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RESEARCH ARTICLE

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Key Points:

- N and P varied at the individual lake scale but were synchronous at the regional scale, suggesting control by broad environmental drivers
- Of the drivers examined, declines in lake DIN were most closely related to declines in atmospheric DIN deposition, especially in the north
- Declining lake DIN:TP promoted a transition from P and NP toward N limitation in some lakes and intensified N limitation in others

Supporting Information:

- Supporting Information S1

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Recent Synchronous Declines in DIN:TP in Swedish Lakes

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Abstract Declining atmospheric nitrogen (N) deposition in northern Europe and parts of North America, coupled with ongoing changes in climate, has the potential to alter the nutrient limitation status of freshwater ecosystems. In this study we compared time series data of atmospheric N deposition, air temperature, and precipitation with corresponding estimates of dissolved inorganic nitrogen (DIN), total phosphorus (TP), DIN:TP, and total organic carbon from 78 headwater streams and 95 nutrient-poor lakes in Sweden from 1998 to 2013 to assess trends in, and potential drivers of, lake N:P ratios. We found that trends in nutrients were variable at the scale of individual lakes but were highly synchronous at the regional scale, suggesting underlying control by broad-scale environmental drivers mediated by site-specific characteristics. Widespread declines in lake DIN throughout Sweden were correlated with declines in atmospheric N deposition, particularly in northern areas. TP did not have strong directional trends, but interannual variability was synchronous at regional scales, implying that broad-scale climate drivers were affecting these trends. Overall, we observed a significant decline in DIN:TP throughout Sweden over the monitoring period. At the beginning of the study period, 32% of lakes were N limited and 45% colimited by N and P. Proportions increased to 63% of lakes N limited and 20% colimited by N and P at the end of the study period. These results suggest that N limitation is likely to become more widespread in subarctic and boreal areas of Europe in the future if recent trends continue.

Plain Language Summary This article examines the way in which changes in the amount of nitrogen from the atmosphere being delivered to lakes (as a result of fossil fuel combustion) are interacting with global climate change to affect nutrient availability in Swedish lakes. Nitrogen can act as fertilizer in lakes, supporting increased growth of algae and aquatic plants. The amount of nitrogen relative to other important elements such as phosphorus can help to determine which groups of plants and algae dominate lake ecosystems, as well as how much living biomass lakes can sustain. We find that declines in atmospheric deposition of nitrogen, which have resulted from the adoption of policies controlling emissions from fossil fuel combustion, have caused declines in nitrogen concentrations in lakes throughout Sweden. This has changed the balance of nitrogen and phosphorus, which may result in changes to the structure of lake biological communities. At the same time, variability in climate also has subtle but widespread effects on lake nutrient concentrations, suggesting that the availability of nutrients in lakes at northern latitudes is likely to change in the future as the climate warms.

1. Introduction

Human activities have dramatically changed global nitrogen (N) and phosphorus (P) cycles directly through the use of fertilizers and release of N through fossil fuel combustion (Vitousek, 1997) and indirectly through climatic changes affecting nutrient cycling (Jeppesen et al., 2009). These large-scale changes in nutrient cycles are important, because N and P are among the most important resources limiting primary production in both terrestrial and aquatic ecosystems (Elser et al., 2007; Harpole et al., 2011). In lakes specifically, the absolute concentrations of N and P, as well as the ratio of N:P, frequently control plankton community composition, total primary production, and total biomass (Brauer et al., 2012; Downing et al., 2001; Guildford & Hecky, 2000; Smith, 1983). If anthropogenic pressures impact the transport and cycling of N and P differently, lake N:P will change, resulting in corresponding shifts in nutrient limitation (Isles et al., 2017; Penuelas et al., 2013; Yan et al., 2016).

In arctic and boreal regions, which generally have low human population densities and contain the highest density of lakes (Verpoorter et al., 2014), N and P concentrations are likely to be most strongly influenced by diffuse anthropogenic drivers operating at broad spatial scales (Holtgrieve et al., 2011; Vitousek, 1997). These drivers include atmospheric N deposition (Bergström & Jansson, 2006), atmospheric P deposition (Wang

et al., 2014), atmospheric sulfur deposition (Gerson et al., 2016), and global climate change (Williamson et al., 2009). Of these drivers, changes in N and P deposition may have the most obvious direct impacts on lake nutrient concentrations (Hessen, 2013; Sickman et al., 2003). Atmospheric P deposition remains poorly characterized, but may be increasing, and has been suggested as a potential driver of recent increases in lake total phosphorus (TP) in the United States (Stoddard et al., 2016; Wang et al., 2014). High and increasing atmospheric N deposition can raise N concentrations in lakes, resulting in changes in lake stoichiometry and increased P limitation (Elser et al., 2009; Hobbs et al., 2016; Weyhenmeyer et al., 2014). However, the responses of lakes to atmospheric N deposition are strongly mediated by catchment characteristics (Arvola et al., 2015; Hessen et al., 2009; Meunier et al., 2015), such as size, land cover, slope, and vegetation dynamics (Canham et al., 2012), as well as by lake characteristics such as morphometry and color (Deininger et al., 2017). Atmospheric N deposition (where N is usually measured as dissolved inorganic nitrogen (DIN), NH_4^+ , and NO_x) has declined recently in North America and Europe as a result of increased controls on NO_x from fossil fuel combustion beginning in the 1970s and 1980s (Lajtha & Jones, 2013), although DIN deposition is increasing in other areas (Liu et al., 2013).

Many lakes impacted by recent decreases in DIN deposition have also experienced recent increases in dissolved organic matter (DOM) inputs (also referred to as lake browning), which are generally attributed to recovery from sulfur deposition as well as climate warming (Monteith et al., 2007; Weyhenmeyer & Karlsson, 2009) (although a number of other factors contribute to DOM inputs, including increased production of catchment vegetation (i.e., catchment greening) (Finstad et al., 2016) and wetter climate (de Wit et al., 2016)). Increases in DOM inputs can directly affect N and P concentrations because both nutrients are complexed with DOM (Kissman et al., 2016; Kortelainen et al., 2006; Seekell et al., 2015). In addition to direct impacts, DOM inputs to lakes can impact N and P concentrations indirectly by inducing light limitation, stimulating bacterial production, and lowering oxygen concentrations (Karlsson et al., 2009; Zwart et al., 2016). Several studies have also suggested that changes in C:N ratios resulting from increased dissolved organic carbon may affect denitrification rates (Taylor & Townsend, 2010; Weyhenmeyer & Jeppesen, 2009). Taken together, changes in mass inputs of DIN from deposition and DOM from browning may lead to substantial changes in lake N:P stoichiometry.

