LIMNOLOGY
and
OCEANOGRAPHY

ASLO

Limnol. Oceanogr. 63, 2018, 2250–2265

doi: 10.1002/lno.10936

Regional variability in the atmospheric nitrogen deposition signal and its transfer to the sediment record in Greenland lakes

N. J. Anderson,[1*] **C. J. Curtis**,[2,a] **E. J. Whiteford**,[1,b] **V. J. Jones**,[2] **S. McGowan**,[3] **G. L. Simpson**,[2,c] **J. Kaiser**[4]

[1]Department of Geography, Loughborough University, Loughborough, U.K.
[2]Environmental Change Research Centre, Department of Geography, University College London, London, U.K.
[3]School of Geography, University of Nottingham, Nottingham, U.K.
[4]Centre for Ocean and Atmospheric Sciences, School of Environmental Science, University of East Anglia, Norwich, Norfolk, England.

Abstract

Disruption of the nitrogen cycle is a major component of global environmental change. $\delta^{15}N$ in lake sediments is increasingly used as a measure of reactive nitrogen input but problematically, the characteristic depleted $\delta^{15}N$ signal is not recorded at all sites. We used a regionally replicated sampling strategy along a precipitation and N-deposition gradient in SW Greenland to assess the factors determining the strength of $\delta^{15}N$ signal in lake sediment cores. Analyses of snowpack N and $\delta^{15}N$-NO_3 and water chemistry were coupled with bulk sediment $\delta^{15}N$. Study sites cover a gradient of snowpack $\delta^{15}N$ (ice sheet: $-6‰$; coast $-10‰$), atmospheric N deposition (ice sheet margin: $\sim$ 0.2 kg ha^{-1} yr^{-1}; coast: 0.4 kg ha^{-1} yr^{-1}) and limnology. Three ^{210}Pb-dated sediment cores from coastal lakes showed a decline in $\delta^{15}N$ of ca. $-1‰$ from $\sim$ 1860, reflecting the strongly depleted $\delta^{15}N$ of snowpack N, lower in-lake total N (TN) concentration ($\sim$ 300 μg N L^{-1}) and a higher TN-load. Coastal lakes have 3.7–7.1× more snowpack input of nitrate than inland sites, while for total deposition the values are 1.7–3.6× greater for lake and whole catchment deposition. At inland sites and lakes close to the ice-sheet margin, a lower atmospheric N deposition rate and larger in-lake TN pool resulted in greater reliance on N-fixation and recycling (mean sediment $\delta^{15}N$ is 0.5–2.5‰ in most inland lakes; $n = 6$). The primary control of the transfer of the atmospheric $\delta^{15}N$ deposition signal to lake sediments is the magnitude of external N inputs relative to the in-lake N-pool.

The disruption of the nitrogen cycle is now a major component of global environmental change and is an unambiguous indication of anthropogenic impact during the last 150–200 yr (i.e., the Anthropocene) (Wolfe et al. 2013; Waters et al. 2016). Industrial fixation of reactive nitrogen (Nr) plus that resulting from fossil fuel burning is now more than that fixed naturally and continues to increase rapidly (Galloway et al. 2008). The impacts of Nr on ecosystem structure and productivity are well known, largely through studies on lowland, agriculturally dominated landscapes (Smith et al. 2006). However, arctic and alpine ecosystems are nutrient poor, highly sensitive to disturbance and adapted to low N availability (Dormann and Woodin 2002). Although far removed from the main Nr sources with correspondingly low N-deposition rates (generally $\sim$ 0.5–1 kg ha^{-1} yr^{-1}; Burkhart et al. 2004; AMAP 2006), the susceptibility of these remote high latitude ecosystems to chronic long-term N-additions is not well constrained (Street et al. 2015).

Although nonvascular cryptogam species (including mosses and lichens), which are an important component of terrestrial ecosystems at high latitudes have been shown to be highly efficient at utilizing available N (Curtis et al.

*Correspondence: N.J.Anderson@lboro.ac.uk

Present address:
[a]School of Geography, Archaeology & Environmental Studies, University of the Witwatersrand, Johannesburg, South Africa

[b]School of Science and Technology, Nottingham Trent University, Nottingham, U.K.

[c]Department of Biology, Institute of Environmental Change & Society, University of Regina, Regina, Saskatchewan, Canada

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2005), it is still not fully understood how biological N retention and physiological function is affected by low and sustained N enrichment over decades. Many experiments use N additions that represent greater inputs than local atmospheric Nr deposition rates and are typically short-term (< 5 yr). Tye et al. (2005) found that up to 60% of N added experimentally (as N-NO_3^-) to snowpack on Svalbard was immobilized (mainly as organic N). However, with up to 40% of the added N "lost" to the system, there was a high possibility of N-transfer to aquatic systems. This is especially likely at high altitudes where reduced vegetation cover (e.g., fell-field) limits the ability of the terrestrial system to retain or process excess Nr. Although N deposition rates across the Arctic are low (Burkhart et al. 2004; AMAP 2006), they are sustained over decades and the sensitivity of these ecosystems to additional inputs of N above natural background rates suggests that critical loads at which ecosystem change occurs will also be low (Pardo et al. 2011; Saros et al. 2011).

The timescale of anthropogenic disruption of the N-cycle is broadly known (Galloway et al. 2004) and is confirmed by modeling exercises and ice core records (Posch et al. 2012; Wolff 2013). Records of N concentration in large ice sheets represent temporally accurate records of atmospheric N deposition at the hemispheric scale (i.e., the Greenland record) (Mayewski et al. 1986) while ice caps are more spatially variable and perhaps reflect regional variability in emissions and deposition (Goto-Azuma and Koerner 2001). At a small spatial scale (e.g., local, 1–10 km^2), it is more problematic to determine atmospheric N-loads delivered to lakes and their catchments from ice core records because of the inherent variability of retention, processing, and transfer by soils and vegetation, as well as the vagaries of hydrology, topography/morphometry, and precipitation (Seastedt et al. 2004; Catalan et al. 2009). While it is clear that sensitive arctic and alpine lakes have been affected by increased N deposition from anthropogenic sources (Wolfe et al. 2013), the exact mechanisms that give rise to observed changes in N biogeochemistry of remote lakes throughout the northern hemisphere remain undetermined and requires further work to elucidate the processes involved. The stable isotope composition of lake sediment (δ^{15}N organic matter) has been used as an indicator of N deposition loads in Arctic lakes (Holtgrieve et al. 2011) but a central problem remains why do some remote lakes record a 20th Century δ^{15}N depletion signal while others (often neighboring sites) do not? This is an important issue as δ^{15}N in lake sediments offers one of the few means of identifying a N-effect on remote lakes over historic timescales and is central to the climate-nutrient debate (Wolfe et al. 2013). Lake sediment records indicate that there is substantial community change over the last 100–150 yr in arctic lakes but the main driver is unclear: is it regional warming or long-range atmospheric deposition of nutrients (Catalan et al. 2009)?

Because of the impact of industrial N-fixation on the N-cycle, volatile N produced by industrial activity is isotopically depleted (Heaton 1986; Hastings et al. 2009); it is widely assumed that this depleted δ^{15}N-Nr signal will be transferred to and recorded in lake sediments. However, lake δ^{15}N is a function of both atmospheric inputs, catchment, and in-lake processing (Talbot 2001). Sediment records reflect these more local/regional signals of changing N inputs to aquatic ecosystems but can also be problematic to interpret due to the dynamic nature of N following incorporation in sediments (Kendall et al. 2007). Change in sediment δ^{15}N from remote lake paleorecords is now considered to be a faithful indicator of increased Nr input to lakes over the last 150 yr, although this relationship has not been demonstrated unequivocally (Holtgrieve et al. 2011). At many sites, the timing of change in the δ^{15}N record in lake sediments agrees with variation in biological indicators of ecological change (e.g., diatom community turnover, algal pigment concentration) and is broadly interpreted as evidence of increasing aquatic production. However, there are a similar number of sites where the δ^{15}N sediment record is ambiguous (Curtis and Simpson 2011; Hobbs et al. 2016) and does not correlate with historical patterns of atmospheric Nr deposition and/or ecological change. Although it has been suggested that this variability reflects interactions between Nr deposition and climate variability (Hobbs et al. 2016), in-lake processing (i.e., anoxic hypolimnia, denitrification, and trophic interactions) and/or post-depositional diagenesis, these processes are not well understood and are variable among individual sites. For example, lakes in similar geographic regions can show variable trends in the sediment δ^{15}N record (Curtis and Simpson 2011). At remote, high latitude sites, where long term Nr deposition records do not exist, there are few, if any studies, that have attempted to match contemporary δ^{15}N signals in precipitation and trace it through into the lake sediment record (Hobbs et al. 2016). The divergent lake δ^{15}N records within a region has been taken by some as an indication of a lack of an atmospheric N pollution signal at high latitudes and has therefore stymied the discussion about what is driving the ecological change (Catalan et al. 2013).

