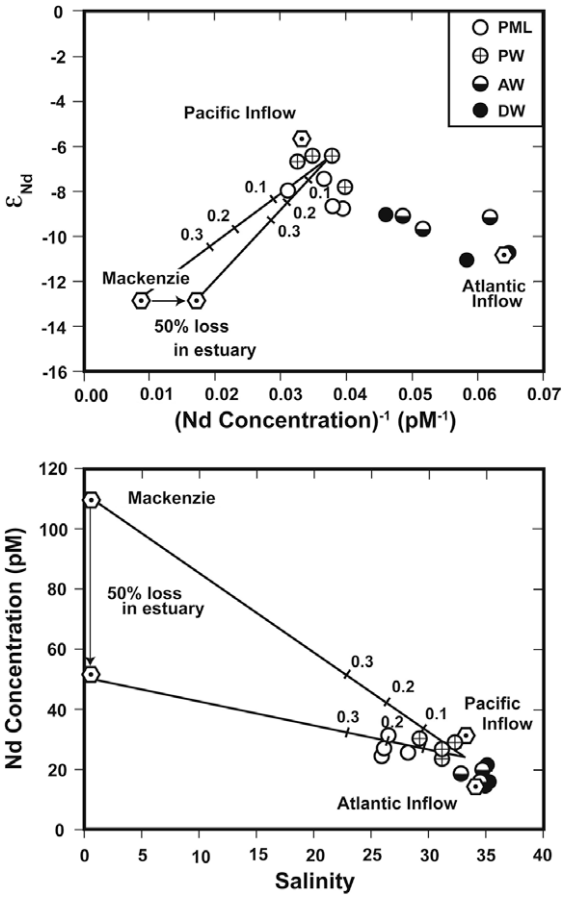

Fig. 7. Waters in the Canada Basin typically have Nd concentrations and ε_{Nd} values (upper panel) that are higher than Atlantic inflow waters (Andersson et al., 2008). Pacific waters have Nd characteristics that can be explained by addition to Pacific inflow water (Dahlqvist et al., 2007) of a small proportion of Atlantic water; somewhat reduced salinities (lower panel) indicate inputs from river waters. PML waters are displaced towards lower ε_{Nd} values than in underlying Pacific waters, consistent with mixing with ~2–10% Mackenzie water. These waters also have lower salinities (lower panel), consistent with a riverine input (Andersen et al., 2007), but are consistent with mixing with a Mackenzie River input that has been reduced in Nd concentration by at least 50%, presumably by estuarine losses. The required fraction of river water is then correspondingly increased to ~5–20%.

4.3. Sources of shallow waters and mixing

The relationships between Nd concentrations and isotope compositions for PML waters of the Canada Basin are also shown in Fig. 7. Like underlying Pacific inflow waters, PML waters have Nd concentrations between those of the Pacific and Atlantic inflow waters, and mixing between these waters is consistent with T and S data (Fig. 3). However, the Nd data are clearly displaced from a mixing line between these waters towards lower ε_{Nd} values. This can be explained by mixing with waters from the nearby Mackenzie River. Although measurements of Nd across the estuary are not available, mixing relationships observed between river and seawater components

further out in the basin can identify the concentration of the river component that results from the net result of estuarine processes that modify Nd water concentrations. A representative mixing line is shown in Fig. 7, with mixing of ~2–8% Nd from Mackenzie River water with seawater inflows explaining the range of shallow water compositions. The spread of data away from a single line can be explained by differences in the ratio of Pacific to Atlantic inflow waters. The lower salinities of these shallow waters are lower due largely to riverine inputs rather than meltwater, as deduced from O isotopes (Andersen et al., 2007). However, mixing of these same components is not compatible with the relationship between Nd concentration and salinity (Fig. 7), where the shallow waters appear to have lower Nd concentrations than might be expected based on salinity. These waters do lie around lines representing mixing between a mixture of Pacific and Atlantic inflow water and a Mackenzie River component that had a Nd concentration that is at least ~50% lower, and so it appears that a considerable fraction of Nd is lost in the Mackenzie estuary. The fraction of Nd from river water in each shallow water sample would then be correspondingly greater, of ~4–16% (Fig. 7). As an interesting comparison, an earlier study used data from the same samples as those in this study to conclude that Mackenzie River water had also lost ~65% of its U prior to mixing into the ocean basin (Andersen et al., 2007), so it appears that U and Nd are removed to similar extents in the Mackenzie estuary, presumably due to the scavenging by flocculating Fe and humic acids at low salinities (e.g. Andersson et al., 2001).

