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Long-term declines in stream and river inorganic nitrogen (N) export correspond to forest change

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Abstract. Human activities have exerted a powerful influence on the biogeochemical cycles of nitrogen (N) and carbon (C) and drive changes that can be a challenge to predict given the influence of multiple environmental stressors. This study focused on understanding how land management and climate change have together influenced terrestrial N storage and watershed inorganic N export across boreal and sub-arctic landscapes in northern Sweden. Using long-term discharge and nutrient concentration data that have been collected continuously for over three decades, we calculated the hydrologic inorganic N export from nine watersheds in this region. We found a consistent decline in inorganic N export from 1985 to 2011 over the entire region from both small and large watersheds, despite the absence of any long-term trend in river discharge during this period. The steepest declines in inorganic N export were observed during the growing season, consistent with the hypothesis that observed changes are biologically mediated and are not the result of changes in long-term hydrology. Concurrent with the decrease in inorganic N export, we report sustained increases in terrestrial N accumulation in forest biomass and soils across northern Sweden. Given the close communication of nutrient and energy stores between plants, soils, and waters, our results indicate a regional tightening of the N cycle in an already N-limited environment as a result of changes in forest management and climate-mediated growth increases. Our results are consistent with declining inorganic N efflux previously reported from small headwater streams in other ecosystems and shed new light on the mechanisms controlling these patterns by identifying corresponding shifts in the terrestrial N balance, which have been altered by a combination of management activities and climate change.

Key words: boreal forest; climate-mediated growth increases; forest management; soil N storage; Sweden; terrestrial biogeochemistry; terrestrial N retention.

Introduction

The biogeochemical cycle of nitrogen (N) is coupled to the cycles of other elements through the metabolic pathways that enable life on Earth (Falkowski et al. 2008). For instance, carbon (C) uptake and synthesis by plants is mediated by enzyme systems rich in N, thereby linking C and N biogeochemical cycles in living plants, as well as during the decomposition of detrital tissues. During the past century, human activities have caused the deposition of reactive N to the terrestrial biosphere to increase by three- to fivefold (Galloway et al. 2008). Coincident with N increases, concentrations of CO_2 and mean global surface temperatures have also shown increasing annual trends (IPCC 2007), jointly creating conditions that have far-reaching effects on long-term forest dynamics and biogeochemical cycling worldwide. Understanding how N availability and turnover interacts with other facets of environmental change, including increases in temperature, altered precipitation regimes, and transitions in land management has therefore become an important goal in ecological and environmental research (e.g., Falkowski et al. 2000, Finzi et al. 2011).

Despite global increases in the loading of reactive N (Galloway et al. 2008), several recent observations have revealed long-term declines in inorganic N export from small streams draining forests of the northeastern USA and eastern Canada (Aber et al. 2002, Goodale et al. 2003, 2005, Bernhardt et al. 2005, Huntington 2005, McLauchlan et al. 2007, Kothawala et al. 2011, Bernal et al. 2012, Argerich et al. 2013). In some cases, observed declines in hydrologic export and reductions in N availability to forests have occurred during periods of increasing

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N deposition (Driscoll et al. 2003). Such patterns suggest increases in terrestrial N retention that have been unexpected, particularly for watersheds composed of mature forests, where decreases in the biological demand of late-successional ecosystems are expected to weaken the terrestrial N sink of the landscape (Vitousek and Reiners 1975, Aber et al. 1998).

The goal of this study was to examine long-term trends in terrestrial N storage and retention in high-latitude ecosystems. We focused our work in managed boreal forest and sub-arctic ecosystems in northern Sweden for multiple reasons. First, these areas are some of the very few locations worldwide where biogeochemical data have been consistently collected over decades, with sufficient temporal and spatial resolution to permit ecological inference relevant over a regional scale. Second, data collection in many of these areas commenced prior to the acceleration of acidic deposition in other areas of the world (Stoddard et al. 1999) and the locations of the sites we included are far enough north that the N cycle within these forests have received relatively low and constant levels of atmospheric N deposition over at least the last 30 yrs (Rodhe and Rood 1986, Lucas et al. 2013). Third, management intensity of these forests is increasing to meet rising energy and fiber needs (Egnell et al. 2011), but key knowledge gaps remain regarding N use and retention which need to be filled in order to avoid undesirable consequences for forest production, soil fertility, and water quality (Laudon et al. 2011, Futter et al. 2012). Because forests, soils, and surface waters are closely connected and readily exchange nutrients, water, and energy, we use a watershed approach to quantify terrestrial pools and aquatic fluxes of N (Vitousek and Reiners 1975). This work was motivated by a previous analysis of streams and rivers across Sweden which suggested that hydrologic N export may be declining nationwide, as indicated by decreasing trends in the concentrations of nitrate (NO_3^-) between 1990 and 2010 (Lucas et al. 2013).