Given the problem of complex ecological responses to multiple anthropogenic stressors/drivers which operate over the same temporal scale (e.g., warming, greening, atmospheric pollution), across the Arctic, it is important to have an independent measure of changing N inputs at remote sites to allow for a nuanced assessment of what is driving ecological change. The natural environmental experimental framework provided by SW Greenland (no mid-late-20th century warming) (Mernild et al. 2014) allows some of these possible drivers to be assessed in a controlled, systematic manner. In an attempt to resolve the issue of the impact of change in Nr deposition load on remote lakes and the transfer of the depleted δ^{15}N signal from precipitation to lake sediments, we studied lakes in three groups located across a precipitation (climate) and N deposition gradient in SW Greenland. We used an integrated, regionally replicated sampling strategy across gradients of precipitation, vegetation

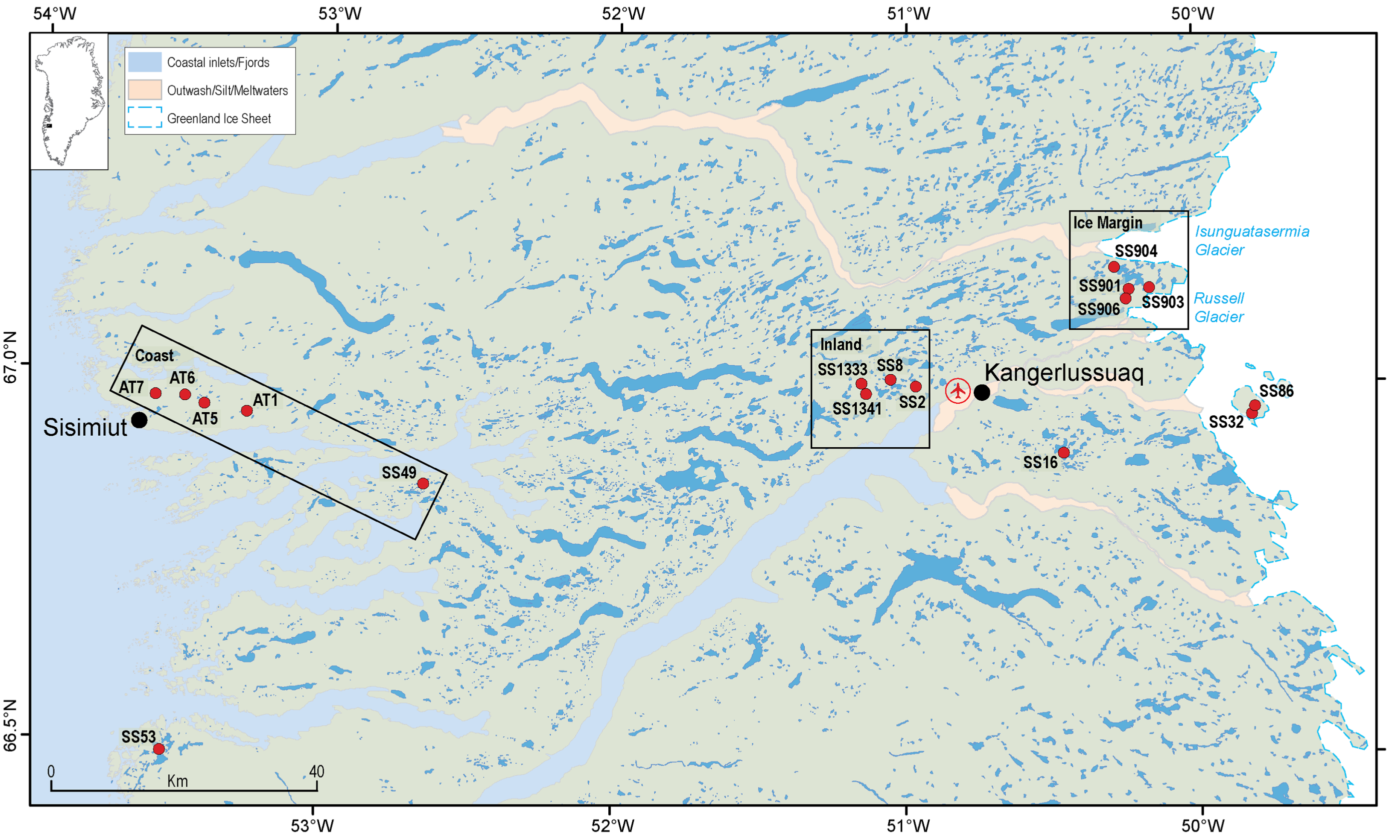

Fig. 1. Location of the study site clusters (coast, inland, and ice margin). Supplementary sites referred to in the text (SS49, SS16, SS86, SS32) are also shown. [Color figure can be viewed at wileyonlinelibrary.com]

and limnology and incorporate snow-pack chemistry, in-lake N concentration and stable isotope analyses of ^{210}Pb-dated lake sediments. Through the analysis of 12 lake-catchments located across the Kangerlussuaq region that cover differences in nitrogen isotopic composition of precipitation and N flux (Curtis et al. 2018), we tested three hypotheses: first that the δ^{15}N record of organic matter in sediment cores is a direct reflection of the input δ^{15}N signature; second, that the regional variation in bulk δ^{15}N in sediment cores and their change over time reflects differences in limnology rather than N input fluxes and third, that hydrological connectivity between lakes and their catchment is of limited relevance.

Methods

Study area

The Kangerlussuaq region between 66.5–67.2°N and 50–53.5°W forms part of the widest ice free margin in SW Greenland (Fig. 1). There is a natural climate gradient: mean annual precipitation increases from < 250 mm yr^{-1} close to the ice sheet margin to > 600 mm yr^{-1} at the coast (Mernild et al. 2015). Across the region, terrestrial vegetation is classified as dwarf shrub tundra, dominated by *Betula nana*, *Salix glauca*, *Empetrum* spp., and *Vaccinium* spp. with grasses and cryptogams also common. Close to the ice sheet margin, *Ledum palustre* heath, *S. glauca* heath, and *Carex stupina* steppe are, however, more abundant, while toward the coast, *Empetrum* spp., bryophytes, and lichens are increasingly common. The geology is relatively uniform, principally composed of granodioritic gneisses and the region is at the southernmost zone of continuous permafrost in Greenland (Nielsen 2010).

The study was based on three discrete clusters of lake catchments along a precipitation and N deposition gradient from the ice sheet margin to the coast, hereafter referred to as ice sheet, inland, and coastal sites (Fig. 1). The distances between study locations are an order of magnitude greater than the distance between lake-catchments within each location; the inland sites are ~ 60 km from the closest coastal site and 34 km from the closest ice sheet site (Fig. 1).

Study lakes and limnology

The region is a major lake district containing > 20,000 lakes. The study lakes are all small (< 40 ha), glacially scoured basins which are oligotrophic (0.45 ± 0.07 μg Chl *a* L^{-1}) and chemically dilute (< 600 μS cm^{-1}; c. 35–2000 μg TN L^{-1}; 3–30 μg total phosphorus (TP) L^{-1}; Whiteford et al. 2016) (Table 1). Reference is also made to four previously

Table 1. Annual nitrate deposition as a percentage of lake TN pool calculated using four methods with varying assumptions as outlined below. See also main text. Cmt, catchment.

				N pools and inputs (kg)					Inputs as % of lake TN pool				Sediment
Region	Lake	δ^{15}N lake snow	δ^{15}N cmt	Lake TN	Lake snow	Cmt snow	Lake deposition	Cmt deposition	Lake snow	Cmt snow	Lake deposition	Cmt deposition	^{15}N decline
Ice sheet	SS901*	−7.7	−7.5	490	0.11	2.33	1.32	17.27	0.02	0.48	0.27	3.53	No
Ice sheet	SS903	−6.4	−6.7	3095	0.40	1.89	4.18	12.87	0.01	0.06	0.14	0.42	No
Ice sheet	SS904	−9.1	−8	316	0.04	0.71	1.36	4.68	0.01	0.22	0.43	1.48	No
Inland	SS2	−3.6	−3.5	1587	0.56	3.02	2.94	17.36	0.04	0.19	0.19	1.09	No
Inland	SS8	−7.2	−5.6	765	0.18	3.92	1.17	22.24	0.02	0.51	0.15	2.91	No
Inland	SS1341*	−6.2	−5.6	364	0.10	0.56	0.56	2.98	0.03	0.15	0.15	0.82	No
Coast	AT7	−12.3	−11.8	194	0.13	6.55	0.85	26.39	0.07	3.38	0.44	13.62	No
Coast	**AT1**	**−13.1**	**−11.6**	51	0.17	1.86	1.04	6.46	**0.34**	**3.64**	**2.04**	**12.68**	**Y**
Coast	**AT6***	**−12.1**	**−11.3**	198	0.26	15.37	1.44	60.58	**0.13**	**7.76**	**0.73**	**30.62**	**Y**
Coast	**SS49***	**−12.1**	**−11.3**	196	0.49	7.98	2.69	31.46	**0.25**	**4.07**	**1.37**	**16.05**	**Y**

* No site specific snowpack data, regional means used.
SNOWPACK ESTIMATES
Lake snow: nitrate pool in lake ice snowpack as % of lake TN pool.
Catchment snow: nitrate pool in catchment snowpack as % of lake TN pool.
Direct deposition: estimated annual nitrate deposition to lake surface as % of lake TN pool.
Catchment deposition: estimated annual nitrate deposition to whole catchment as % of lake TN pool.

studies lakes in the region (SS49: coastal; SS16: inland and SS86, SS32 ice margin; Perren et al. 2009; Reuss et al. 2013). The regional climate gradient directly affects hydrological connectivity, precipitation/evaporation balance, and lake water residence times, which in turn influence limnological variation across the region (Table 1; Whiteford et al. 2016). All the lakes have a simple biological structure with higher trophic consumers limited to aquatic coleopterans and occasional predation by birds although some lakes support stickleback and Arctic Char. Ice-off generally occurs first in lakes inland: lakes in this area are typically ice-free by mid-June. Spring ice melt is typically c. 1 week later at the ice sheet margin and delayed by 3 weeks at the coast compared to inland (Whiteford et al. 2016). Study lakes were visited on three occasions during a 12 month period: July–August 2010 (25 July 2010–12 August 2010), April–May 2011 (26 April 2011–23 May 2011), and June–July 2011 (19 June 2011–29 July 2011). During each sampling occasion, all lakes were visited within a 3 week period. Methods for chemical analysis are detailed in Whiteford et al. (2016). Values presented are means of all three sampling occasions. Total N (total particulate and dissolved N) is used as a representative measure of the total in-lake N-pool.