In addition to changes in DIN deposition and lake browning, climate change may also impact catchment inputs and processing of nutrients within lakes. Delivery rates of N, P, and C from catchments can be affected by both temperature and precipitation changes (Jeppesen et al., 2009, 2010; Weyhenmeyer & Karlsson, 2009). Changes in precipitation are likely to affect lake nutrient stoichiometry by changing both the timing and magnitude of nutrient inputs. In subarctic and boreal regions, nutrient loading to lakes is likely to occur earlier in the year as more nutrients are delivered in the winter months (Bouraoui et al., 2004). Warmer temperatures and longer growing seasons may lead to increased catchment greening and increased watershed retention of N (Hessen et al., 2009; Lucas et al., 2016). Catchment greening may also affect P loading; however, insufficient attention has been given to P dynamics in undisturbed boreal catchments in Scandinavia. Within lakes, warming may affect water temperature and the length of the stratified period (Read et al., 2014), potentially resulting in reduced oxygen, increased internal P loading, and increased denitrification (Myrsten et al., 2016; Veraart et al., 2011). All of these interacting DIN deposition and climate-driven changes are likely to affect lake nutrient stoichiometry.

Understanding the effects of large-scale environmental drivers is difficult because site-specific features related to catchment properties or lake morphometry may mask the signal of these drivers at the scale of individual lakes (Read et al., 2015). Because many sources of variation affect N and P concentrations in individual lakes, comparable data sets from multiple lakes are needed to identify long-term impacts of diffuse environmental drivers. Sweden, which straddles boreal and subarctic biomes, has large gradients of DIN deposition, lake color, temperature, and precipitation, making it well suited for studying of the impact of large-scale climatic and depositional changes on lake nutrient stoichiometry (Bergström et al., 2005; Seekell et al., 2015). Sweden also has an extensive water-quality monitoring program, with data from hundreds of lakes and headwater streams analyzed using consistent methods. Moreover, most Swedish lakes are nutrient poor, making them sensitive to changes in nutrient concentrations (Karlsson et al., 2009), and phytoplankton are frequently found to be N limited in both boreal and subarctic areas in Sweden (Bergström et al., 2015, 2008; Elser et al., 2009). Climate is also warming faster in Scandinavia than in other

parts of Europe (Vautard et al., 2014). As a result of all of these factors, Swedish lakes may be particularly sensitive to changes in atmospheric N deposition as well as climate (Elser et al., 2009).

In this study, we compared both spatial and temporal trends in DIN, TP, DIN:TP, and total organic carbon (TOC) in 78 headwater streams and 95 nutrient-poor Swedish lakes with corresponding estimates of DIN deposition, air temperature, and precipitation from 1998 to 2013. While several studies have observed declining DIN deposition in southern Sweden (Lucas et al., 2016) and impacts of changes in DIN deposition on lake DIN concentrations (Elser et al., 2009; Weyhenmeyer et al., 2007; Weyhenmeyer & Jeppesen, 2009), to our knowledge none have addressed these changes in the context of simultaneous changes in lake P over time, or the consequences of these changes for N:P and lake nutrient limitation. We hypothesized that (1) changes in DIN deposition would be correlated with changes in lake DIN; (2) changes in TP would be positively correlated with changes in lake TOC as a result of cotransport of phosphorus with DOM; and (3) because of these two processes, lake DIN:TP would decline in areas with browning and declining DIN deposition.

2. Methods

2.1. Meteorological Data

Estimates of daily air temperature and precipitation were downloaded from the interpolated Luftweb data product (Swedish Meteorological and Hydrological Institute; <http://luftweb.smhi.se>) for grid cells corresponding to the lake monitoring sites. Mean annual temperature (MAT; °C) and mean annual precipitation (MAP; mm d⁻¹) were calculated for each lake. Because air temperature and precipitation were being compared to July–September averages of lake data, and to capture possible effects on antecedent winter conditions on lake biogeochemistry, we used water year rather than calendar year in our calculation of MAT and MAP (so, e.g., water year 2000 is 1 October 1999 to 31 September 2000).

2.2. Atmospheric DIN Deposition Data

Gridded, interpolated estimates of both NH₄⁺ and NO_x total (wet + dry) deposition were downloaded from the Swedish Meteorological and Hydrological Institute (<http://www.smhi.se/klimatdata/miljo/atmosfarskemi>) as shapefiles for each calendar year from 1998 (the first year data were available) to 2013. Data were extracted for grid cells corresponding to the monitored lake and stream sites. DIN deposition (DIN_{dep}) was calculated as the sum of NH₄⁺ and NO₃⁻ deposition. Lakes were divided into four regions corresponding to quantiles of DIN deposition (“low,” 0–25th percentiles; “medium,” 25–50th percentiles; “high,” 50–75th percentiles; and “highest,” 75–100th percentiles). DIN deposition thresholds corresponding to these quantiles were used to assign the lake and stream sites to corresponding DIN deposition regions.

2.3. Lake and Stream Data

Summary information about catchment characteristics and lake size is provided in Table 1. Data on nitrate–nitrite N (NO_x), ammonium (NH₄⁺), TP, and TOC from 1998 to 2013 were downloaded from the trend lakes, reference lakes, trend streams, and reference streams data sets collected by the Swedish long-term monitoring program (<http://miljodata.slu.se/mvm/Default.aspx>). Lakes were selected that had low proportions of agricultural land (1 lake had 17%; all others were below 9%) and developed land (consisting of urban development, roads, mining, and recreational parks; maximum 4.5%, most below 2%) in their catchments and that had data for the entire study period. Only epilimnetic data from lakes were used. Lakes with average TP > 25 μg L⁻¹ were excluded from the analysis to minimize the effects of anthropogenic eutrophication on the results (cf. Bergström et al., 2005).

For lakes, data were averaged from July to September (hereafter referred to as summer). This time window excludes a substantial fraction of the growing season in southern Sweden; however, many northern lakes do not become ice free until May or June, making it difficult to compare spring data across regions. Furthermore, biogeochemical data during the spring snowmelt period are highly variable at short time scales and sensitive to specific melt events (Pellerin et al., 2011; Sebestyen et al., 2008), so individual samples are unlikely to represent true seasonal averages (particularly in small lakes with short residence times), and the high seasonal variance may mask long-term trends.