Despite substantial work on the controls over terrestrial N cycling (Virtanen 1945, Wijler and Delwiche 1954, Stewart et al. 1967, McElroy et al. 1977, Aber et al. 1989, Schimel and Bennett 2004, Barichivich et al. 2013), there is no generally accepted mechanism to explain observed decreases in stream inorganic N export, although numerous alternatives have been proposed (Bernal et al. 2012, Johnson and Turner 2014). Given concurrent increases in tree biomass (Cory et al. 2013) and soil C storage (Ortiz et al. 2013) across Sweden, we tested two hypotheses linked to terrestrial N retention that may explain observed declines in hydrologic export. The first hypothesis is that increased N storage due to enhanced tree growth has resulted in decreased hydrologic exports of inorganic N. The second, but not mutually exclusive, hypothesis is that declines in hydrologic N export are driven by an accumulation of N in forest soils which exceeds inputs via N deposition and N_2 fixation (Huntington 2005, Bernal et al. 2012). To evaluate these hypotheses, we combined long-term data on stream and river N export, with spatially extensive inventories of forest growth, and soil storage throughout northern Sweden beginning in the early 1980s.

MATERIALS AND METHODS

Stream and river inorganic N export

To best characterize regional variation, we quantified changes in terrestrial N storage and hydrologic export for nine watersheds in northern Sweden that represent a wide range of drainage sizes, elevations, mean annual temperatures, and runoff patterns. Inorganic N concentration data were collected from monitoring stations in these watersheds serviced by the Swedish Environmental Protection Agency (Table 1). The date when data collection began varied between 1967 and 1985 among watersheds (Table 1). In this project, we used concentration data that were collected between 1 January 1985 and 31 December 2011. In order to decrease the influence of

TABLE 1. Characteristics of each watershed used in this study.

Watershed name	Start (yr)	Size (km^2)	Elev. (m)	Lat.	Long.	Forest or natural (%)	Ag. (%)	Urban (%)
Abiskojokk Röda Bron	1982	569	377	68°21′ N	18°46′ E	76	0	0
Övre Lansjärv	1985	1281	79	66°38′ N	22°14′ E	72	1	0
Kukkasjärvi	1985	494	60	66°7′ N	23°19′ E	73	1	0
Ostvik	1985	149	15	64°54′ N	21°4′ E	87	6	0
Vindelälven Maltbrännan	1967	10 622	192	64°35′ N	19°17′ E	79	1	0
Rickleån Robertsfors	1970	1591	50	64°12′ N	20°49′ E	80	4	0
Västersel	1985	1501	50	63°26′ N	18°18′ E	81	3	0
Indalsälven Hammarstrand	1975	23 842	120	63°7′ N	16°21′ E	67	2	0
Ljusnan Funäsdalen	1967	299	629	62°33′ N	12°34′ E	87	1	0

Notes: Start is the year continuous stream nutrient data collection began, elev. is elevation, lat. is latitude, long. is longitude, forest or natural is the percentage of land area in each watershed covered by forests or natural vegetation, ag. is the percentage of land area in each watershed covered by agriculture, urban is the percentage of land area in each watershed covered by urban development. Note that forest/natural vegetation, ag, and urban do not add to 100%. The difference should be considered an other category and is comprised mainly of rock, wetlands, lakes, and tundra.

trends in atmospheric N deposition, all monitoring sites were located in northern Sweden, within the boreal and sub-arctic vegetation zones (Fig. 1). Water samples were collected from the central part of each channel using clean polyethylene bottles, generally at semi-weekly intervals during periods of peak flow (e.g., every two weeks during snow melt) and monthly during periods of normal flow. Samples were transported to a centralized lab facility at the Swedish University of Agricultural Sciences and processed upon arrival. Nitrate (NO_3^--N) and nitrite (NO_2-N) were determined following cadmium reduction on unfiltered samples preserved with H_2SO_4, and are hereafter referred to as NO_3^-. Ammonium (NH_4^+-N) was analyzed on unpreserved samples using the phenate method. Analytical methods for NO_3^-/NO_2^- and NH_4^+ concentrations considered in this study followed protocols as defined by the International Organization for Standardization (ISO 5667-1:2007, ISO 5667-3, ISO 5667-6: 2005).