Snowpack chemical sampling and deposition estimates

Three lake-catchments were chosen in each subregion on the basis of previous studies and their suitability for (paleo-) limnological studies. Eight replicated snowpack samples were collected within each catchment, five from contrasting sites around the catchment slopes and three from the snowpack on the frozen lake surface ($n = 24$ per region). The snowpack sampling methods and derivation of the N deposition estimates are described in Curtis et al. (2018). Briefly, they reported snowpack chemistry, NO_3^- stable isotopes and net deposition fluxes for the Kangerlussuaq region of SW Greenland. The δ^{15}N of snowpack NO_3^- shows a significant decrease from inland regions to the coast which is attributed to post-depositional processing rather than variable sources. Curtis et al. (2018) concluded that the δ^{15}N of coastal snowpack is most representative of snowfall in West Greenland. Importantly, however, post-depositional processing (photolysis, volatilization, and sublimation) lead to isotopic enrichment of the snowpack, with the greatest effects in inland areas where precipitation is low and sublimation losses higher than at the coast.

Deposition vs. lake chemistry comparisons

Without detailed catchment hydrology and chemical data, it is not possible to accurately determine the deposition input fluxes to the study lakes. At the catchment scale, there are likely to be major differences in the proportion of deposition reaching lakes through surface runoff, which is negligible in the inland lakes but could be much more important at the coast. Here, we estimate the range of possible NO_3^- and NH_4^+ deposition inputs to lakes relative to total N (TN) pools in the lakes using four methods with two contrasting assumptions. Under the first scenario, we assume that a pulsed input of snowmelt represents the key input of atmospheric NO_3^- into the system, as nitrate accumulated over the winter period is delivered in a relatively short period of intense biological activity. Although the proportion of accumulated catchment snowpack which enters the lake as meltwater is not known, we can estimate the minimum and maximum bounding values as follows: first, the minimum snowpack estimate assumes that all snowpack NO_3^- on catchment slopes is either lost by sublimation/volatilization or immobilized in catchment soils, so that only the NO_3^- in snowpack accumulated on the lake ice enters the lake as a spring melt pulse, although the proportion which percolates through candled ice into the lake rather than exiting the catchment through lateral meltwater flow over frozen lake ice is unknown. Second, the maximum bounding value for snowpack inputs to the lake would occur if the soil is frozen and biological activity is minimal, so that all snowmelt could drain into the lake basin and for closed basins, would then be delivered as the lake ice melts. In reality, losses through surface flow out of the catchment may occur, but our estimate provides the theoretical maximum input from snowpack.

Alternatively, if it is assumed that the key determinant of atmospheric NO_3^- inputs is the annual deposition flux, then we can estimate the minimum and maximum bounding fluxes in a similar way, based on direct deposition to the lake surface and deposition to the entire catchment. In this third approach, NO_3^- deposited directly to the lake surface through the whole year (rainfall and snowpack) is estimated using lake area and regional deposition flux from Curtis et al. (2018) (but ignoring the meltwater routing issue raised above). Finally and fourth, annual nitrate deposition onto the whole lake catchment provides the maximum bounding flux for atmospheric NO_3^- entering lake systems, while recognizing that a large proportion will in reality be taken up and/or immobilized in catchment soils and vegetation before reaching the lake during the summer when soils are not frozen.

Using these four estimates of inputs, the ratio of annual inputs to the lake TN pool may be expressed as a percentage to provide an index of the strength of the input signal. Lake TN pools are estimated using mean lake water TN concentration, lake surface area, and mean depth estimated as 0.5× maximum depth. Catchment and lake areas were obtained from manually digitized boundaries on topographic maps.

Sediment cores

Sediment profiles were taken in 2010 and 2011 from the deepest part of each lake using a HON-Kajak gravity corer (Renberg 1991). Intact sequences from the sediment-water interface to a depth of ~ 30 cm were recovered and were extruded on site at a resolution of 0.25 cm or 0.5 cm intervals. All samples were stored frozen until further analysis. Following standard analyses for percent dry weight and organic content (loss-on-ignition at 550°C), samples were freeze dried. Each profile was analyzed for ^{210}Pb, ^{226}Ra, ^{137}Cs, and ^{241}Am by direct gamma assay and chronologies derived using the CRS model (Appleby 2001). Details of the ^{210}Pb chronologies are provided in the Supporting Information Figs. S1–S12. Chronologies for cores from lakes SS2 and SS903 are taken from Sobek et al. (2014) while those for the four lakes which were previously analyzed for δ^{15}N, the ^{210}Pb results can be found elsewhere (Bindler et al. 2001*a,b*; Perren et al. 2009; Reuss et al. 2013).

Sediment stable isotope analyses

Subsamples of sediment were milled to a fine powder using a Retsch mixer mill. Approximately 1 mg of milled sediment was transferred to pre-weighed tin capsules, which were then sealed. The amount of dried sediment in each capsule was recorded. The samples were analyzed for TN, and ^{15}N/^{14}N on a Flash EA 1112 connected to a Thermo Finnigan Delta V isotope ratio mass spectrometer via a CONFLO IV at the Bloomsbury Isotope Facility (University College London). The isotopic ratio of ^{15}N/^{14}N is expressed using the delta (δ) notation in parts per thousand (or per mille, ‰), where $\delta^{15}N‰ = [(R_{sample}/R_{standard}] - 1] \times 1000$, where R is the ^{15}N/^{14}N ratio in the measured sample or the appropriate standard. The standard for nitrogen is the δ^{15}N of atmospheric nitrogen (commonly referred to as Air).

Results

Snowpack and deposition

Snowpack nitrate concentration varies regionally; from 1.5 μeq L^{-1} at the coast to 2.4 μeq L^{-1} at the ice margin. Snowpack depth is greatest at the coast (> 180 mm snow water equivalent [SWE]) compared to inland (< 44 mm SWE) and the ice sheet margin (36 mm SWE) (Curtis et al. 2018). This leads to inverse fluxes along the regional transect: 0.13 kg NO_3^--N ha^{-1} yr^{-1} at the coast, compared to 0.08 kg NO_3^--N ha^{-1} yr^{-1} inland and 0.11 kg NO_3^--N ha^{-1} y^{-1} at the ice sheet margin (Fig. 2). There are statistically significant regional differences in wet NH_4^+ deposition, ~ 0.24 kg N ha^{-1} yr^{-1} at the coast and 0.05 kg ha^{-1} yr^{-1} and 0.09 kg ha^{-1} yr^{-1} at the inland and ice margin lakes, respectively. As a result, the total dissolved inorganic nitrogen (DIN) flux is twofold greater at the coast compared to the ice margin (0.37 kg ha^{-1} yr^{-1} vs. 0.19 kg ha^{-1} yr^{-1}). The δ^{15}N in snowpack ranged between −7.5‰ and −5.7‰ at the inland and ice margin sites, and −11.3‰ at the coast. There are clear regional differences in the relative magnitude of snowmelt inputs and deposition fluxes compared to lake TN pools (Table 1). If the complexities of snowmelt pathways are ignored, snowpack fluxes of NO_3^- to lake surfaces at the coast are at least 3.7× greater as a proportion of lake TN than at the inland sites while the NO_3^- pool in the entire catchment snowpack relative to lake TN pools is at least 7.1× greater for coastal lakes (Table 1). For annual deposition fluxes, direct deposition to the lake surface as a proportion of lake TN pool is at least 1.7× greater at the coast, while using total deposition to the entire catchment gives ratios at least 3.6× larger for coastal lakes.