For streams, data were averaged from May to September. May and June data were included in these averages because most nutrient loads to lakes are delivered during the spring snowmelt pulse (restricting our analysis

Table 1*Mean Catchment Characteristics for Lakes in the N Deposition Regions (See Figure 1 for Region Definitions)*

Region	% forest	% wetland	% agriculture	Lake surface area (km ²)	Catchment area (km ²)	Catchment/lake area
Low	45.6 (22.7)	10.1 (9.8)	0.4 (0.6)	2.6 (4.0)	166.6 (502.3)	20.4
Medium	64.0 (8.8)	3.1 (4)	1.1 (1.9)	0.8 (0.6)	15.1 (22.2)	11.3
High	68.4 (7.2)	1.5 (2.3)	1.9 (3)	1.4 (3.0)	8.8 (14.4)	7.3
Highest	71.9 (10)	2.2 (3.6)	3.2 (4.1)	0.8 (1.2)	15.4 (38.2)	11.5

Note. Values are expressed as arithmetic mean (st. dev) for all variables except for catchment:lake area ratio, which is expressed as the geometric mean.

of stream data to July–September low-flow months would only reflect a small proportion of annual nutrient loading to most Swedish lakes), and spring nutrient loads have been found to be particularly important for the development of summer phytoplankton populations (e.g., Kohler & Hoeg, 2000; Stumpf et al., 2012). Most Swedish lakes have residence times <100 days (Lindström et al., 2006), so input water from May to September likely represent a substantial fraction of the nutrients available to summer production. DIN_{lake} , TP_{lake} , TOC_{lake} , and $\log_{10}(\text{DIN:TP})_{\text{lake}}$ refer to annual summer averages (July–September); $\text{DIN}_{\text{stream}}$, $\text{TP}_{\text{stream}}$, $\text{TOC}_{\text{stream}}$, and $\log_{10}(\text{DIN:TP})_{\text{stream}}$ refer to annual May–September averages for all subsequent analyses of spatial gradients, trends, and synchrony unless otherwise stated. For completeness, z scores of April–June averages of chemical variables in both lakes and streams are also presented in supporting information Figure S1.

N to P ratios were represented as molar ratios of DIN:TP. DIN:TP has been shown to be a robust predictor of phytoplankton nutrient limitation in Swedish lakes (Bergström, 2010) and in other areas with high DOM (Axler et al., 1994). DIN in lakes and streams was calculated as the sum of NO_x and NH_4^+ . To avoid averaging errors resulting from the nonnormal distribution of ratio data, DIN:TP was calculated as $\log_{10}(\text{DIN:TP})$ of daily observations. The use of log transformations is generally appropriate when calculating statistics with ratio data (Keene, 1995) and is necessary when averaging ratio data to accurately represent nutrient limitation status (Hillebrand et al., 2013; Keene, 1995).

2.4. Data Analysis and Statistical Methods

Total changes in lake nutrients ($\Delta\text{TP}_{\text{lake}}$, $\Delta\text{DIN}_{\text{lake}}$, $\Delta\text{TOC}_{\text{lake}}$, and $\Delta\log_{10}(\text{DIN:TP})_{\text{lake}}$) at individual sites were assessed by calculating the slope of the Thiel-Sen regression line using the R package rkt (Marchetto, 2017) and multiplying by the length of the study period. In addition, to quantify the changes in lake nutrient limitation status over the monitoring period and percent changes in lake nutrients, the difference between the average of the first 5 years of the study period (1998–2002) and the average of the last 5 years (2009–2013) was calculated for DIN_{lake} , TP_{lake} , TOC_{lake} , and $\log_{10}(\text{DIN:TP})_{\text{lake}}$. Temporal changes in each variable were used to address hypothesis 3 (DIN_{lake} would decline in areas with increasing TOC_{lake} and decreasing DIN_{dep}).

To test hypothesis 2 (changes in TP_{lake} will be correlated with changes in TOC_{lake}) at both spatial and temporal scales, correlations between TP_{lake} (and DIN_{lake}) and TOC_{lake} were first calculated across the entire data set (including all sites and years). To investigate temporal correlations while controlling for spatial gradients, correlations were also calculated between TP_{lake} and TOC_{lake} within each individual lake and between DIN_{lake} and TOC_{lake} within each individual lake. The averages of the individual lake correlations were then calculated. Average correlations greater than 1.96 standard errors from 0 were considered statistically significant.

Synchrony (S) is a measure of the degree to which different sites show the same patterns of year-to-year variability. To quantify S in temporal trends in individual variables, a full correlation matrix among lakes was calculated for each of the chemical variables (TP_{lake} , DIN_{lake} , TOC_{lake} , and $\log_{10}(\text{DIN:TP})_{\text{lake}}$). For each variable, S was calculated as the average of the correlation matrix. S can range from slightly negative (~ -0.1) to 1.0 in the case of perfectly correlated time series among all lakes (Vogt et al., 2011). High values of S suggest that lakes are responding to similar drivers. All correlations were calculated using Pearson's product-moment correlation. To calculate the significance of S values for each variable, a permutation test was used following Vogt et al. (2011). The observed average S was compared to a distribution of r values derived from 1,000 randomizations of the original time series, and values below the 2.5th or above the 97.5th percentiles were considered significant. For all variables, S was significant when $S > 0.04$.

To compare relative trends over time across sites with different nutrient concentrations and stoichiometry at the regional scale, nutrient concentrations and log ratios were standardized to z scores for each variable within each lake and stream. For each nutrient N in year i , the z score was calculated as $(N_i - \text{mean}(\mathbf{N}))/\text{st. dev}(\mathbf{N})$. Standardized values were then averaged across lakes and streams for each year within each N deposition region (defined in section 2.1). Regional averaging was performed to allow comparisons between stream and lake data (because sampled stream sites were not paired with sampled lake sites) and to filter out lake-specific differences to detect underlying regional trends. Regional average z scores were also used to test hypothesis 1 (DIN_{lake} will be positively correlated with DIN_{dep}). To determine temporal trends in each variable, the regional Kendall test was used on regional averages by year, using region as a blocking variable. This nonparametric regression was used to minimize the effects of outliers on the results. Thiel-Sen slopes were also calculated to estimate the rates of increase or decline in nutrients. Kendall tests and Thiel-Sen slopes were calculated using the R package *rkt* (Marchetto, 2017).