Daily mean water discharge data were obtained from stage and water velocity measurements at eight out of the nine watersheds from 1985 to 2011. Measurement stations are operated and maintained by the Swedish Meteorological and Hydrological Institute (SMHI). In general, all rivers were gauged at a stable cross-section (i.e., near a bridge) and calibrated using current meters. Sampling locations for discharge and stream water chemistry differed by a few tens or hundreds of meters for all locations except the Vindelälven, where discharge measurements took place 42 km downstream from the water chemistry measurement location. Modeled discharge data are available at the water chemistry measurement location on the Vindelälven from 1999 to 2011 from SMHI, which is what we used in our analysis. In order to estimate discharge at the water chemistry monitoring station located on the Vindelälven from 1985 to 1998, we calculated a correction factor determined from a comparative analysis of measured and modeled discharge from 1999 to 2011. Based on this analysis, discharge data were categorized into 11 different groups (100–200, 200–300, 300–400, 400–500, 500–600, 600–700, 700–800, 800–900, 900–1000, 1000–1100, and >1100 m^3/s), and a correction factor was applied to each group in order to improve data accuracy. Measured discharge data were not available at Rickleån Robertsfors, thus we instead used the modeled discharge data available for 1999–2011. The modeled discharge of Rickleån Robertsfors was carried out by SMHI using S-Hype model (Strömqvist et al. 2012).

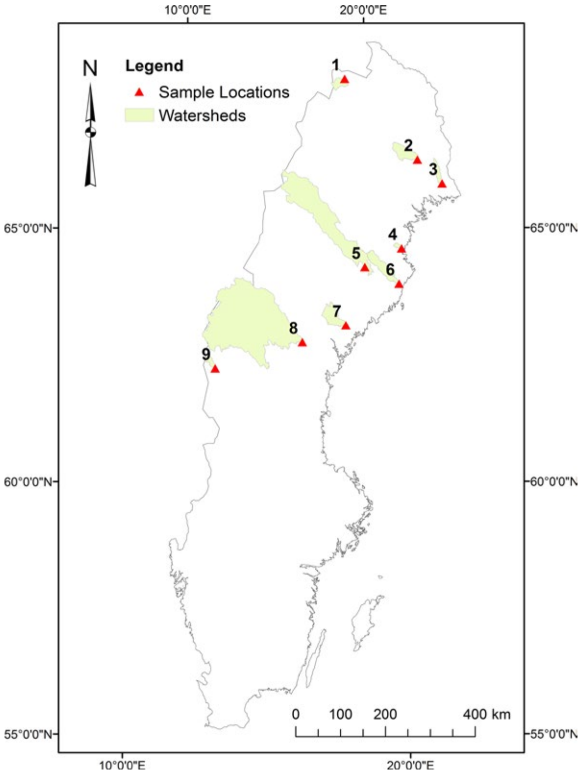

Fig. 1. Map showing locations of the nine watersheds in northern Sweden used in this analysis. Numbers correspond to those listed in Table 1.

Stream inorganic N export was estimated by first calculating the daily flux rate (μg/s) as the product of the sum of the NH_4^+ and NO_3^- concentrations (μg/L) and discharge (L/s) on the day samples were collected. N flux calculated on this day was treated as the mean rate for the time interval between subsequent water sampling events. This daily flux was then multiplied by the number of days within a sampling interval, and each of these intervals within a year were summed to yield an estimate of the annual N flux (kg/yr). Annual inorganic N export was then determined by dividing the flux by the watershed area to yield estimates in kg $N \cdot ha^{-1} \cdot yr^{-1}$.