As shown in Fig. 5, there is a strong widespread Nd enrichment in the surface waters across the rest of the Arctic. Salinities are also lower in these waters; O isotope data for some of these samples indicate that melt water can only account for a small fraction of the freshwater (Andersen et al., 2007), leaving river water as the cause of the decreased salinity. Nd isotope characteristics of Nd in all samples from the Amundsen and Makaraov are shown in Fig. 8. All of the PML and CHL samples of the Makarov, and many of those from the Amundsen, fall on a trend extending from the Atlantic inflow towards much higher Nd concentrations, but similar ε_{Nd}, and so suggesting that the Nd in these samples are the result of mixing between Atlantic waters and a Nd-rich component with $\varepsilon_{Nd} \sim -11.5$. The input of freshwater from the Siberian Rivers has been identified in this area based upon other tracers (Anderson et al., 2004). While none of the individual rivers measured in this study have the required ε_{Nd} value, a mixture containing water from the Lena and either 12% water from the Ob (producing a mixture with 830 pM Nd) or 62% water from the Yenisey (producing a mixture with 340 pM Nd) could provide a suitable endmember component with $\varepsilon_{Nd} \sim -11.5$. However, it is the Lena–Yenisey component that is compatible with the relationship between the salinity and Nd concentrations of the samples (Fig. 8). Between 5 and 10% of this river water mixture would be required. Similar contributions of river water to the shallow waters and halocline of the region have been documented using O isotopes (Schlosser et al., 2000). Of course, other mixtures involving as yet unmeasured river waters may also yield a

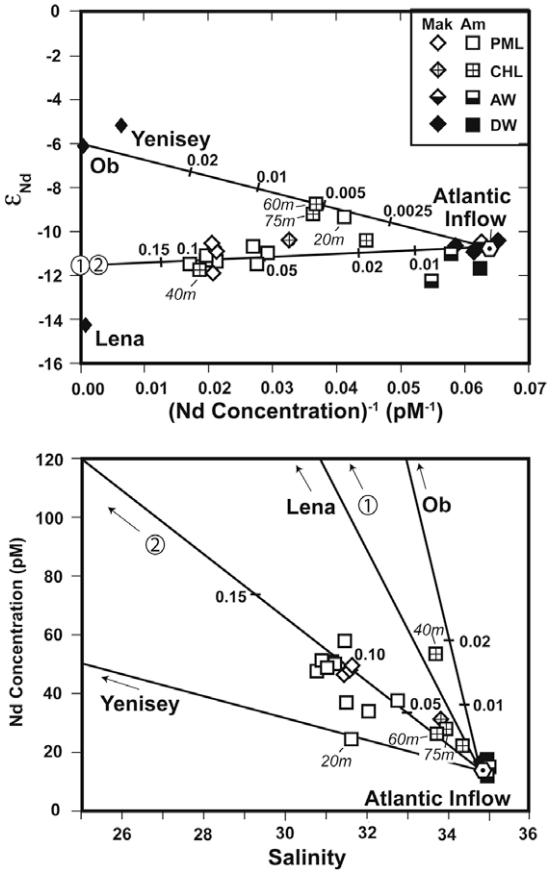

Fig. 8. Waters from the Amundsen and Makarov Basins generally have greater Nd concentrations than the Atlantic inflow (Andersson et al., 2008), and ε_{Nd} values that are both greater and smaller than the inflow (upper panel), indicating several different Nd sources. In the Makarov Basin, PML and CHL waters generally cluster around a mixing line between the Atlantic inflow composition and a mixture of either Lena and Ob waters (composition 1) or Lena and Yenisey waters (composition 2). The fraction of the river water composite 2 is marked along the mixing line. A similar mixing line is compatible with the Nd-salinity data (lower panel). Composite 1, with a higher Nd concentration, results in mixtures that are too saline. Some CHL and PML samples are not consistent with this mixing relationship (identified with a sampling depth) and require different river contributions.

component with the appropriate Nd composition of $\varepsilon_{Nd} \sim -11.5$ and ~400 pM Nd. However, it is clear that if a significant fraction of water from the Ob is involved, much of its Nd must be lost prior to entering the Arctic Ocean interior. An exception to these relationships is the CHL sample from Station 22 (40 m) in the Amundsen, which has a higher salinity than these other samples, and so is consistent with greater contributions of Nd from the Lena.