2.5. Estimating the Importance of Direct Deposition

To estimate the contribution of DIN_{dep} to a typical lake (i.e., direct DIN input via DIN deposition onto the lake surface) relative to surface water inputs, a simple model was developed based on median DIN deposition (DIN_{dep} , $\text{mg m}^{-2} \text{ lake yr}^{-1}$), area-normalized lake volume based on average lake depth (expressed as z , $\text{m}^3 \text{ m}_{\text{lake}}^{-2}$; the volume of a column of water with surface area of 1 m^2 and depth z), median stream DIN concentrations ($[\text{DIN}_{\text{stream}}]$, mg m^{-3}), and published estimates of lake turnover times in Sweden (T_{lake} , yr^{-1}) (Lindström et al., 2006). First, surface water input volumes V_{stream} ($\text{m}^3 \text{ m}_{\text{lake}}^{-2} \text{ yr}^{-1}$) were calculated as the product of lake turnover time and lake depth (equation (1)). Stream load ($\text{DIN}_{\text{streamLoad}}$, $\text{mg m}_{\text{lake}}^{-2} \text{ yr}^{-1}$) was then calculated as the product of V_{stream} and $[\text{DIN}_{\text{stream}}]$ (equation (2)). Percent DIN inputs from direct DIN deposition for a hypothetical lake (%Deposition) were then calculated from DIN deposition rates and estimated surface water fluxes (equation (3)). %Deposition was calculated across a gradient of lake sizes and depths using median $[\text{DIN}_{\text{stream}}]$ and median DIN deposition from this data set. A graphical explanation of the model is provided in supporting information Figure S2.

$$V_{\text{stream}} = T_{\text{lake}} \times z \quad (1)$$

$$\text{DIN}_{\text{streamLoad}} = V_{\text{stream}} \times [\text{DIN}_{\text{stream}}] \quad (2)$$

$$\% \text{Deposition} = \text{DIN}_{\text{dep}} / (\text{DIN}_{\text{streamLoad}} + \text{DIN}_{\text{dep}}) \times 100 \quad (3)$$

A similar model was constructed to estimate the importance of direct total atmospheric N (organic + inorganic) deposition relative to total N (TN) inputs using the median stream concentration of TN from 2007 to 2016 (when data were available). Assuming that 15% of total atmospheric N deposition is organic (based on the low end of estimates for Europe reported in Cornell et al., 2003), TN_{dep} was calculated as $\text{DIN}_{\text{dep}}/0.85$. Both DIN and TN models ignored contributions from groundwater, N fixation, and lake sediments.

3. Results

DIN_{dep} had a strong spatial gradient, with high deposition in the southeast, moderate deposition in central Sweden and along the Baltic coast, and low deposition in the north (Figure 1). Average DIN_{dep} decreased significantly over the study period in each defined DIN deposition region (p values of regressions of deposition $\times$ year were 0.0019, 0.0006, 0.0004, and 0.00001 in the low, medium, high, and highest DIN deposition areas, respectively). The average rate of decline ranged from $-10.74 \text{ mg N m}^{-2} \text{ yr}^{-1}$ in the low deposition region to $-32 \text{ mg N m}^{-2} \text{ yr}^{-1}$ in the highest deposition region; however, proportional declines (relative to the mean of the first 5 years) were greatest in the low region (43%), followed by medium (38%), highest (37%), and high (30%).

Spatial patterns were different for $\Delta \text{DIN}_{\text{lake}}$, $\Delta \text{TP}_{\text{lake}}$, and $\Delta \text{TOC}_{\text{lake}}$ (Figure 2). DIN_{lake} declined in most sites over the monitoring period (12 increasing, 7 no trend, and 76 decreasing; Figure 2). The largest absolute declines were in the southeast, where DIN deposition was highest and where DIN_{lake} concentrations were also highest. More lakes had declining TP_{lake} than increasing TP_{lake} (28 increasing, 28 no trend, and 39 decreasing), but there was no clear spatial pattern across Sweden distinguishing areas with increases or decreases. Most lakes had inclining TOC_{lake} (83 increasing, 3 no trend, and 9 decreasing), but there was a strong spatial gradient, as reported in previous studies (Weyhenmeyer & Jeppesen, 2009). TOC_{lake} increased substantially over the

Figure 1. Annual dissolved inorganic nitrogen (DIN) deposition ($\text{mg N m}^{-2} \text{yr}^{-1}$) at Swedish long-term monitoring sites, separated into four groups (low, 0–25%; medium, 25–50%; high, 50–75%; and highest, 75–100%). Locations of lakes in each category are shown in map to the right.

study period in the south and southeast in areas where DIN deposition was highest, but lakes in the northeast tended to have no change or declining TOC_{lake} (Figure 2). $\log_{10}(\text{DIN:TP})_{\text{lake}}$ declined in most lakes (22 increasing, 1 no trend, and 72 decreasing), resulting in a shift in the distribution of lakes toward greater N limitation. In the first 5 years of the study period, 32% of lakes were primarily N limited and 45% were colimited by N and P during the summer using thresholds from Bergström (2010) (i.e., N limited below 3.3; NP limited between 3.3 and 7.5; and P limited above 7.5); however, in the last 5 years, the percentage increased to 63% of lakes primarily N limited and 20% colimited by N and P during the summer (Figure 3).

The degree of synchrony among lakes was significant for all variables (DIN_{lake} , TP_{lake} , TOC_{lake} , and $\log_{10}\text{DIN:TP}_{\text{lake}}$; Figure 4), but values of S differed among chemical variables and were relatively low compared to other studies of synchrony in lake time series (Rusak et al., 1999; Vogt et al., 2011). DIN_{lake} had the lowest synchrony ($S = 0.12$), and 40% of pairwise correlations were negative. Synchrony in TP_{lake} was somewhat higher ($S = 0.20$). Correlations of TP_{lake} among lakes were more likely to be positive (71%) than negative (29%). TOC_{lake} had a similar degree of synchrony to TP_{lake} ($S = 0.23$), and 72% of lakes had positive correlations. $\log_{10}\text{DIN:TP}_{\text{lake}}$ had low synchrony ($S = 0.08$) resulting from the low synchrony in DIN_{lake} .

Although annual $\log_{10}\text{TP}_{\text{lake}}$ and $\log_{10}\text{TOC}_{\text{lake}}$ across the entire data set (including all sites and all years) were significantly correlated ($r = 0.55$, $p < 0.0001$; Figure 5a), the average of the correlations of TOC_{lake} and TP_{lake} within individual lakes was significantly different from 0 (average $r = 0.068$, $p = 0.02$; Figure 5c), but still low, suggesting that while TOC_{lake} and TP_{lake} covary spatially, they were not responding strongly to the same drivers over time. Furthermore, long-term changes in TP_{lake} and TOC_{lake} ($\Delta\text{TP}_{\text{lake}}$ and $\Delta\text{TOC}_{\text{lake}}$) were not significantly correlated ($r = 0.039$, $p = 0.71$; Figure 2). Annual $\log_{10}\text{DIN}_{\text{lake}}$ and $\log_{10}\text{TOC}_{\text{lake}}$ were not correlated across the entire lake data set ($r = 0.04$, $p = 0.17$; Figure 5b) nor were DIN_{lake} and TOC_{lake} correlated within individual lakes (average $r = -0.041$, $p = 0.20$; Figure 5d). The total changes over time ($\Delta\text{DIN}_{\text{lake}}$ and $\Delta\text{TOC}_{\text{lake}}$) across all lakes were also not significantly correlated ($r = -0.20$, $p = 0.055$; Figure 2).