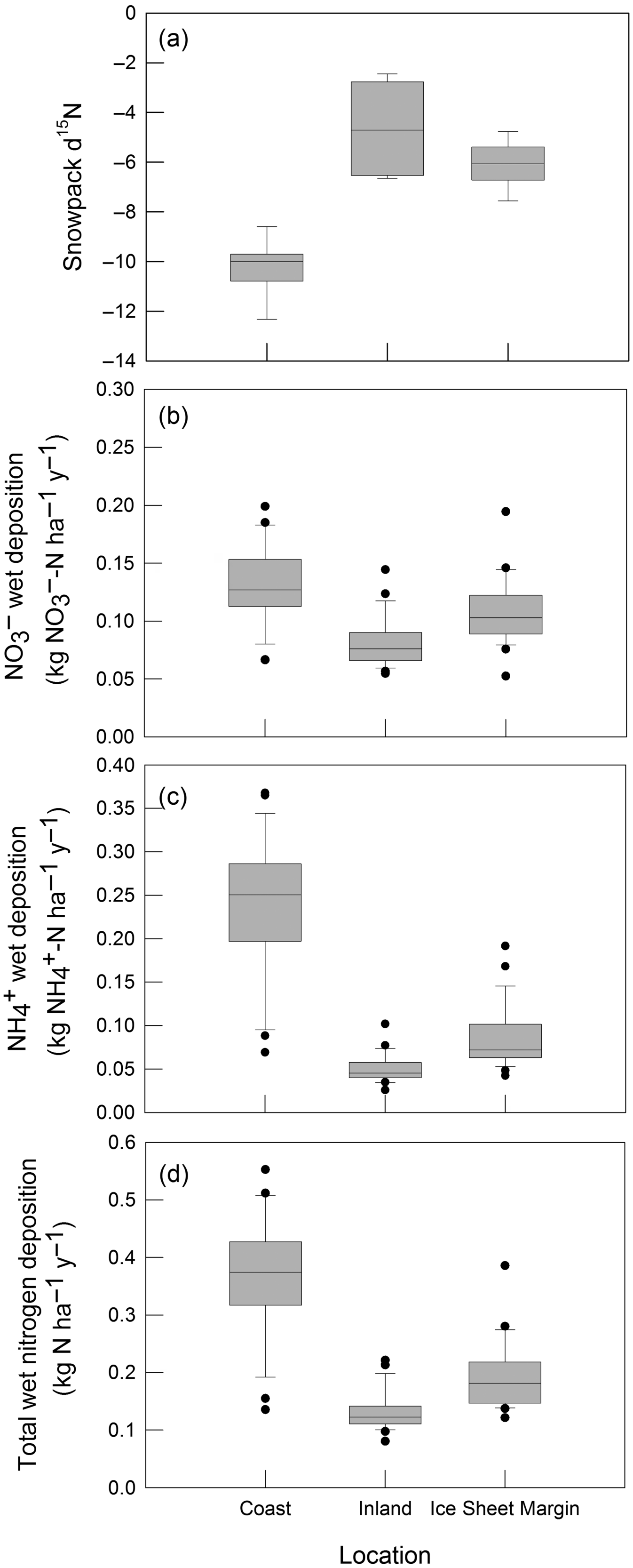

Fig. 2.

Fig. 2. Snowpack δ^{15}N and bulk N deposition (based on snowpack) for the three regions summarized as box and whisker plots; (**a**) δ^{15}N; (**b**) NO_3^--N; (**c**) NH_4^+-N; and (**d**) TN deposition, all as kg ha^{-1} yr^{-1}. The horizontal line is the median; the box represents the 25th and 75th percentiles; whiskers the 10th and 90th percentiles.

Limnology

There are strong regional differences in the water chemistry and physical limnology along the ice margin-coast transect, most notably the higher conductivity and dissolved organic carbon (DOC) concentrations observed in the inland lakes at the head of the fjord (close to Kangerlussuaq) (Table 2) (see also Anderson et al. 2001; Whiteford et al. 2016). These lakes also have the highest TN concentration, strong stratification, and hypolimnetic anoxia with increased NH_4^+ at depth. Both the coastal and ice margin lakes are oligotrophic and dilute with oxic hypolimnia, but there are statistically significant differences between these lake groups lakes in terms of pH (coastal lakes are slightly acidic), DOC and TN, which are higher inland (Table 2). While mean lake NO_3^- concentration is higher at the coast, there is a distinct seasonal pattern in DIN availability in all three lake clusters: the concentration of NO_3^- and NH_4^+ is highest under-ice and both ions are rapidly depleted with the onset of increased biological production.

Sediment dating and δ^{15}N

Sediment accumulation rates are low at all sites (generally around 0.05 cm yr^{-1} or less) (Supporting Information Table 1) but are particularly low in the coastal and ice margin lakes. The ice margin lakes all have positive δ^{15}N sediment values (1.2–1.9‰) and there are few systematic temporal trends although at SS903, δ^{15}N sediment decreased slightly from 2.3‰ to 1.9‰ (Fig. 3). Two of the three inland lakes (SS8 and SS1341) exhibit little trend in δ^{15}N over the past $\sim$ 100 yr, varying between 0.46‰ and 1.3‰ while at SS2, δ^{15}N increased in the sediment record from 1.8‰ $\sim$ 1800AD to 2.7‰ at the surface (2010 AD) (Fig. 3). Three of the coastal lakes show systematic decreases of $\sim$ 1‰ δ^{15}N from 1880–1900 AD to present, although the background values vary: AT6 decreases from 3.5‰ to 2.5‰, AT1 from 2.5‰ to 1.5‰, and SS49 from 1.4‰ to 0.5‰. There is no trend at AT7 apart from a positive increase of 0.6‰ from $\sim$ 1994 AD onward, a pattern which is also observed at AT6 (Fig. 3). The C% and δ^{13}C profiles from the cores are shown in Supporting Information Fig. S13.

Discussion

The NO_3 concentration and δ^{15}N-NO_3^- record from the Greenland Summit ice core (Hastings et al. 2009) shows a significant increase in nitrate concentration beginning in the late 1800s and early 1900s and a substantial depletion in δ^{15}N of NO_3^-, falling from a pre-industrial average of $\sim$ 11‰ to around 0‰ by the 1970s and has remained at this low level since (Fig. 4). However, of the 16 lakes that have been

Table 2. Chemical properties of study lakes. Values provided represent site-averages (± standard deviation) of three measurements taken at a depth of 2 m between August 2010 and July 2011. SS49 is from June 1999 and a single visit at SS906 was made in August 2010. Mean values in bold represent analysis of data available for all sampling occasions ($n = 5$–12).

Location	Lake	pH	Alkalinity (μeq L^{-1})	Conductivity (μS cm^{-2})	TP (μg L^{-1})	SRP (μg L^{-1})	TN (μg L^{-1})	NO_3^- (μg L^{-1})	NH_4^+ (μg L^{-1})	DIN : SRP (molar)	TN : TP (molar)	SiO_3^{2-} (μg L^{-1})	DOC (mg L^{-1})
Ice sheet	SS901	7.6 ± 0.7	1466.7 ± 1159.0	104 ± 2.8	7.9 ± 6.7	5.1 ± 2.6	563.0 ± 95.5	0	4.9 ± 8.4	2.0 ± 3.5	219.8 ± 109.3	199.7 ± 187.1	8.1 ± 0.9
Ice sheet	SS903	7.7 ± 0.5	1766.7 ± 404.1	179 ± 13.0	3.9 ± 5.0	2.0 ± 1.8	542.7 ± 194.4	0	2.8 ± 3.6	1.3 ± 0.9	247.5 ± 188.7	359.3 ± 289.3	8.0 ± 0.3
Ice sheet	SS904	7.4 ± 0.4	2166.7 ± 2626.9	77.0 ± 2.1	17.4 ± 10.3	3.9 ± 5.6	447.0 ± 142.2	0	10.4 ± 17.9	3.3 ± 4.7	63.5 ± 17.9	81.5 ± 73.7	5.7 ± 0.43
Ice sheet	SS906	7.3	1900	69.0	4.8	1.2	525.0	0	0	0	242.0	0	5.7
Ice sheet	**Mean**	**7.5 ± 0.4**	**1810 ± 1397**	**116.2 ± 49**	**9.2 ± 8.6**	**3.4 ± 3.4**	**518.3 ± 132.2**	**0**	**5.4 ± 10.2**	**2.3 ± 2.9**	**176.3 ± 121.6**	**192.2 ± 212.4**	**7.1 ± 1.3**
Inland	SS2	8.1 ± 0.5	4650.0 ± 3694.9	365 ± 31.2	7.6 ± 6.8	2.1 ± 2.5	770.3 ± 476.9	4.2 ± 7.3	6.2 ± 8.9	1.8 ± 2.6	143.6 ± 98.9	380.3 ± 277.6	26.5 ± 4.0
Inland	SS8	8.1 ± 0.8	4166.7 ± 2720.9	351 ± 51.4	8.1 ± 8.4	4.1 ± 4.3	1007.5 ± 655.8	1.1 ± 1.8	7.1 ± 12.3	3.2 ± 4.5	240.9 ± 18.7	883.0 ± 508.1	33.3 ± 10.7
Inland	SS1333	n/a	6383.3 ± 3801.4	n/a	5.3 ± 6.4	2.0 ± 1.7	1090.7 ± 242.7	0	13.5 ± 8.9	6.3 ± 0.2	420.1 ± 248.7	333.2 ± 269.1	23.9 ± 3.0
Inland	SS1341	8.1 ± 0.4	4966.7 ± 2983.9	346 ± 27.7	10.2 ± 6.4	0.2 ± 0.3	650.3 ± 365.1	1.5 ± 2.5	14.5 ± 19.4	33.2	217.9 ± 203.4	130.9 ± 219.5	18.4 ± 3.7
Inland	**Mean**	**8.1 ± 0.5**	**5041.7 ± 2969.5**	**354 ± 32.4**	**7.8 ± 6.3**	**2.1 ± 2.7**	**879.7 ± 434.4**	**1.7 ± 3.8**	**10.3 ± 11.8**	**8.0 ± 11.5**	**251.4 ± 172.7**	**431.9 ± 408.0**	**25.5 ± 7.7**
Coast	AT1	7.0 ± 0.8	266.7 ± 208.2	24.7 ± 11.8	16.9 ± 18.5	3.3 ± 1.2	93.0 ± 115.3	1.1 ± 1.8	14.3 ± 22.5	8.0 ± 12.6	6.7 ± 9.5	840.3 ± 765.6	1.3 ± 0.02
Coast	AT5	6.9	500.0 ± 565.7	37	7.2 ± 10.2	3.7 ± 1.1	184.0 ± 178.2	6.7 ± 9.5	0	5.1 ± 7.2	47.6	553.9 ± 89.2	2.2
Coast	AT6	7.0 ± 0.3	433.3 ± 288.7	47.3 ± 6.5	2.7 ± 4.6	1.7 ± 2.2	155.3 ± 104.4	19.4 ± 18.1	0.5 ± 1.0	48.1 ± 51.7	52.3	1177.3 ± 539	1.9 ± 0.1
Coast	AT7	6.7 ± 0.4	333.3 ± 208.2	41.3 ± 3.2	1.0 ± 1.7	1.4 ± 2.1	298.3 ± 269.6	82.9 ± 132.2	3.6 ± 6.2	102.3 ± 58.8	219.4	917.7 ± 506.1	1.5 ± 0.4
Coast	SS49	6.5	94	31	6.0	2.0	180.0	0	n/a	n/a	66.4	n/a	12.2
Coast	**Mean**	**6.9 ± 0.4**	**349.5 ± 271.9**	**37.1 ± 11.2**	**6.8 ± 10.8**	**2.4 ± 1.7**	**182.3 ± 162.9**	**27.0 ± 66.6**	**5.0 ± 12.1**	**34.9 ± 41.7**	**66.8 ± 78.7**	**901.2 ± 524.9**	**3.0 ± 3.8**