Tree N storage

To estimate N storage in trees at the watershed scale, we combined tree biomass estimates available from the Swedish National Forest Inventory (NFI; Fridman et al. 2014) with a database of nutrient concentrations contained in various components of Swedish trees (i.e., stems, bark, needles, branches, and stumps; Lucas et al. 2014). The NFI data have been collected to be representative of forests throughout the whole of Sweden since 1923, although we used data beginning in 1953 for this analysis. The modern incarnation of the NFI utilizes approximately 30 000 permanent sampling plots distributed throughout Sweden that were established in 1983, each permanent plot being inventoried every 5–10 yr (Fridman et al. 2014). Plots are located in systematic clusters across the entire country to represent forests on all land area in Sweden (~45 million ha) except the high mountains in the northwest that are not covered with trees (~2.3 million ha) and urban areas (~1.1 million ha). At each sample plot, tree species, diameter at breast height, total height, living and dead tree volumes, age classes, ownership, land use, and other data are collected from circular plots with a 10-m radius. The original purpose of the collected NFI data was to monitor timber production and environmental factors within Sweden. We used NFI data from sample plots located within our nine focal watersheds of interest between the years 1983–2012 to estimate tree N storage at the watershed scale. Biomass estimates of individual tree components (i.e., stems, bark, needles, branches, and stumps) were calculated using single-tree biomass equations by Marklund (1988). There was an average of 1506 plots per watershed, with 7295 plots in the largest watershed (Indalsälven Hammarstrand) and 52 plots in the smallest watershed (Ljusnan Funäsdalen). We used 12 046 plots in total. The NFI does not currently conduct tree measurements in the Abiskojokk Röda Bron watershed because this watershed is situated in the sub-arctic region of the northwest and does support merchantable timber operations. Additionally, we used NFI data to present a general estimate of standing tree volume and the proportion of the landscape that has been clear-cut annually, averaged across the two northern-most counties of Sweden (Norrbotten and Västerbotten) from 1953 to 2012. We expect that the county-scale estimates of tree growth and harvest patterns are directly applicable to each of the individual watersheds, given that each has actively managed forests and has been subject to the standard forestry practices common to northern Sweden.

Concentrations of N in various tree components (i.e., stems, bark, needles, branches, and stumps) were obtained from a database of nutrient concentrations compiled from published values for different tree species in Sweden (Lucas et al. 2014). Tissue N concentrations were selected from the database for inclusion in this study according to the following criteria: (1) values were derived from trees growing in Sweden, (2) at least one concentration was reported for N in any plant tissue component type of *Pinus sylvestris, Picea abies, Betula* spp*.*, or *Pinus contorta*, and (3) trees were not fertilized.

Tree N storage was estimated by multiplying the biomass of a given tree tissue component type (i.e., stems, bark, needles, branches, or stumps) by its corresponding N concentration, summing over all tree tissue component types and then dividing by the watershed size to yield normalized estimates in kg N/ha. We estimated the annual accumulation rate of N in aboveground biomass from the slope of the relationship between N storage in tree biomass and year. The N accumulation rates in tree biomass we report are from >10 000 individual measurements made since 1983 which are representative of the general forest condition in northern Sweden.

Soil N storage

Soil N storage was estimated using data from the Swedish Forest Soil Inventory (SFSI). The SFSI data have been collected annually since 1983 from the same 30 000 permanent sampling plots used for the NFI, but soil data are not available in the nine watersheds used in the present study at the same temporal resolution as the tree data. Starting in 2003, each permanent plot has been inventoried every 5–10 yr (Ortiz et al. 2013). Prior to 2003, soil data are more sparsely distributed within the boundaries of the nine watersheds. Consequently, we analyzed the soil data in aggregate in order to best characterize soil N and C pools in northern Sweden and minimize heterogeneity among samples. Our approach was to pool all SFSI sites located north of approximately 64° N and thus take advantage of additional soil samples collected outside of the nine watersheds included in this study. There were 934 SFSI sites used in this analysis collected between 1983 and 2009, the most recent year data were available at the time of analysis. We confined our analysis to Podzol soils (WRB soil classification system, *Spodosols* under the USDA Soil Taxonomy) from forested sample sites with predominantly mor-type humus because this is the dominant soil type in northern Sweden (Mazhitova 2006) and covers a majority of the land area in our selected watersheds (Table 1). The SFSI samples were collected by field crews over the three decade data collection period, using standardized protocols. The organic soil horizon (the mor layer) was collected by first removing the Oi horizon (i.e., fresh litter, moss, and lichen), and then collecting the entire Oe and Oa horizons (i.e., partially or fully decomposed organic matter), using a 10 cm diameter corer. Up to nine soil cores were bulked together to form a composite sample from each plot, with a target volume of approximately 1.5 L of humus material. In the lab, soil samples were sieved (2.0 mm), homogenized, and dried to a constant weight at 40°C. During the years 1983–1987, total N content was determined by the Kjeldahl method and total C content was determined by wet oxidation using chromic acid. From 1988 onward, total N and C content were analyzed via dry combustion using an elemental analyzer (LECO CNS-1000,LECO, St. Joseph, Missouri, USA) on 0.03–0.5 g of soil depending on the organic matter content. In order to facilitate a robust comparison of C and N data determined using the different analytical methods, an analysis of 2740 archived soil samples was made using dry combustion. This comparison showed that both methods yielded comparable N concentrations, whereas C concentrations were 8% greater using the dry combustion method. The dry combustion bias for C has been previously reported and is well known (Craft et al. 1991). Accordingly, original C concentration estimates made in the years 1983–1987 were corrected using the following expression.

$$C_{\text{corrected}} = -0.0110 \times C^2_{\text{original}} + 1.557 \times C_{\text{original}} - 2.50$$

Soil pools of N and C were calculated based on the dry weight of the organic matter sampled in the O horizon, the corresponding C or N concentration in the organic matter, and the sampling area corresponding with the composite soil samples collected from each plot.