Although synchrony among individual lakes was relatively low for all chemical variables (Figure 4), averages of standardized variables (i.e., z scores) within each N-deposition region behaved very similarly on a year-to-year basis (Figure 6). All pairwise correlations between each region were significant for DIN_{lake} and TP_{lake} (Table 2). Correlations were almost always stronger for regional averages between adjacent regions than distant regions (e.g., low was most strongly correlated with medium, and highest was most strongly

Figure 2. Changes in TP_{lake} , DIN_{lake} , TOC_{lake} , and $\log_{10}(DIN:TP)_{lake}$ over the monitoring period based on the slope of the Thiel-Sen regression line for each lake. Areas of the circles are proportional to changes in concentration. Increases are shown in blue, decreases in red. Sites with significant ($p < 0.05$) Sen's slopes are outlined with dark black lines. Sites with no trend are marked with $\times$.

Figure 3. Density plot showing the distribution of $\log_{10}(\text{DIN:TP})_{\text{lake}}$ in the first and last 5 years of the study period, indicating shifts in nutrient limitation status of Swedish lakes. The red line represents the boundary previously identified as the median percentile dividing N and P limited lakes in Sweden; the gray area represents the 25th and 75th percentiles or the potentially colimited range (Bergström, 2010).

correlated with high); this was particularly true with TOC_{lake} , where all correlations between adjacent regions were highly significant, but correlations between distant regions were nonsignificant (but still positive; Table 2). DIN_{lake} in all regions initially increased until 2003–2005, then decreased consistently for the remainder of the study period (Figure 6). Although absolute declines in DIN_{lake} were greatest in the southeast where DIN deposition was highest, proportional decreases in DIN_{lake} relative to the mean of the first 5 years were greatest in the low deposition region (35.6%), followed by highest (30.4%), medium (24.4%), and high (19.8%). These declines were similar to the proportional declines in DIN_{dep} in each region (low 43%, highest 37%, medium 38%, and high 30%). The correlation between DIN_{lake} and DIN_{dep} was strongest in the northern low deposition region ($r = 0.61$, $p = 0.013$) and positive but not significant in the medium ($r = 0.39$, $p = 0.133$), high ($r = 0.40$, $p = 0.120$), and highest ($r = 0.44$, $p = 0.088$) deposition regions.

There was a significant decline in mean TP_{lake} z scores across all DIN deposition regions using the Kendall test with region as a blocking variable ($p = 0.041$). Although long-term temporal trends in TP_{lake} were weaker than trends in DIN_{lake} , there was considerable interannual variability, and average z scores in different regions had very similar patterns of variability, resulting in high correlations between regions (Table 2). The most striking example of this is 2007; in this year, TP_{lake} was extremely low in all regions (Figure 6). TOC_{lake} generally declined in all regions until 2003–2004, then increased through the remainder of the study period.

In contrast to DIN_{lake} , $\text{DIN}_{\text{stream}}$ did not decline significantly over the monitoring period (Figure 6). Generally, DIN behaved similarly in streams and lakes, increasing in the first several years of the monitoring period and declining after 2005–2006, but the trends diverged in the last several years of the study period (Figure 6). In the highest deposition region, $\text{DIN}_{\text{stream}}$ behaved differently from other regions or from lakes. $\text{TP}_{\text{stream}}$ behaved similarly to TP_{lake} , with the highest TP in 2005 in most regions, and the lowest in 2007. DIN:TP

Figure 4. Histogram of between-lake pairwise correlations of summer DIN_{lake} , TP_{lake} , TOC_{lake} , and $\log_{10}(\text{DIN:TP})_{\text{lake}}$ for all lakes. Average correlations ($\bar{r}$) for each variable are shown as thick black lines. Positive correlations are pictured in orange, negative concentrations in blue.

Figure 5. Top: Scatterplots of (a) summer mean TOC_{lake} (mg L^{-1}) versus TP_{lake} ($\mu\text{g L}^{-1}$) and (b) TOC_{lake} (mg L^{-1}) versus DIN_{lake} ($\mu\text{g L}^{-1}$). Each point represents a mean summer concentration for a single lake in a single year. Bottom: Histograms showing all correlations of (c) $\text{TOC}_{\text{lake}} \times \text{TP}_{\text{lake}}$ and (d) $\text{TOC}_{\text{lake}} \times \text{DIN}_{\text{lake}}$ over time within individual lakes.

had a similar temporal pattern in streams as in lakes in the low, medium, and high deposition regions (rising initially until 2003–2005, then falling) but did not decline as steeply at the end of the study period in the streams. In the highest deposition region the trend in $\text{DIN:TP}_{\text{stream}}$ was very different from the other regions, and $\text{DIN:TP}_{\text{stream}}$ was highly variable and increasing. TOC appeared to follow similar patterns in streams and lakes, with low or declining concentrations in the first few years followed by increasing concentrations for most of the study period. Average z scores of DIN, TP, and DIN:TP were highly significantly correlated between streams and lakes on an annual basis in the low deposition region (Figure 6 and Table 3). However, average annual z scores of DIN and TP were generally only weakly correlated between lakes and streams in other regions. In the highest deposition region, average annual z scores of DIN were uncorrelated between streams and lakes ($r = -0.127$). For TOC, average annual z scores were most highly correlated between lakes and streams in the medium and highest deposition regions (cf. Table 3). Trends in spring (April–June) data were similar to summer trends for all stream variables and for TP_{lake} and TOC_{lake} , but DIN_{lake} and $\text{DIN:TP}_{\text{lake}}$ did not decline at the end of the monitoring period during the spring as they did during the summer (supporting information Figure S1). However, DIN_{lake} concentrations were much higher and more variable during the spring, and this variability may have obscured long-term trends. TP_{lake} was very low in 2007 in the spring as well as in the summer data.