In the Amundsen Basin, waters from 60 m to 75 m in the Cold Halocline Layer (CHL), as well as this PML water (from Station 22, 20 m), have significantly higher ε_{Nd} values than Atlantic waters, suggesting involvement of Nd from either the Ob or Yenisey without input from Lena waters. Based upon the relationships between Nd concentration and salinity (Fig. 8), the PML water can be explained by

mixing between Atlantic waters and Yenisey River water. The CHL waters have higher salinities and are more compatible with addition of Nd from the Ob, which has a much higher Nd concentration and so will result in a much smaller concomitant decrease in salinity. However, identifying specific mixtures of river components to explain each sample outside of the main trend described above is underconstrained, especially without more comprehensive data on river inputs to the Arctic. In addition, it is possible that during formation of halocline water from Atlantic water on the shelves (Rudels et al., 1996), Nd is obtained from underlying sediments derived from river discharges. Although evidence from other tracers suggest that Pacific inflow water is also present in these waters (Jones et al., 1998; Taylor et al., 2003), it would have to comprise an unrealistically large proportion (based on *T*–*S* relationships, Fig. 3) of these samples to have a significant influence on the Nd budget. Interestingly, isotopically distinct PML and CHL samples are interleaved in the Amundsen profiles, with waters at 20 and 60 m in Station 22 and 75 m at Station 21 having an average of $\varepsilon_{Nd} = -9.1$ and 26 pM Nd, while waters at 10, 40, and 100 m in Station 22 have significantly lower ε_{Nd} values. There are no prominent indications of these different waters in the potential temperature–salinity profiles (Fig. 3). It is likely that there are significant variations in the areal distributions of riverine Nd in shallow waters as currents flow away from the Siberian shelves (Fig. 1), and sampling of the layering seen at these stations occurred before mixing destroyed the Nd concentration and isotopic differences.

An interesting comparison is possible with an earlier study of Arctic Ocean U concentrations and isotope variations (Andersen et al., 2007) It was found that the U in the same samples used in this study can be explained by mixing of Atlantic inflow water and Yenisey River water; there was no evidence for losses of riverine U in the Yenisey estuary, nor of contributions from the Lena or Ob. Nd evidence for the presence of water from the Lena therefore requires appreciable losses of its U.

Some of the scatter in the Nd data away from simple mixing lines (Figs. 7 and 8) might be due to the contributions from shelf sediments. These sediments are likely to have similar Nd isotope compositions to those of the Siberian Rivers, and so would be capable to provide a similar mixture of Nd, although without affecting salinity. Indeed, the transport of Nd across the estuaries might involve extensive removal, followed by release as sediment is transported and salinities vary (e.g. Sholkovitz and Szymczak, 2000), effects that might change dramatically between periods of ice cover and thawing (MacDonald, 2000).

The Nd evidence for the impact of Arctic River inputs on seawater can be compared to that of the Mediterranean. Tachikawa et al. (2004) found evidence for Nd derived from the Nile in the eastern part of the basin, although the riverine component has a greater Nd concentration than what could be accounted for by dissolved constituents. Therefore, it was inferred that partial dissolution of river-derived particles occurred to release additional Nd. Interestingly, surface waters here are also not all depleted, suggesting that in relatively restricted basins, higher inputs to surface waters from rivers relative to the rate of removal by scavenging have driven up the concentrations of surface waters. In contrast, in the larger ocean basins, the residence time of Nd in surface waters with respect to river inputs is much greater and so greater integrated depletion is achieved.