Growing season and climate

In order to quantify a potential change in the length of the growing season, we calculated the number of growing degree days from 1 May to 30 September of each year using 10°C as the base temperature (T_{base}; McMaster and Wilhelm 1997). Growing degree days (gdd) were calculated as: ($gdd = (T_{max} + T_{min})/2 - T_{base}$), where T_{max} is the maximum daily temperature and T_{min} is the minimum daily temperature. Temperature data were obtained from 56 independent meteorological stations that are operated and maintained by the Swedish Meteorological and Hydrological Institute (SMHI). Data are from 1983 to 2011.

Statistical analyses

Rates of change in hydrologic N fluxes, terrestrial N or C pools, stand age, mean annual temperature, growing degree days, and peak date of discharge were estimated from calculations of the slope and the corresponding 95% CIs of the ordinary least squares (OLS) linear regression between the variable of interest and year. All data satisfied the standard parametric assumption required for OLS regression. The significance of long-term trends in annual stream and river discharge was assessed using the non-parametric Mann-Kendall test which is well suited to distinguish between random fluctuations and monotonic trends. Analysis of variance was used to determine significant differences between rates of change of seasonal DIN export. We used $\alpha = 0.05$ to determine if trends or comparisons were significantly different from zero. All statistical calculations were done in R version 2.13.1 (R Development Core Team 2011).

Results

There has been a subtle, yet consistent, decline in the hydrologic export of inorganic N in streams and rivers across northern Sweden from 1985 to 2011. All nine watersheds included in this study display a similar decreasing pattern of inorganic N export (Fig. 2A). The slope of the inorganic N export for individual watersheds ranged from −0.001 to −0.037 $kg \cdot ha^{-1} \cdot yr^{-1}$, being significant at $\alpha = 0.05$ for five out of the nine watersheds and marginally significant for two additional watersheds (Table 2). Treating each watershed as an independent replicate, and thus accounting for expected variation in natural systems, we combined export estimates from all sites, normalized by watershed size, and estimate that the overall annual decrease of inorganic N export is 0.01 $kg \cdot ha^{-1} \cdot yr^{-1}$ (i.e., the slope of the overall trend line including data from all watersheds was −0.01 $kg \cdot ha^{-1} \cdot yr^{-1}$ with lower and upper 95% confidence intervals being respectively −0.003 and −0.012 $kg \cdot ha^{-1} \cdot yr^{-1}$; $P < 0.001$).

The decrease in the inorganic N export did not appear to be caused by dilution or a change in hydrology given that annual discharge from 1985 to 2011 did not exhibit significant long-term temporal trend in any of the nine watersheds included in this study (Appendix S1), indicated by the non-significant estimates of Kendall's tau which provide a distribution-free test of the strength of dependence between discharge and year (Appendix S2). As additional support for the lack of a hydrological effect, hydrographs for each watershed are provided for each watershed (Appendix S3). No discharge data prior to 1985 were available. The decreasing pattern of N export we report may have begun much earlier than 1985, as suggested by the declining pattern of inorganic N concentrations in the three watersheds for which data collection began in 1967 or 1970 (Appendix S4).

Examining hydrologic N exports seasonally, the strongest declines over the 1985–2011 period were observed during the growing season months (June–September). The mean rate of change (i.e., the slope of the overall trend line including data from all watersheds treated as independent replicates) of inorganic N export exhibited a declining trend in all seasons, but the magnitude of decline was significantly greater during growing season (−0.018), when compared to the spring (−0.012) and winter (−0.009; $P < 0.05$; Table 3). Due to the steep decline in inorganic N export during the growing season, the relative proportional rate of change in seasonal export expressed as a fraction of the total annual export actually increased during the winter and spring seasons (Table 3; Appendix S5). These proportional rates of change indicate than an ever declining fraction of the annual N export is derived from the growing season.