SRP, soluble reactive phosphorus.

analyzed for sediment δ^{15}N in the Kangerlussuaq area (this study and Sobek et al. 2014), only three coastal lakes show a decline in δ^{15}N similar to the trend recorded in the Greenland ice core record (Fig. 4). Regional replication of lake sediment δ^{15}N profiles, especially at high latitudes has been limited to date despite the ecological sensitivity of lakes in these regions to global change processes. Here, we examine the regional variability of δ^{15}N profiles in terms of key biogeochemical processes: regional variability in hydrology, in the source input of δ^{15}N (snow pack deposition) and external N load, catchment controls and, limnological variability (including in-lake processing, and post-depositional diagenesis).

Snow pack chemistry and regional N-deposition rates

There are few data on N deposition rates in the Arctic (AMAP 2006) and what are available are mainly derived from deposition models (Posch et al. 2012). Also, rates are generally assumed to be low and inconsequential for ecosystem functioning. There is, however, a paucity of data for recent atmospheric N-deposition rates across Greenland, although there have been studies of snowpack and ice core records of pollutants on the Greenland ice sheet (e.g., Dye3, 400 km distant from the present study site; Dye2, 550 km; Summit, 800 km distant) (Burkhart et al. 2006). The N deposition flux rates estimated for the Kangerlussuaq area of SW Greenland are low (< 1 kg ha^{-1} yr^{-1}) (Curtis et al. 2018) and compare with a mean derived from snow pits at Summit of 0.5 kg NO_3^- ha^{-1} yr^{-1} (0.11 kg N-NO_3^- ha^{-1} yr^{-1}; Burkhart et al., 2004); the latter are double the pre-industrial values for both wet and dry NO_3^- deposition (Fischer et al. 1998).

While the inorganic N deposition rates in this study are low, they exhibit a distinct regional pattern, reflecting broad precipitation patterns (Mernild et al. 2015) with a depleted δ^{15}N signal at the coast (Fig. 2). Higher deposition of N at the coastal sites, despite lower snowpack concentrations of NO_3^-, is due to greater precipitation at the coast, which not only increases rates of pollutant scavenging by wet deposition (Bindler et al. 2001*b*). Moreover, the high snowpack accumulation also reduces re-emission of NO_3^- to the atmosphere and minimizes the post-depositional processing which would otherwise tend to enrich the snowpack δ^{15}N signal (Curtis et al. 2018). The high concentration of ammonium in coastal snowpack is most likely to be due to marine emissions and aerosol deposition onto the snowpack, which the sea salt data show to be a critical process (Jones 1999; Pomeroy et al. 1999). While this makes a major contribution to lake total inorganic N inputs, there is no evidence from ice core records of increasing NH_4^+ deposition over the past ~ 200 yr (Savarino and Legrand 1998), hence we focus here on NO_3^- inputs and assume there has been no temporal trend in NH_4^+ deposition.

The regional gradient in snowpack δ^{15}N (Fig. 2) suggests strong fractionating processes between the coastal and inland/ice sheet margin sites. Higher levels of volatilization

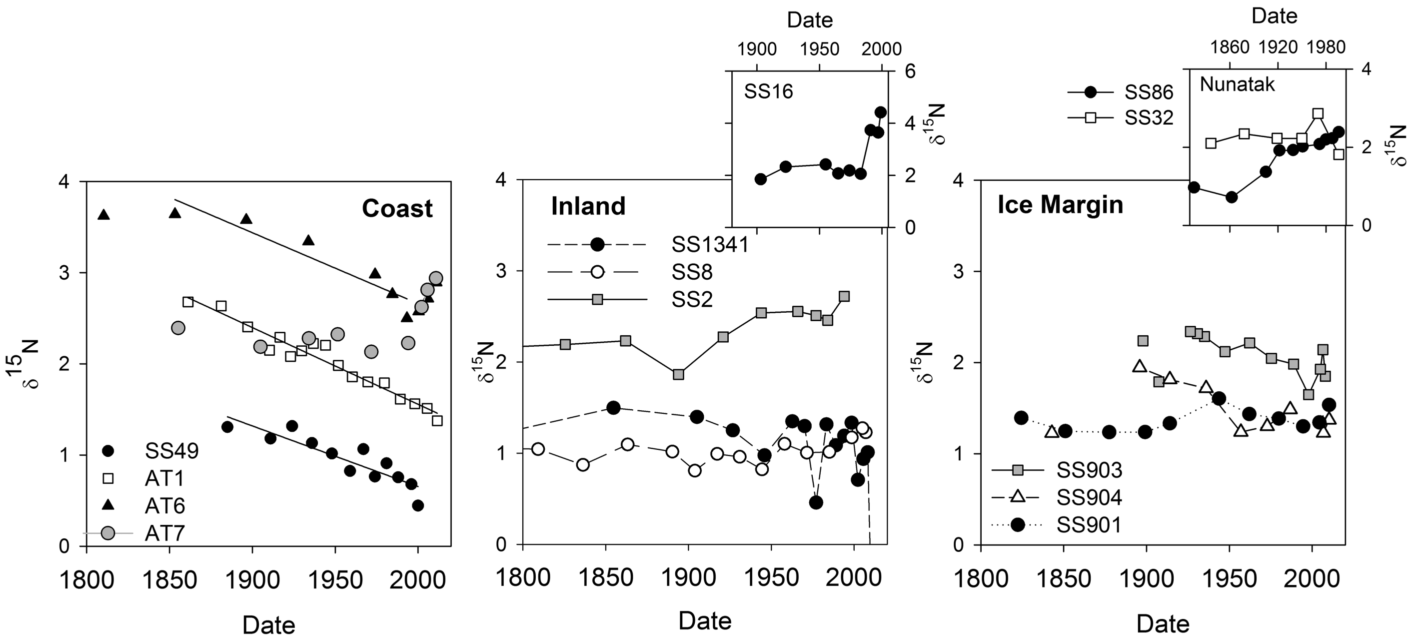

Fig. 3. Sediment core δ^{15}N vs. time (since 1850) by lake cluster. Three of the four coastal lakes show significant declines in δ^{15}N as indicated by the regression lines. The two supplementary plots (upper right) for the inland and ice margin lake clusters show δ^{15}N profiles from supporting, ancillary lakes (see main text for details).

of NO_3^- at the two inland locations with greater snow sublimation and lower precipitation may lead to enrichment of snowpack ^{15}N compared with the coastal sites. Heaton et al.(2004) and Morin et al. (2008) suggested that post-deposition processing of snowpack nitrate would lead to isotopic enrichment, so while these processes cannot account for the low coastal values, they could account for the higher inland values if it is assumed that fresh snow in all locations was deposited with a similar δ^{15}N value of c. $-11‰$. Since snow photochemistry is a major driver of NO_3^- re-emissions, the effects of post-depositional processing should be maximal in spring when UV exposure is highest and there is still snowpack present (Morin et al. 2008). The greater snowfall at the coast increases the burial rate of NO_3^- in the snowpack and hence reduces re-emission rates and photolysis. Importantly, this more depleted δ^{15}N in the snow remaining on the lake ice at the coast is more effectively transferred to the lake water as snow melts in a relatively unaltered state due to the reduced fractionation.