4.4. Waters in the Atlantic layer

Atlantic layer water is clearly seen in all three basins, based on salinity and temperature (Figs. 2 and 3). Andersson et al. (2008) found that while waters from the main, eastern branch of Atlantic inflow as sampled in the Barents Sea had Nd characteristics ($\varepsilon_{Nd} = -10.7$ and 15.5 pM) consistent with waters from the North Atlantic (Piepgras and Wasserburg, 1987), waters from the western branch sampled north of Svalbard had concentrations up to 30 pM and ε_{Nd} values as low as −12.2, suggesting that some Atlantic waters may be modified while entering the Arctic due to the water–shelf interactions. Water from the Atlantic layer at a depth of 500 m in the Makarov profile, with $\varepsilon_{Nd} = -10.9$ and 16.3 pM, has the characteristics of north Atlantic waters. In contrast, data for waters from the Atlantic layer in the Amundsen Basin include a slightly lower value of $\varepsilon_{Nd} = -11.6 \pm 0.4$ and a slightly higher concentration of 17.3 pM indicating some involvement of waters that have been modified by water–shelf interaction.

In the Canada Basin, Atlantic layer waters in the three profiles from depths of 400–500 m have $\varepsilon_{Nd} = -9.6$ to −9.1, and concentrations of 16–21 pM. A value of $\varepsilon_{Nd} = -9.1$ can be obtained by mixing of 20% Pacific inflow water and 80% Atlantic water (Fig. 7), and this mixture would yield a concentration of 19 pM. Such mixtures could plausibly account for the Atlantic water at Stations 3 and 5. This would then imply that the Atlantic water endmember has a somewhat higher temperature than seen in Atlantic water found in the other profiles (Fig. 3). However, the sample from Station 4, with a concentration (16 pM) that is not greater than Atlantic inflow water, is not consistent with mixing between measured Atlantic and Pacific inflow waters. However, it is possible that the Nd composition of the latter may vary spatially or temporally due to the variations in water–shelf interactions in the Chukchi Sea and so provide a reasonable mixing component. Alternatively, interactions with the shelf or slope within the Arctic may have modified the Nd isotope composition of the Atlantic layer water.

4.5. Deep waters of the Arctic Ocean

Although deep waters in the Arctic Ocean clearly are derived largely from Atlantic waters, there are important differences. These waters must be modified on the shelves to produce higher salinity water (Swift et al., 1983). Further, since deep waters are warmer than shelf waters at freezing, descending waters need to mix with Atlantic layer water (Aagaard et al., 1985). Such descending waters can also carry a considerable flux of constituents found in shallow waters (e.g. Anderson et al., 1999). Nd characteristics observed in deep waters that vary from those of Atlantic waters could be explained by inputs from such waters.

A characteristic feature of the deeper waters of the Arctic is a general enrichment in Nd concentration compared to Atlantic inflow waters. Waters from depths $\geqslant$1000 m have concentrations of 15–22 pM. The highest concentration is found for the deepest sample near the slope in the Canada Basin, and may represent only water that has locally been added, since it is distinctly higher than the other deep Canada Basin sample, which was collected further away from the continental shelf. The lowest value, from 1000 m in the Makarov Basin, is the only sample with a concentration below that of the Atlantic inflow and so has not had any Nd enrichment. The deepest waters in the Canada and Makarov Basins have slightly greater Nd concentrations than deep waters somewhat higher in the water column, suggesting that the formation of the densest brines ventilating Arctic deep waters may also involve somewhat greater enrichment in Nd.