Forests in northern Sweden have concurrently shown changes in total biomass and in estimated N content over the last three decades. In eight out of the nine watersheds, the mean stem volume measured on the outside of the bark significantly increased (Fig. 2B). Combining stem volume measurements from each watershed into single regression analysis, the slope of the overall regression line was 1.06 $m^3 \cdot ha^{-1} \cdot yr^{-1}$ (lower and upper 95% CIs were 0.575 and 1.540 $m^3 \cdot ha^{-1} \cdot yr^{-1}$, respectively), indicating a significantly increasing trend in tree biomass. Using the NFI data at a county-scale, we estimate that the increasing trend in tree biomass accumulation began in 1968 (Fig. 3A). More importantly, the estimated storage of N in the tree biomass significantly increased in eight out of the nine watersheds (Fig. 2C). The slope of the overall trend representing the estimated annual N accumulation in tree biomass was 4.53 kg $N \cdot ha^{-1} \cdot yr^{-1}$ (lower and upper 95% CIs were 1.695 and 7.367 kg $N \cdot ha^{-1} \cdot yr^{-1}$, respectively) and was significantly greater than zero (Fig. 2C). No tree biomass data exist for the ninth watershed (Abiskojokk Röda Bron) because this watershed is situated in the alpine region were the NFI does not currently conduct tree measurements.

Over the last decades, forests of northern Sweden have exhibited substantial turnover. The mean stand age in the eight watersheds with NFI data has remained unchanged, with no increase between 1985 and 2012 (Fig. 4; lower and upper 95% CIs surrounding the slope

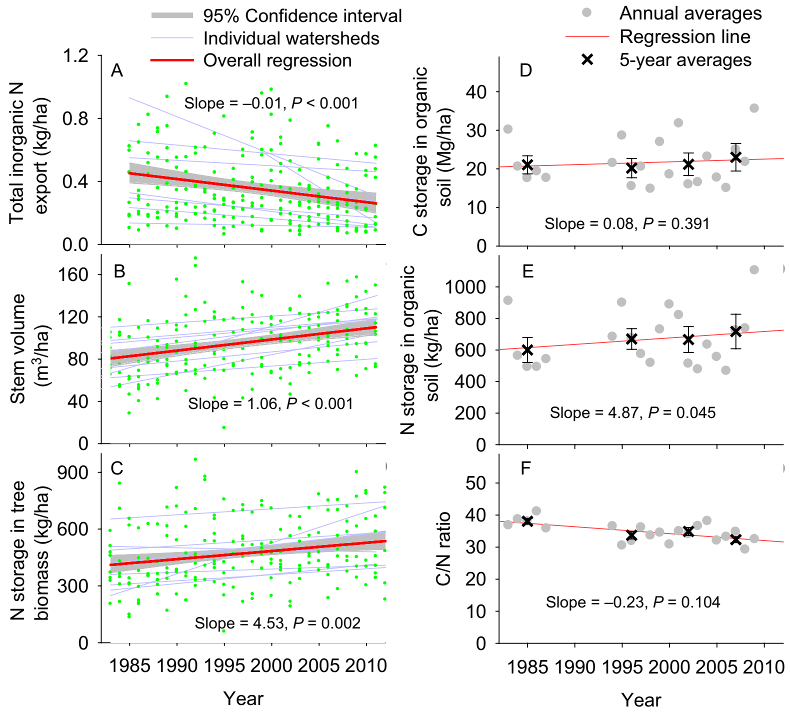

FIG. 2. Long-term trends in northern Sweden. Panels show (A) mean annual hydrological export of inorganic N from nine watersheds in northern Sweden, (B) mean annual stem volume outside the bark in nine watersheds in northern Sweden, (C) mean annual N storage in tree biomass, and (D–F) five-yr mean C storage, N storage, and C/N ratios, respectively in podzols with more decomposed mor-type humus in Sweden north of latitude 64° N. Gray dots in panels (A–C) represent annual estimate for an individual watershed. Gray circles in panels (D–F) represent estimate of means for an individual year aggregating over all available soil collection plots in Sweden north of latitude 64°, black crosses (bars represent standard error) represent group mean of years 1983–1987, 1994–1998, 2000–2004, 2005–2009, respectively and were calculated to reduce the variability in the soil data. Regressions in panels (A–C) were calculated using all data, regressions in panels (D–F) were calculated from group means. 95% confidence intervals for all slope estimates may be smaller than the symbol and are given in the text.