Sediment δ^{15}N in lakes

Nitrogen cycling in lakes is complex. The N isotope signal of bulk sediment is influenced by the primary source of organic matter, fractionation during ammonia and nitrate assimilation from DIN, N fixation (direct assimilation of atmospheric nitrogen) as well as internal recycling of the organic nitrogen-DIN pool, in particular the processes of ammonification, nitrification, and denitrification (Teranes and Bernasconi 2000; Talbot 2001).

There are two main mechanisms proposed that could cause the observed depletion in δ^{15}N in sediment cores from remote, arctic lakes; (1) increased biological utilization of DIN and (2) increased deposition of isotopically light N from emission sources of fossil fuel combustion. The first mechanism is likely to result from an increased availability of N to lake biota until P becomes limiting, at which point, discrimination between the heavy and light isotopes would begin with incomplete usage of the DIN pool and the Rayleigh distillation effect can act to increase discrimination between reactant and product (Kendall et al. 2007). This process is unlikely to be relevant in SW Greenland lakes because of their oligotrophic nature. There is a clear N : P gradient, increasing from the ice margin to the coast (Table 2), which reflects the greater P load (form dust) close to the ice sheet (Hawkings et al. 2016) and increased N-load at the coast. Despite this increased N-load (Curtis et al. 2018) which has likely alleviated nutrient stress of primary producers to some extent, the coastal lakes remain nutrient poor (see next section). Regionally, all lakes are very nutrient poor and are N + P co-limited with all available DIN utilized very quickly by in-lake biological processes. Phytoplankton growth yield is greater with addition of both N and P (Whiteford unpubl.), supporting the assumption that algal discrimination of N is not a dominant process.

The second mechanism is one of isotope mixing, with an increased amount of "light" ^{14}N acting to reduce the overall isotopic values of the DIN pool in lakes. Holtgrieve et al. (2011) hypothesized that the switch to isotopically lighter

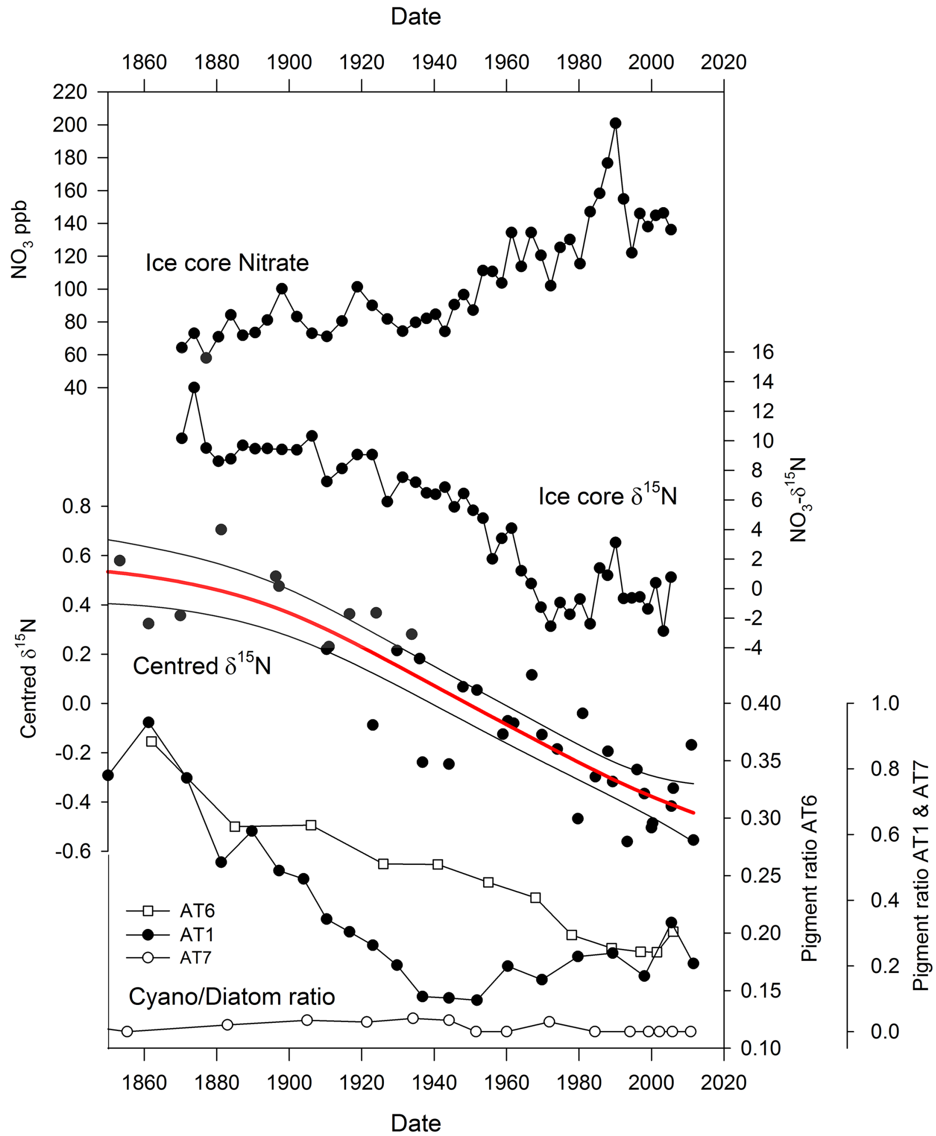

Fig. 4. Summary plot showing the temporal relationship between ice core NO_3^- and $\delta^{15}N$ and sediment $\delta^{15}N$ and trends in pigment-inferred decline in N-fixing algae in two coastal lakes. Two upper most profiles: ice core NO_3^- and $\delta^{15}N$ (data from Holtgrieve et al. 2011); lower central are centered values of $\delta^{15}N$ from the three lakes at the coast with significant declines in $\delta^{15}N$ (*see* Fig. 3). The fitted smooth (red line) is the result of a generalized additive model (GAM) fitted to the original, noncentered $\delta^{15}N$ values. The lowermost profiles are pigment ratios for three coastal lakes (AT1, AT6, and AT7) illustrating the relative change in the abundance of canthaxanthin (calculated as the ratio of canthaxanthin to diatoxanthin and alloxanthin concentrations; *see* Supporting Information) as a decline in potentially N-fixing cyanobacteria as the atmospheric N-load increases. The lack of change at AT7 is discussed in the main text. [Color figure can be viewed at wileyonlinelibrary.com]

organic matter in lake sediments is due directly to the large depletion in atmospheric NO_3^- (as recorded in the Summit ice core). This process results in the incorporation of an isotopically light source of NO_3^- by organisms, thereby causing the isotopic signature of the organic matter becoming progressively lighter ($\delta^{15}N$ depleted) over time and subsequently stored in the sediment records. This hypothesis gives little consideration to the role of transformations of deposited N in both lakes and catchment vegetation and soils and to the stimulation of aquatic and terrestrial production. Moreover, while it is often suggested that N from fossil fuel combustion sources is depleted in ^{15}N relative to precipitation, there is little evidence of a difference between the $\delta^{15}N$ of N from precipitation in clean and polluted areas (e.g., Heaton 1986). However, this second mechanism does not depend upon an isotopically distinct source of N from fossil fuel sources; observed values for NO_3^- in deposition are lower than pre-industrial $\delta^{15}N$ values observed from bulk sediments and as such, increased deposition of N could lead to the observed $\delta^{15}N$ depletion without a distinct isotopic signature simply by virtue of increasing proportions of this relatively light source of N.

Trophic interactions influence the fractionation of N and hence the δ^{15}N record (France 2012) but the lakes in this study have short food chains (fish are present in only a few lakes) and their low productivity means input of organic debris from cladocerans, etc. is limited. Likewise, remains of aquatic higher plants, while present they are mainly limited to the littoral zone. Sediment cores were taken in the deepest part of the lakes where there is little, if any, in situ growth of macrophytes. The mean C/N ratio of lake sediments around Sisimiut is ca. 12, suggesting only a small terrestrial component of the organic fraction.

The sediment δ^{15}N profiles across SW Greenland show a distinct regional geographic pattern which reflects gradients in N deposition and δ^{15}N in snowpack (Fig. 2). The coastal lakes (with the exception of AT7) cover a large range of background δ^{15}N values (1–3.7‰) and show a ~ 1‰ decline since ~ 1880 whereas the inland and ice margin lakes tend to show little trend with values varying between 1‰ and 2‰ over the past ~ 100–130 yr. Analyses of a late Holocene sequence from lake SS1220 (located in the inland lake area) had δ^{15}N values ranging from 1‰ to 2.6‰ over the last 3500 yr (Olsen et al. 2013) with little obvious trend. However, there are some minor trends at SS2 (inland) (Sobek et al. 2014) which has a positive trend up core, reaching 2.7‰ at the core surface and SS16 (inland; Fig. 3, insets) and SS903 (ice sheet margin) which vary around 2‰ (Fig. 3). The different starting points for pre-industrial δ^{15}N of lake sediments at the coast (Fig. 3) may indicate differing equilibria between natural atmospheric nitrate inputs (+11‰), N-fixation (as indicated by the cyanobacterial pigments; cf. Fig. 4) and allochthonous sources of N generated within lake catchments. The limnological and landscape controls on the regional patterns in δ^{15}N are discussed in the next section.