While most of the Arctic deeper waters have measured Nd isotope compositions that are similar to that of the Atlantic inflow waters (Tables 1 and 2, and Andersson et al., 2008), the Amundsen Basin intermediate- and deep water samples have lower ε_{Nd} values (-11.6 to -12.3) than either Atlantic inflow waters (-10.8) or the adjacent Nansen (~-11.0) and Makarov (-10.6) basins and so require a substantial input from isotopically less radiogenic shelf waters. However, Amundsen waters, with 18 pM and $\varepsilon_{Nd} = -12.3$, cannot be the result of simple mixing or addition of Nd; Atlantic water, with a concentration of 16 pM and $\varepsilon_{Nd} = -10.8$, would require a contribution of 3 pM with a value of $\varepsilon_{Nd} = -21$, which is substantially less radiogenic than any major rivers in the Arctic (Table 3). Rather, the formation of the appropriate water composition is likely to have resulted from exchange between shelf sediments and the overlying waters. This resulted in an isotopic composition that was modified by Nd from material discharging from rivers draining the adjoining continental areas, and may be related to possible recent major deep water renewal in the Amundsen (Björk and Winsor, 2006). Such a process contrasts with the simple mixing of Nd-rich river waters with Arctic surface waters, where Nd enrichment is accompanied by a decrease in salinity; here, exchange with underlying sediments modifies the Nd water signal without evidence for bulk mixing of distinct waters. A similar process of Nd exchange was suggested to explain waters north of Svalbard with $\varepsilon_{Nd} = -12$ while waters in the Barents Sea had a value of $\varepsilon_{Nd} = -11$, indistinguishable from that of the Atlantic (Andersson et al., 2008). Such exchange has also been inferred for other ocean basins (e.g. Lacan and Jeandel, 2005b). Tachikawa et al. (2004) found similar substantial enrichments in Nd in surface waters of the Mediterranean, and that leaching of river-derived particles could provide isotopically distinct Nd to specific water masses. It is possible that shelf waters ventilating deep waters display substantial variations along the Siberian coast, reflecting differences in riverine sources, water–sediment interactions, or sediment compositions. Such differences may explain why deep waters of the Amundsen Basin are isotopically different from those of the Nansen and Makarov Basins.

The Makarov Basin has ε_{Nd} values that are similar to those of Atlantic and Nansen Basin waters, and so distinctive from those of Amundsen Basin waters. It has been argued that there is a significant flux into the Makarov Basin across a gap in the Lomonosov Ridge as deep as 2400 m (Timmermans et al., 2005). However, while the Makarov profile had values of $\varepsilon_{Nd} = -10.5 \pm 0.4$ at 1000 and 2500 m, and -10.7 ± 0.4 at 3500 m, the Amundsen profile had values of -11.6 ± 0.4 at 500 m and -12.3 ± 0.4 at 3900 m. The Nd in Makarov waters clearly matches Atlantic inflow and Nansen deep waters, and show no affinity for Amundsen Nd. More recently, studies of possible flow across the Lomonosov Ridge suggests that flow between the two basins may indeed be more limited, consistent with the Nd data (Timmermans and Garrett, 2006; Björk et al., 2007). The pronounced isotopic contrast between the two basins indicates that Nd might be used to determine when small or intermittent fluxes occur.

The bottom sample from Station 4 in the Canada Basin is also different from Atlantic inflow waters, with a higher value of $\varepsilon_{Nd} = -9.0 \pm 0.4$, along with the highest Arctic deep water concentration of 22 pM. At Station 3, further into the Canada Basin, values again decrease to $\varepsilon_{Nd} = -10.7 \pm 0.4$ at 1000 m and -11.0 ± 0.4 at 3000 m, with more typical deep water concentrations of 16–17 pM. The difference could be explained by the simple addition of Nd with $\varepsilon_{Nd} = -3.8$, though waters with such values are not observed. Rather, the similarity between the Nd isotopic composition in the Station 4 water and the shallower waters in Station 5 on the continental slope suggests that this water was recently supplied by waters that have suffered Nd exchange on the shelf. Further data mapping out Nd isotope distributions could provide clearer evidence for the sources and rates of supply of bottom water.

A tentative budget of Nd in Arctic deep water can be made from the available data. The average concentration of water >1000 m appears to be approximately 16.7 pM, corresponding to an excess over the concentration of Atlantic water of 1.2 pM that must be derived from the shelves. Using a total volume of water below 1700 m of 3.4×10^6 km^3 (Jakobsson, 2002), this corresponds to a total excess of 4.1×10^{18}pmoles. Taking average ventilation times of 250 years for the bottom waters of the Eurasian Basin and 450 years for the Canadian Basin (Schlosser et al., 1997), and so using an overall average of 350 years, this requires an input of 1.2×10^{16}pmoles per year. This can be compared to the riverine input. The water flux of 1.4×10^{15} kg/year is relatively well known, although the average Nd concentration is not well constrained. Taking 1000 pM as a likely high estimate of undiluted river water, this yields 1.4×10^{18} pmol per year discharged into the Arctic, over 10^2 times the amount needed to supply the deep waters. As discussed above, Nd that remains dissolved beyond the immediate estuaries is transported along with the freshwater into the interior basins. However, a considerable fraction of Nd may be removed during estuarine mixing to the underlying sediments. Since nominally only 1% of the riverine Nd is required, unaccompanied by the freshwater, to supply deep waters, it is possible that the Nd removed by adsorption or incorporation into flocculating material provides this amount by later interaction with shelf waters ventilating the Arctic interior. Weathering of