TABLE 2. Estimated annual change in $\mathrm{kg \cdot ha^{-1} \cdot yr^{-1}}$ of the hydrologic inorganic N export from nine watersheds in northern Sweden.

Watershed	Slope	*P*
Abiskojokk Röda Bron	**−0.008**	**0.004**
Indalsälven Hammarstrand	*−0.005*	*0.062*
Kukkasjärvi	**−0.005**	**0.046**
Ljusnan Funäsdalen	−0.007	0.122
Ostvik	−0.003	0.549
Övre Lansjärv	−0.001	0.390
Rickleån Robertsfors	**−0.037**	**0.011**
Västersel	**−0.024**	**0.011**
Vindelälven Maltbränn	**−0.005**	**0.001**

Notes: Slope estimates presented in this table correspond to regression lines for individual watersheds presented in Fig. 2A. Watersheds displayed in bold have a significant decreasing pattern of inorganic N export at $\alpha = 0.05$, watersheds displayed in italics have a marginally significant decreasing pattern.

estimate were −0.296 and 0.220 per yr, respectively). Timber extraction histories for each individual watershed were not available, but can be estimated from the NFI data at a county-level. For the two northern-most counties of Sweden (Norrbotten and Västerbotten), the proportion of land area that has been clear-cut annually between 1978 and 2012 has remained relatively unchanged at 0.7%, decreasing from 1.0% of the land area that was clear-cut between 1953 and 1977 (Fig. 3B).

Soil in northern Sweden has also experienced significant changes over the last three decades. The C pool in the organic horizon showed an increasing, but non-significant trend (Fig. 2D; lower and upper 95% CIs surrounding the slope estimate were −0.226 and 0.380 $\mathrm{Mg\ C \cdot ha^{-1} \cdot yr^{-1}}$, respectively). The N pool in the organic horizon showed a significant increase (Fig. 2E), accumulating N at a rate of 4.87 kg $\mathrm{ha^{-1}\ yr^{-1}}$ (lower and upper 95% CIs were 0.291 and 9.447 kg N $\mathrm{ha^{-1}\ yr^{-1}}$, respectively). Due to the faster accumulation of N relative to C, the C/N ratio of the organic soils showed a declining but non-significant trend, remaining above 30 during the whole period (Fig. 2F; lower and upper 95% CIs of the slope were −0.580 and 0.117 per yr, respectively).

Climate in northern Sweden has also changed over the last three decades. From 1983 to 2011, the mean annual temperature measured from 56 weather stations scattered throughout northern Sweden has increased

TABLE 3. Estimated seasonal rate of change in dissolved inorganic N (DIN) export from nine watersheds in northern Sweden from 1985 to 2011.

Season	Seasonal rate of change of DIN export ($kg \cdot ha^{-1} \cdot yr^{-1}$)	SE	Mean proportional rate of change (% per yr)	SE
Growing	−0.018	0.010	−0.004 a	0.001
Spring	−0.012	0.007	0.002 b	0.001
Winter	−0.009	0.007	0.003 b	0.001

Notes: Rate of change was estimated as the slope of the linear relationship between DIN export (kg/ha) in each season of a given year (using three seasons) or as the relative proportional change of the seasonal DIN export expressed as a percentage of the annual export. Spring is March–May and encapsulates all runoff from snowmelt. Growing season is June–September. Winter is October–February. Seasonal estimates sharing a letter are not significantly different at α = 0.05.

by 1°C, adding on average 0.03°C per yr (Fig. 5A). The number of growing degree days also significantly increased (Fig. 5B), indicating an increase in the cumulative energy received above a 10°C threshold. Additionally, the average date of peak snow melt discharge has shifted nine days earlier from 24 May in 1985 to 15 May in 2011 (Fig. 5C).