The model developed to estimate proportional contribution of direct DIN deposition to the lake surface relative to catchment DIN inputs suggests that for a typical lake in this study (median depth 3.3 m, average DIN deposition $686 \text{ mg N m}^{-2} \text{ yr}^{-1}$, median stream nutrient concentration 25 mg m^{-3} , residence time 50–100 days; Lindström et al., 2006), direct DIN deposition likely accounts for 50–70% of annual DIN inputs

Figure 6. Left column: Regional summer means (July–September) of standardized total phosphorus (TP), dissolved inorganic nitrogen (DIN), total organic carbon (TOC), and log DIN:TP in lakes (grouped by N deposition). Z scores are first calculated from summer averages within each lake and stream across the monitoring period, then averaged for each year across lakes in each region. Right column: Regional spring–summer averages (May–September) for streams in corresponding N-deposition regions. Circles = low deposition, squares = medium, triangles = high, and diamonds = highest.

and between 10 and 20% of TN inputs (excluding N fixation and sediment loading). The importance of direct DIN deposition increases with lake residence time and is greater for shallow than for deep lakes (Figure 7).

MAT ranged from 0.86°C in the northern low deposition region, to 7.12°C in the southern highest deposition region. MAP did not have such a strong regional gradient and ranged from 2.1 mm d⁻¹ in the medium deposition region to 2.7 mm d⁻¹ in the highest deposition region. Although there is a strong air temperature gradient from southern to northern Sweden, MAT was highly synchronous throughout Sweden, and warmer years in one area were warm everywhere ($S = 0.90$; Figure 8). MAP was much more variable than MAT

Table 2

Pairwise Correlations Between DIN Deposition Regions of Average z Scores for Summer DIN_{lake} , TP_{lake} , and TOC_{lake}

		Medium	High	Highest
DIN_{lake}	Low	0.717**	0.682**	0.558*
	Medium		0.593*	0.636**
	High			0.704**
TP_{lake}	Low	0.786***	0.673**	0.530*
	Medium		0.677**	0.721**
	High			0.771***
TOC_{lake}	Low	0.752***	0.559*	0.171
	Medium		0.759***	0.463
	High			0.690**

* $p < 0.05$. ** $p < 0.01$. *** $p < 0.001$.

($S = 0.50$); however, lakes generally followed similar patterns of wet and dry years (Figure 8). Although air temperatures have been increasing in Scandinavia in recent decades (Kivinen et al., 2017), there was no clear temporal trend in MAT or MAP during the 16 year study period; however, there was considerable interannual variability. Neither MAT nor MAP was significantly correlated with regional means of DIN_{lake} or TP_{lake} in any of the DIN deposition regions.

4. Discussion

Declining $DIN:TP_{lake}$ was observed throughout Sweden, indicating that N limitation in Sweden is becoming more widespread and severe (Figures 2, 3, and 6). This finding supports previous research suggesting that DIN deposition may alter lake N:P stoichiometry (Elser et al., 2009; Hessen, 2013). However, whereas previous studies using

space-for-time approaches highlighted the role of DIN deposition in southern Sweden (Bergström et al., 2005; Elser et al., 2009), using time series data, we found the strongest effect of declining DIN deposition in northern Sweden where deposition rates have been historically relatively low. In addition to overall declines in $DIN:TP_{lake}$, we also found a surprisingly high level of synchrony in year-to-year variability in all of the chemical variables when considered at the regional scale (Figure 6 and Table 2). This synchrony suggests that lake stoichiometry is being partially controlled by climatic and environmental drivers operating at large spatial scales.

There was considerable variability in summer concentrations of chemical variables at the scale of individual lakes ($S < 0.3$ for all chemical variables; Figure 4). Variability observed at the scale of individual lakes may be the result of either catchment or lake properties. Catchment variability was likely important in filtering (cf. Hessen et al., 2009) the effects of changes to mass inputs such as DIN_{dep} (which is known to be retained efficiently by many terrestrial ecosystems) (Lucas et al., 2016) and runoff (which is highly dependent on catchment size and composition) (Blenckner, 2005; Leavitt et al., 2009; Vogt et al., 2011). Changes in energy inputs, such as increasing air temperatures, are expected to have more synchronous impacts on lakes (Read et al., 2014); however, the impacts of energy fluxes on chemical variables are mediated by individual lake morphometry and ecological status (Arnott et al., 2003). Hence, the high variability of DIN_{lake} , TP_{lake} , and TOC_{lake} at the scale of individual lakes in this study was likely the result of heterogeneity in both catchment and lake properties (Figure 4 and Table 1).

Despite the relative lack of synchrony at the individual lake scale, DIN_{lake} , TP_{lake} , TOC_{lake} , and $DIN:TP_{lake}$ all behaved synchronously at the scale of the four DIN deposition regions (Figure 6 and Table 2). The coherent behavior of nutrients at the regional scale suggests that although there are many lake- and catchment-specific differences, there seems to be an underlying signal from broad-scale environmental drivers such as climate or DIN deposition that lakes are responding to at the landscape scale. Geographic patterns of air temperature, precipitation, and acidic deposition further follow a similar north-south and east-west gradients to DIN deposition (Figure 1) (Weyhenmeyer, 2008). The fact that for all variables trends were almost always more highly correlated with neighboring regions than with distant regions (Table 2) further reinforces the conclusion that broad-scale drivers were responsible for the observed trends.

DIN_{lake} declined throughout Sweden (Figures 2 and 6) as has been observed previously (Weyhenmeyer et al., 2007), and declining DIN_{dep} probably contributed to this trend (Figure 1). In absolute numbers, DIN_{lake} declines were greatest in the highest deposition region (Figure 2), which also had the highest average absolute decline in DIN_{dep} ($-32 \text{ mg N m}^{-2} \text{ yr}^{-1}$). However, the proportional declines in DIN_{lake} were greatest in the low deposition region and smallest in the southeastern high deposition region. DIN_{dep} had a similar geographic pattern, with the greatest proportional declines in the low deposition region and smallest proportional declines in the high deposition region. Although the magnitude of proportional declines

Table 3

Pearson Product-Moment Correlations Between Average z Scores of Lake and Stream Nutrients in the Four DIN Deposition Regions

	DIN	TP	TOC	$\log_{10}(DIN:TP)$
Low	0.802***	0.805***	0.478*	0.757***
Medium	0.349	0.374	0.796***	0.520*
High	0.611*	0.425	0.340	0.713**
Highest	-0.127	0.487*	0.537*	0.045

* $p < 0.1$. ** $p < 0.05$. *** $p < 0.01$. **** $p < 0.001$.