detrital material within the sediment to release additional Nd is not required but is still possible, as suggested elsewhere (Tachikawa et al., 2004). The added Nd component only constitutes about 10% in the Nd in the deep waters and so does not cause a significant isotopic shift, regardless of the likely average riverine value. Although this budget is uncertain due to the limited data available for characterising Nd in the Arctic deep waters and riverine inputs, additional data is unlikely to change the sense of these conclusions. This provides an interesting contrast with other locations where boundary exchange results in isotopic shifts but only minor changes in Nd concentration (e.g. Jeandel et al., 1998; Lacan and Jeandel, 2005b).

5. CONCLUSIONS

The Nd isotope and concentration data presented here and in Andersson et al. (2008) are the first data for the Arctic Ocean. A pattern of high Nd concentrations at the surface that gradually diminish with depth is seen throughout the Arctic, and is distinct from that seen in other oceans where surface waters are generally relatively depleted. A range of isotopic variations across the Arctic and within individual depth profiles reflects the different sources of waters. The dominant source of water, and so Nd, is the Atlantic Ocean. Distinct radiogenic isotope Nd signatures can be traced in Pacific water flowing into the Canada Basin and further into the Eurasian Basin. Waters from rivers draining older terrains provide very unradiogenic isotope Nd that can be traced in surface waters across much of the Eurasian Basin.

The relationships between Nd characteristics and salinity reveal that on the Arctic shelves, Nd behaviour ranges from conservative mixing between river discharge and seawater, Nd losses from the water column during estuarine mixing, and Nd addition to overlying waters from the shelf sediments. Mixing relations of shallow waters in the central Arctic indicate that the freshwaters from the Siberian Rivers have not lost significant amounts of Nd, while there have been substantial losses in the Mackenzie river estuary. Data is not available to determine the causes of the varying estuarine behaviour, although it is likely that this is due to the differences in complexing organic matter and concentrations of constituents such as Fe and humic acids that flocculate and scavenge trace elements in estuaries. Such differences may vary both spatially and seasonally across the Arctic.

A distinct feature of the Arctic is the general influence of the shelves on the Nd concentrations of waters flowing into the basins, either from the Pacific across the Chukchi Sea, or from across the extensive Siberian shelves. This results in an increase in Nd concentration in essentially all waters in the Arctic. On the shelves, interactions with underlying sediments, through processes that are not yet well understood, modify Nd concentrations without necessarily directly affecting salinity. In estuarine regions other processes modify the Nd signal of freshwater inputs into the ocean basin, and possibly also add to the Nd in the sediments that is involved in later interactions.

Deep waters in the Arctic have been enriched in Nd, apparently by addition of Nd to waters from the shelves that ventilate the deep basins. These enrichments generally have not resulted in major shifts in the isotopic compositions of the deep waters in either the Makarov or Nansen Basins (Andersson et al., 2008), although distinctive Nd signatures were found near the margin of the Canada Basin, possibly reflecting a recent local input. Also, the Amundsen has deep waters that are isotopically different from there other basins, likely due to the ventilation of shelf waters from regions with distinctive Nd isotope signatures, and this difference limits the amount of possible inflow into the Amundsen from the adjacent Makarov Basin across the Lomonosov Ridge.

The Nd isotope variations in the Arctic are likely to have been different in the past due to the climatic and tectonic changes, and these have been recorded in Arctic sediments (Winter et al., 1997; Tütken et al., 2002; Haley et al., 2008). Changes in factors such as water inflow and brine formation, sea level, and current velocities are all expected to affect water–shelf interactions and shallow water Nd signatures. The isotopic composition of water replenishing deep basin waters will also change (Haley et al., 2008) and be recorded in deep sediments. Records of the distinctive Pacific water signature will provide information on the distribution of waters entering through the Bering Sea. A better understanding of the spatial and temporal variations in Nd isotopes on the shelves as well as in the deep waters of the various Arctic basins is therefore essential for unravelling Arctic circulation in the past.

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