The declining δ^{15}N trend in three of the coastal lake sediment records agrees well with the ice core δ^{15}N profile (Fig. 4) and suggests an increasing input of isotopically light NO_3^- over the last ~ 100–130 yr. This trend is only apparent at the coast where the proportional input of most depleted ^{15}N to the TN pool is greatest (Table 1). While there are greater marine inputs of NH_4 at the coast, there is little evidence for changing NH_4 deposition over the time period covered by the sediment cores (*see* Curtis et al. 2018 for a detailed discussion of NH_4 inputs and post-depositional processing). Moreover, the TN inputs are higher at the coast due to greater extent of thin soils and low vegetation cover (with the exception of the AT7 catchment; see below). The different trajectories of change in sediment δ^{15}N over the past ~ 150 yr in coastal lakes investigated could reflect difference in the relative importance of fossil fuel sources of NO_3^- to the total in-lake N pool at this location; the annual N flux relative to water column N pool is greatest at the coast (Table 1). Another factor that might be amplifying ^{15}N depletion in the coastal lakes is that they are more hydrologically flushed [open] systems. Compared to the long retention time of the inland and ice margin lakes, where all inputs must be internally recycled and hence not fractionated, coastal lakes can lose NO_3^- through surface outflows. Osburn et al. (2017) recently highlighted the importance of hydrological linkage for DOC quality in coastal lakes. Further support for a hydrological component comes from the strong relationship between the high DOC concentration and organic N in the long-retention time lakes of the inland area (Anderson and Stedmon 2007).

Direct NO_3^- deposition as a proportion of the in-lake pool is greatest at the coast (Table 1), a detailed study of catchment snowpack (Curtis et al. 2018) found that sublimation and/or wind redistribution of snow resulted in an enhanced gradient from ice sheet to coast in terms of SWEs sitting on the lake ice, from 9–41 mm at the ice sheet and 43–53 mm at inland sites to 91–139 mm at the coast. Hence NO_3^- in the snowpack sitting on lake ice is at least 3.7× greater at the coast than inland, when expressed as a proportion of the lake TN pool. Coastal sites showing sediment δ^{15}N depletion signals (AT1, AT6) have 2–5× more NO_3^- in lake snowpack (as a proportion of the lake TN pool; Table 1) than the only coastal site (AT7) which does not show δ^{15}N depletion. Coastal site AT7 is more similar to those inland sites that show no trend in δ^{15}N, in terms of the lake ice snowpack contribution to the TN pool. This this site has a much greater in-lake NO_3^- concentration than other coastal lakes (Table 1) which means that the ratio of inputs to lake TN pool is much smaller (Table 1).

The recent (post-1996) upturn in sediment δ^{15}N seen at AT6 and AT7 must either reflect a change in dominant atmospheric N sources (e.g., from fossil fuel and agriculture; cf. the ice core nitrate reported in Hastings et al. 2009) or the presence of diagenetic effects and perhaps differing sedimentation rates (see below). Ice core records show a clear decrease in fossil-fuel derived sulfate since around 1980 AD which is not so apparent in NO_3^-, potentially derived from other nonfuel sources which are increasing as fossil fuel emissions decrease (Fischer et al. 1998; Geng et al. 2014). When comparing the similar NO_3^- deposition to the whole catchment (i.e., maximum possible input) relative to the lake TN pools (both c. 13% per year) (Table 1) at AT1 (δ^{15}N decline) and AT7 (no δ^{15}N decline), the differing δ^{15}N profiles cannot be explained. It is speculated that the presence of a morainic wetland upstream of AT7 is very likely attenuating the atmospheric NO_3^- signal conveyed to the downstream lake. There are also small lakes/ponds feeding AT7, which together with the wetland may increase catchment retention.

Limnological controls

There are number of aspects of the limnological gradients in the Kangerlussuaq study region that are relevant to the discussion of N-cycling: the TN pool, NO_3^- seasonality, nutrient limitation, and vertical stratification with associated bottom-water anoxia. Limnologically, the inland lakes are the most distinct with high DOC and conductivity (Table 2), hypolimnetic anoxia is common and there is a greater total in-lake N pool (TN > 900 μg L^{-1}) compared to the coastal lakes (< 300 μg L^{-1}). Much of the TN in the inland lakes is organic N (correlation of DOC and TN: $r = 0.78$; *see* Anderson and Stedmon 2007); in August 2001, > 95% of the TN pool was in organic form (Anderson unpubl. data). Importantly, the inland lakes have a much lower atmospheric input of less depleted δ^{15}N (Fig. 2) but also have substantially longer retention times (Leng and Anderson 2003) which allows for microbial processing of the in-lake N pool. This greater recycling in the Inland region may account for the uniform δ^{15}N in the sediment profiles of these lakes (Fig. 3) (McCarthy et al. 2007).

Although the ice sheet margin and coastal lakes do not differ in terms of conductivity and hypolimnetic O_2 availability (Table 2) (*see* Whiteford et al. 2016 for details), there are significant differences in NO_3^- concentration, which is greater in coastal lakes and rarely above detection limits at inland and ice margin sites. However, there was also seasonal variability across the region with maximum lake water NO_3^- concentration recorded under ice or immediately after ice-out. Seasonally, nutrient limitation of phytoplankton yield differed significantly among sampling occasions; there were more occurrences of P-limitation under ice. Regional and seasonal variations in nutrient limitation status are linked to patterns of atmospheric Nr delivery and recycling of organic N under ice. Lowest rates of Nr delivery close to the ice sheet margin (Fig. 2), combined with elevated P inputs (via glacially derived dust) (Pulido-Villena et al. 2008; Bullard and Austin 2011; Hawkings et al. 2016) likely drive an imbalance in phytoplankton N : P stoichiometry (Table 2), generating increased growth-led demand for N. At the coast, where increased delivery of Nr accumulated in winter snowpack is released to lakes upon thaw, the N : P stoichiometric imbalance drives an increased demand for P (as recorded in spring under ice cover).

As many Arctic lakes are oligotrophic, low resource systems, where production is tightly controlled by nutrient supply (Bonilla et al. 2005), any increase in nutrient (e.g., Nr) supply could have a marked impact on phototrophic growth, stimulating primary production and impacting whole lake ecological structure and function. One adaptation to N scarcity in Arctic lakes (as with Arctic terrestrial ecosystems), is the abundance of N-fixing organisms (e.g., some cyanobacteria, lichens) which play a crucial role in alleviating ecosystem N-limitation. *Nostoc* are common in lakes throughout the study region, but particularly so in the central and ice marginal lakes and contain large amounts of the carotenoid canthaxanthin. Some indication of the increased N supply to the coastal lakes is provided by decreases in the relative abundance of canthaxanthin, a pigment associated with potentially N_2-fixing cyanobacteria in the coastal lakes AT6 and AT1, over the past ~ 100 yr (Fig. 4; see also Supporting Information Fig. S14). This decrease (which mirrors changes in the ice core NO_3^- concentration and coastal lake sediment δ^{15}N) suggests that an increased input of Nr helped to alleviate N-limitation in diatoms and cryptophytes or reduced the competitive benefits associated with N-fixation. At lake AT7, the low relative abundance of canthaxanthin (compared to AT1 and AT6) and absence of a depletion signal in the δ^{15}N profile are consistent with less N-limitation at this site; the lake has the highest NO_3 concentration of any lake in the study (Table 1). The lack of change in the pigment ratio over the last 150 yr at AT7 together with high total carotenoid concentrations would suggest that this lake has always had abundant and higher than average algal production. The local NO_3^- source is unknown but is presumably derived from a wetland/glacial deposit located in its catchment.

Anoxia and diagenesis

Diagenesis can lead to marked changes in the isotopic values of nitrogen in organic matter post deposition (Talbot 2001). Most often this occurs in anoxic environments which result in denitrification, although other processes are known to affect isotopic values (Teranes and Bernasconi 2000; Burgin and Hamilton 2007). The anaerobic processes of denitrification and ammonification both heavily discriminate against ^{15}N resulting in enrichment of the DIN reservoir and consequently also of subsequently derived organic matter (Wolfe et al. 1999; Olsen et al. 2013). If these processes (ammonification and denitrification) were dominant, the resultant δ^{15}N values should be higher than the δ^{15}N observed values in the inland lakes where anoxia is prevalent. Interestingly, the geographic pattern in regional δ^{15}N profiles does not correspond with limnological O_2 gradients: the lakes with the lowest hypolimnetic O_2 concentrations (the inland cluster) have the most stable δ^{15}N profiles (Fig. 3) while the coastal and ice margin lakes which have differing δ^{15}N profiles have similar hypolimnetic O_2 concentrations (*see* Whiteford et al. 2016).