Figure 7. Proportional contribution of direct deposition to the lake surface relative to estimated catchment inputs for dissolved inorganic nitrogen (DIN) (top) and total N (bottom). Numbers in the plot refer to different lake depths (m). The larger red circles represent the median site depth from the lakes used in this study (3.3 m). The model assumes constant DIN deposition ($686 \text{ mg m}^{-2} \text{ yr}^{-1}$) and constant stream nutrient concentrations ($25 \text{ mg m}^{-3} \text{ yr}^{-1}$).

in DIN_{lake} and DIN_{dep} were similar throughout Sweden, annual DIN_{dep} was only significantly correlated with DIN_{lake} in the low deposition region.

The lack of significant correlations of DIN_{dep} with DIN_{lake} in medium, high, and highest deposition regions at the annual scale is likely the result of different catchment characteristics, which can strongly influence N loading (Crowley et al., 2012; de Wit et al., 2008; Hu et al., 2014). Boreal forests, which are usually N limited, retain DIN efficiently, although some added DIN may be subsequently released as dissolved organic nitrogen (DON) later (Lucas et al., 2016; Sponseller et al., 2017); this may mask the annual signal of DIN deposition. Many lakes in the low deposition region were in mountainous areas or in the sub-arctic, so surrounding catchments contained less forest cover than in other regions (Table 1); this may help to explain the stronger response of the northern lakes to changes in DIN deposition. The signal of reduced deposition in the southern regions may also be obscured by increased DON loading in areas where browning has occurred. In boreal catchments in Scandinavia, 74–95% of TN may be associated with DOM (Kortelainen et al., 2006; Sponseller et al., 2014); if even a relatively small fraction of this organic N is mineralized to DIN, this may be sufficient to mask the signal from DIN deposition. Increasing TOC_{lake} may also promote denitrification in lakes with low C:N_{lake} ratios (Weyhenmeyer & Jeppesen, 2009); however, the spatial patterns of changes of TOC_{lake} and DIN_{lake} are very different (Figures 2 and 6), particularly in the north where the largest DIN_{lake} declines occurred but where TOC_{lake} was unchanged or decreasing. Temporal patterns were also different, and we saw no significant correlation of DIN_{lake} and TOC_{lake} within individual lakes (Figure 5). Taken together, the similar long-term trajectories and proportional declines of DIN_{dep} and DIN_{lake} support hypothesis 1 (declining DIN_{dep} was a contributor to declining DIN_{lake}) although the effects of DIN_{dep} were

spatially variable and likely modified by catchment processes and lake morphometry for individual lake sites.

$\text{DIN}_{\text{stream}}$ followed similar temporal patterns to DIN_{lake} in some regions (Figure 6 and Table 3) but generally declined less than DIN_{lake} over the monitoring period (although declining DIN has been observed in northern Swedish streams in previous studies over longer time scales; Lucas et al., 2016). As with DIN_{dep} , $\text{DIN}_{\text{stream}}$ was

Figure 8. Box plots showing relative changes in mean annual temperature (MAT) and mean annual daily precipitation (MAP) at all 92 lake locations. MAT and MAP are standardized to z scores at each site before plotting. Water year (October–September) is used to calculate MAT and MAP.

most highly correlated with DIN_{lake} in the low deposition area ($r = 0.80$), suggesting that there is a closer coupling between DIN_{depr} , $\text{DIN}_{\text{stream}}$, and DIN_{lake} in northern than southern areas of Sweden. By contrast, in the highest deposition region, $\text{DIN}_{\text{stream}}$ and DIN_{lake} were completely uncorrelated (Table 2). Where $\text{DIN}_{\text{stream}}$ did decline, these declines were less dramatic than the declines in DIN_{lake} (Figure 6). Several factors may help to explain greater declines in DIN_{lake} relative to $\text{DIN}_{\text{stream}}$. Lakes may respond to changes in DIN deposition more directly than streams because higher fractions of the DIN loads are delivered to lakes by direct deposition to the lake surface. Our model (Figure 7) suggests that for a typical Swedish lake experiencing average DIN deposition and stream nutrient concentrations and a residence time of 50–100 days, direct DIN deposition may account for as much as 53–69% of annual DIN inputs. Direct DIN deposition accounts for a substantially lower fraction of TN inputs (9–16%, similar to the proportion observed in Canham et al., 2012), but much of the organic N pool is likely unavailable (Stepanauskas et al., 2000). Even assuming that 25% of stream DON may be bioavailable (Stepanauskas et al., 2000), DIN deposition would still account for 28–44% of bioavailable N inputs. In contrast, essentially all nutrients delivered to streams must be filtered through the catchment.

In-lake processes may also contribute to the larger declines in DIN_{lake} relative to $\text{DIN}_{\text{stream}}$. Previous studies have found that warming increases denitrification rates in Swedish lakes directly (Myrstener et al., 2016), which combined with browning may stimulate denitrification and further reduce DIN_{lake} (Weyhenmeyer & Jeppesen, 2009). Increased production may also reduce DIN_{lake} as a result of phytoplankton and bacterial uptake. Hence, while some of the interannual variability in DIN_{lake} can be attributed to stream inputs, it is likely that direct DIN deposition and in-lake processes further modified DIN_{lake} concentrations. The relatively greater declines in DIN_{lake} during summer than during spring suggest that in-lake removal processes during the growing season may have also contributed to the observed declines.

While the declines in DIN_{lake} can be at least partly attributed to declining DIN deposition, the drivers of interannual variability in TP_{lake} were less clear. Surprisingly, $\Delta\text{TP}_{\text{lake}}$ was not correlated with $\Delta\text{TOC}_{\text{lake}}$ ($p = 0.79$; Figure 2). This result did not support hypothesis 2 (changes in TP_{lake} would be correlated with changes in TOC_{lake}). Although TP_{lake} and TOC_{lake} covary spatially (Figure 5a), when examined within individual lakes temporal trends in TP_{lake} and TOC_{lake} were not strongly correlated (Figure 5c) suggesting that lake browning is not a dominant driver of changes in TP_{lake} in Swedish lakes. It is surprising that TP_{lake} and TOC_{lake} dynamics appear to be uncoupled, given that a previous study found that TOC_{lake} export explains 61–73% of the variance in TP_{lake} (Kortelainen et al., 2006), and other studies have suggested a link between changes in TP_{lake} and TOC_{lake} in Sweden (Huser et al., 2018). It is likely that changing climatic conditions have affected hydrological properties of catchments and as a result the mobility of terrestrial P, which is closely linked to soil moisture (Macrae et al., 2005). Enhanced forest growth and catchment greening as a result of warming may reduce P export by increasing forest demands for P as well as for N, which has been demonstrated previously (Finstad et al., 2016; Lucas et al., 2016). It is also possible that declining DIN in N-limited lakes may reduce phytoplankton biomass (Deininger et al., 2017), resulting in decreased particulate P and TP. Changes in atmospheric P deposition may also be important, as has been suggested in the United States (Stoddard et al., 2016), although a recent study identified no significant relationship between modeled atmospheric P deposition and lake P concentrations in Swedish lakes (Huser et al., 2018).