Some indication of the role played by limnology and anoxia in δ^{15}N processing are, however, shown by the two adjacent lakes on a nunatak at the ice margin and SS16 (*see* Anderson et al. 2001) (Fig. 3, insets). These lakes differ in terms of their thermal stratification and chemical characteristics (Anderson et al. 2001) and the δ^{15}N profiles are different (Fig. 3, inset). The strong positive trend in δ^{15}N at SS86 may be associated with changing abundance of phototrophic bacteria (Reuss et al. 2013). In contrast, at SS1220 (an inland lake) where changing strength of long-term thermal stratification was inferred from geochemical proxies (Olsen et al. 2013), the fluctuations between strong and weak thermal stratification (and hence O_2 availability) did not show in the δ^{15}N record.

If all available N from the DIN pool is assimilated by algae, the $\delta^{15}N$ of organic matter would approach that of DIN (Meyers and Lallier-Verges 1999; Talbot 2001; Brahney et al. 2006). The rapid depletion of the DIN pool in all lakes following ice out (*see* Whiteford et al. 2016) would tend to suggest that this is the case. Using compound specific analyses, Enders et al. (2008) interpreted the decreasing ^{15}N of the algal component as being driven by changes in N delivery, i.e., a shift in N cycling caused by increasing anthropogenic N. They also suggest that diagenetic effects are less than the fingerprint of regional changes in N biogeochemistry, so that bulk sediments reflect the signal of the ^{15}N inputs, i.e., diagenetic effects in the sediment are minor. The generally low sediment $\delta^{15}N$ values (1–2‰) at the inland and ice margin lakes suggest that the organic matter may be derived from cyanobacteria fixing atmospheric nitrogen ($\sim$ 0‰) (Talbot 2001). Sobek et al. (2014) found a strong correlation between the sediment $\delta^{15}N$ value and O_2 exposure time in a group of Greenland lakes, with $\delta^{15}N$ increasing from 0.5‰ to +3‰ as O_2 exposure increased from $\sim$ 2 yr to > 10 yr. The O_2 exposure of organic matter is largely controlled by the sediment accumulation rate and the O_2 content of the hypolimnion, suggesting that more positive $\delta^{15}N$ values should be found in lakes with slow sediment accumulation rates and oxic hypolimnion (i.e., Coastal and Ice Margin). Two coastal lakes (AT6, AT7) show a positive upturn in $\delta^{15}N$ values around the year $\sim$ 2000 AD and it is possible that this is the result of greater O_2 penetration (cf. Sobek et al. 2014), perhaps associated with changed thermal stratification patterns (Saros et al. 2016). Some further support for a recent diagenetic signal at AT7 is suggested by the pigment profiles which indicate changed preservation conditions at this time ($\sim$ 2000 AD). Today, this lake has highly oxygenated bottom waters, presumably due to abundant macrophytes in the littoral zone.

Landscape perspective

The response of lake nitrogen (N) biogeochemistry to catchment vegetation and soil dynamics change is complex (Baron et al. 2013). Mineralized N is frequently a limiting nutrient in Arctic ecosystems (Dormann and Woodin 2002), the fate of which is key to understanding changes in the availability of nutrients to catchment vegetation and lake biota. Where the permafrost underlying tundra has started to thaw (e.g., North Slope of Alaska) (McClelland et al. 2007; Harms and Ludwig 2016; Harms et al. 2016), there have been dramatic changes in nutrient soil water interactions. Deepening of the active layer leads to enhanced soil organic matter decomposition rates and consequently to greater N mineralization. This increases the local supply of N that occurs during spring thaw, resulting in a temporal mismatch between availability and biological uptake, thereby enhancing availability for leaching into surface waters during snow melt. However, Bartrons et al. (2010) showed how soil type in a catchment contributes to higher $\delta^{15}N$ than that related to snow melt processes, which may reflect local soil dynamics and vegetation effects. Increased decomposition rates associated with warming and/or vegetation changes are also reflected in higher $\delta^{15}N$ in soil organic matter and consequently in NO_3^- leached from soils to lakes, which may be recorded in lake sediments. However, Kangerlussuaq was cooling for much of the 20th century (Mernild et al. 2014) and there is also limited hydrological connectivity at the inland and ice margin sites. In the Kangerlussuaq area of SW Greenland, the thickest organic soils are located close to the head of the fjord associated with the dwarf shrub tundra, while there is clearly more biological processing of terrestrial organic matter in this location, there are few hydrological pathways for soil and vegetation isotopic signatures to be transported into the lakes. As a result, catchment soils and vegetation have minimal influence on the $\delta^{15}N$ in these lakes. At the ice margin, the vegetation is also sparse (predominantly steppe) with the result that soils are thin; there are also large areas of bare ground due to the inhospitable climate, wind speeds can be very high. Importantly, however, here too, hydrological connectivity is limited. Snow sublimation is a major factor at both the inland and ice margin sites which influences the snowpack $\delta^{15}N$ and lake hydrologic budgets.

The coastal catchments also have limited vegetation cover (at altitudes > 400 m they resemble high Arctic systems): soils are thin or absent. Total precipitation and snow cover are also substantially higher (see above) and while hydrological connectivity is much greater compared to the inland sites, the thin soils suggest that catchment processing of N is limited, with the exception of the AT7 catchment (see above; Table 1).

At the coast, atmospheric deposition inputs of DIN, whether in spring snowmelt or annual deposition load (Fig. 2), proportionately represent a much greater fraction of the in-lake pool of TN (Table 1). Rapid hydrological delivery means that the isotopic signature of the atmospherically derived NO_3^- is delivered to a much less modified N pool and captured by in-lake production and then sedimented in the lake. Furthermore, coastal deposition is much more depleted in ^{15}N than inland (Fig. 2). The rapid hydrological transfer means that there is limited microbial processing (and hence isotopic fractionation) (Finlay et al. 2007). Theoretically, the total atmospheric NO_3^- deposition flux to an entire lake catchment defines the total potential load (or an upper limit) for NO_3^- which may be transported into a lake. Here, the relationship to lake TN pools is more complex. At the coastal sites, there is an order of magnitude more NO_3^- deposited onto catchments relative to lake TN pools, with NO_3^- deposition averaging $\sim$ 18% of lake TN pools per annum, compared with < 2% inland (Table 1). At site AT6, annual catchment NO_3^- deposition is equivalent to > 30% of the lake TN pool. If deposited nitrate were completely mobile, then an atmospheric deposition source would account for a third of the lake TN pool each year. However, it is known that terrestrial Arctic systems strongly retain N inputs and only a fraction of deposited NO_3^- is likely to reach lakes (Tye et al. 2005). This proportion must though be largest at the coast where precipitation is much higher and surface flow pathways are much more pronounced. If spring snowpack inputs to lakes reflect the dominant atmospheric source, then the signal should be much more apparent in coastal lakes. Likewise, annual deposition to the whole catchment is much greater at the coast, where the conditions for surface runoff inputs to lakes outside of the snow season are also greatest. Hence, although it is not possible to precisely quantify the proportion of catchment snowmelt or annual deposition that is transported into lakes to be utilized for primary production, all the evidence indicates that the coastal sites showing clear sediment ^{15}N signals are those experiencing the greatest deposition inputs, and where atmospheric nitrate also has the lowest $\delta^{15}N$ values (Fig. 2; Table 1). The lack of a signal in AT7 is not due to lower snowpack or deposition inputs, but rather due to the larger lake TN pool and possible processing of catchment N inputs in an upstream wetland which is unique to this site within the current study.

Synthesis

The majority of lakes analyzed for sediment $\delta^{15}N$ along the regional N deposition gradient in SW Greenland show little agreement with the ice core records of altered atmospheric N dynamics (both quality and total amount) (Hastings et al. 2009; Holtgrieve et al. 2011). This lack of a signal, of course, does not preclude ecological effects driven by N-deposition in this area, merely that there is no sediment $\delta^{15}N$ record of the increased deposition. In contrast, most coastal lakes show the characteristic decline of $\sim$ 1–1.5‰ in $\delta^{15}N$ over the last 100–130 yr that has been observed in selected northern hemisphere lakes (Holtgrieve et al. 2011). The differences in historic $\delta^{15}N$ sediment records observed in this study (over a 170 km transect) cannot be readily explained by limnological processes and/or diagenesis but most likely reflects the interplay between the atmospheric N-load, the more depleted isotopic signature at the coast, N processing in the snow pack and the size of the in-lake N pool. At coastal sites, the rapid incorporation of the isotopically negative, reactive N into organic matter and transfer to the sediment provides an unambiguous record of changing N dynamics in the coastal region. Hobbs et al. (2016) found that the relationship between sediment $\delta^{15}N$ and measured atmospheric deposition relies on catchment N retention. Given the observations in this study and those elsewhere (Curtis and Simpson 2011) variable $\delta^{15}N$ signals are probably the norm and that only lakes and catchments with certain characteristics will record the atmospheric depletion signal. Although the N-load is low in SW Greenland (< 0.5 kg ha^{-1} yr^{-1}), sustained inputs are sufficient to alter the $\delta^{15}N$ signal (and drive ecological change) if the recipient lakes are oligotrophic and sensitive to external forcing having developed under low background N deposition loads.

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Acknowledgments

This work was funded by awards from NERC (UK) (NE/G019622/1 and NE/G019509/1).

Conflict of Interest

None declared.

Submitted 09 August 2017
Revised 13 March 2018; 27 March 2018
Accepted 24 April 2018

Associate editor: Emily Bernhardt