Although the declines in TP_{lake} were less widespread than the declines in DIN_{lake} , the TP_{lake} signal was more synchronous among lakes than the DIN_{lake} signal both at the scale of individual lakes (Figure 4) and at the scale of regions (Table 2), suggesting that broad-scale forcing variables (which may include other climate-related or deposition drivers not tested) were impacting the year-to-year variability in TP_{lake} . Although there were no significant correlations between climate forcing variables (MAT or MAP) and lake chemistry variables, annual averages are rough metrics and may mask important changes at shorter time scales (e.g., seasonal; Palmer et al., 2014, or episodic extreme events; Jennings et al., 2012; Strock et al., 2016). The anomalously low TP in 2007 in lakes and streams throughout Sweden (Figure 6) coincided with the water year with the highest MAT of any year in the study period (Figure 8), as a result of extremely warm temperatures during the fall and winter months. Indeed, most of Europe experienced extremely warm temperatures from fall 2006 to winter 2007 (Luterbacher et al., 2007; Yiou et al., 2007), which might have been an important climate driver contributing to very low soluble reactive P in 2007 noted in several large lakes in the southern Alps (Salmaso et al., 2013). Interestingly, those lakes had similar trends in P concentrations in surrounding years to those observed in regional averages presented here, and interannual variability in that study was

attributed to broad-scale climatic indices affecting winter weather conditions. The hypothesis that winter conditions may be partly responsible for trends in summer TP_{lake} is supported by the data from the spring months, which are similar to summer data and also have low TP_{lake} in all regions in 2007 (supporting information Figure S1). The similarity between spring and summer data suggests that the observed patterns are not driven by summer growing season processes (e.g., decreasing production or changes to internal loading). Winter conditions have been shown to affect plankton and nutrient dynamics in subsequent growing seasons (Hampton et al., 2016; Weyhenmeyer, 2004), and warm winters have been found to decrease lake TP in subsequent years (Blenckner et al., 2007). Further analysis over a longer monitoring period is required to understand the effects of warm winters on TP_{lake} ; however, if this is important, it suggests that TP_{lake} in northern lakes may be expected to decline with continued climate warming. Several previous studies have observed declining TP in forested boreal catchments in North America and Europe (Arvola et al., 2010; Crossman et al., 2016; Eimers et al., 2009; Huser et al., 2018; Stammer et al., 2017), suggesting that the patterns observed in Sweden may be broadly relevant, although contrasting patterns have been observed in the United States (excluding Alaska and Hawaii) where TP in lakes and streams in relatively undisturbed catchments has increased in recent years (Stoddard et al., 2016).

The significant decline in $DIN:TP_{lake}$ throughout Sweden, resulting from declining DIN_{lake} coupled with only modest declines in TP_{lake} , indicates that N limitation in phytoplankton is increasing throughout Sweden (Bergström, 2010) (Figures 2 and 3). This generally supports hypothesis 3 ($DIN:TP_{lake}$ would decline in areas with reduced DIN_{dep} and increased TOC_{lake}), although browning had less effect on the results than we hypothesized. In regions that are already N deficient, particularly in northern Sweden (Bergström et al., 2008), declining DIN_{lake} and $DIN:TP_{lake}$ are therefore contributing to an intensified N limitation, potentially reducing pelagic primary production and biomass accumulation at higher trophic levels in northern lakes (Deininger et al., 2017; Elser et al., 2010). At the same time, reduced DIN loadings could lower phytoplankton (i.e., seston) N:P ratios and increase their quality as prey items especially for P demanding crustacean zooplankton (Hessen et al., 2013). This change in phytoplankton N:P stoichiometry should be of especial relevance in regions with initially P-limited phytoplankton, particularly southern Sweden (Bergström et al., 2008), where declining $DIN:TP_{lake}$ may shift the nutrient limitation status from P toward NP or N limitation. A shift toward N limitation may also promote increases in cyanobacteria populations (Downing et al., 2001), particularly if it is accompanied by significant temperature increases (Paerl & Huisman, 2008), reducing food quality for upper trophic levels (Brett & Müller-Navarra, 1997) and increasing the risk of potential toxic effects (Beversdorf et al., 2015). Trends of increasing cyanobacteria have already been observed in some lakes in southern Sweden (Weyhenmeyer, 2001), and it is possible that declining N:P may exacerbate this trend with potential threats for water quality and lake ecosystem production.

It is uncertain how future changes in climate, atmospheric N deposition, catchment greening, and lake browning may result in further alterations of lake N:P in boreal and subarctic areas. The time period of the current study is too short to definitively ascribe observed changes to long-term trends rather than decadal-scale fluctuations. Some factors that may contribute to the observed trends (decreasing DIN_{dep} and possibly winter warming) are likely to continue with continued controls on NO_x emissions and future climate change, whereas it is not clear for how long browning trends will continue following recovery from acidification (Monteith et al., 2007). The reductions in DIN_{dep} experienced in Sweden are similar to those seen in parts of North America (Simpson et al., 2014). Other parts of the world, particularly east Asia, are experiencing large increases in DIN_{dep} (Liu et al., 2013) and so will likely have increases in $DIN:TP_{lake}$. The impacts of DIN_{dep} may also be affected by the form of N in atmospheric loads; NO_3^- and NH_4^+ may be transported differently through catchments (Dillon & Molot, 1990) and have different effects on phytoplankton nutrient limitation (Blomqvist et al., 1994; Ferber et al., 2004). Recently, the proportion of NH_4^+ relative to NO_x in DIN deposition has increased in North America and Europe (Du et al., 2014; Li et al., 2016; Simpson et al., 2014). The impacts of climate change on P transport and accumulation in surface waters in undisturbed boreal and subarctic/mountain areas have received less attention than N transport, in part because P-centric literature has focused primarily on land use-driven eutrophication. The potential link between warm winters and low TP_{lake} observed here and elsewhere may suggest that northern lakes will become more oligotrophic in the absence of other anthropogenic impacts; however, this topic requires substantial future research using longer time series and a variety of methodological approaches. Ultimately, this study suggests that in

Sweden and probably other parts of Scandinavia, N limitation is becoming more severe. In other areas experiencing different trends in climate and DIN deposition, lakes may respond differently. However, both N and P concentration in lakes appear to be influenced by these broad-scale drivers, so it is likely that lake nutrient limitation will respond to these changes coherently at large regional scales.

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