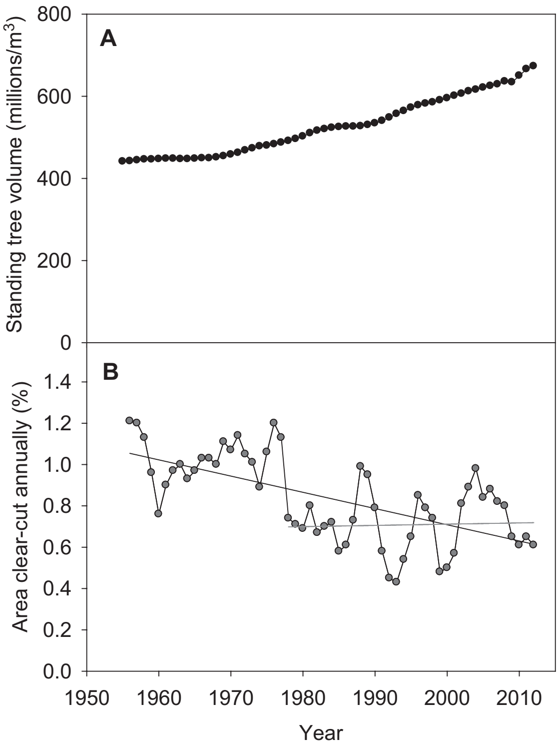

FIG. 3. Summary of all available data from the Swedish National Forest Inventory for the two northernmost counties of Sweden (Norrbotten and Västerbotten) from 1953 to 2011. Panel (A) depicts the estimated standing tree volume on the outside of the bark and (B) represents the proportion of the land area that was clear-felled in a given year. Note that there an was overall decreasing trend in the proportion of land that is clear-cut from 1953 to 2012 (black line), but since 1978 the proportion of the landscape that has been clear-felled remains relatively constant at ~0.7% (light grey line).

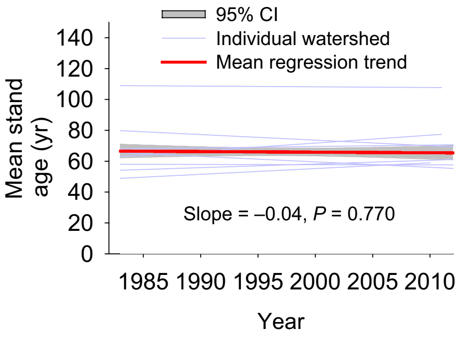

FIG. 4. Mean stand age of forests growing in watersheds of northern Sweden from 1983 to 2012.

DISCUSSION

Results from this study indicate widespread changes in the pools and fluxes of N in northern Swedish landscapes, including declines in the hydrologic export of inorganic N in streams and rivers. There were no directional changes in annual discharge over the three decade period, whereas inorganic N concentrations in stream water decreased, suggesting that greater N retention in terrestrial ecosystems is likely responsible for these trends. Compared to other recent observations of inorganic N declines in streams (e.g., Goodale et al. 2003, Bernhardt et al. 2005), the patterns observed here are unique in that long-term change can be seen for both small and large catchments across the wide geospatial range of northern Sweden (i.e., ~250 000 km^2) which is a region where the N cycle has not been greatly affected by anthropogenic N inputs (Gundale et al. 2011, Lucas et al. 2013). Observed annual declines in hydrologic export of inorganic N varied among catchments and on average (0.01 $kg \cdot ha^{-1} \cdot yr^{-1}$) were of lower magnitude than estimates recently made for Hubbard Brook in the northeastern United States (0.08 $kg \cdot ha^{-1} \cdot yr^{-1}$; Bernal et al. 2012), but were within range of values reported for streams in southern Canada (0.003–0.07 $kg \cdot ha^{-1} \cdot yr^{-1}$; Kothawala et al. 2011).

One mechanism proposed to explain the observed declines of stream inorganic N export in other studies is recovery from historically high rates of anthropogenic N loading (i.e., from N deposition and fertilizer application; Kothawala et al. 2011, Bernal et al. 2012). This mechanism is unlikely to explain the inorganic N export decline in northern Sweden for several reasons. First, the amount of land area covered by agricultural or urban lands in these watersheds was small (average, 2% cover) and is unlikely to have changed significantly through the study period. Further, long-term trends in N export were observed for relatively remote catchments (e.g., Abiskojokk Röda Bron) that are located far from any agricultural activity. Second, atmospheric N deposition in our study region has remained comparatively low (i.e., ~2 kg N·ha^{-1}·yr^{-1}) and relatively constant throughout the three decade study period (Lucas et al. 2013). Third, data from three of the nine watersheds (Ljusnan Funäsdalen, Vindelälven Maltbrännan, and Rickleån Robertsfors) exist as far back as 1967, well before atmospheric N deposition rates across Sweden rose to current levels (Binkley and Högberg 1997, Lucas et al. 2013). The decline in stream and river inorganic N concentration in these three watersheds has been occurring since at least the start of data collection in 1967 and has continued until 2011, despite a rise in atmospheric N deposition rate into the 1980s (Appendix S4). Similar opposing trends in atmospheric N deposition and hydrologic export over time have been described elsewhere (e.g., McLauchlan et al. 2007) and point to changes in terrestrial N cycling processes, not anthropogenic inputs, as likely drivers of long-term watershed output.

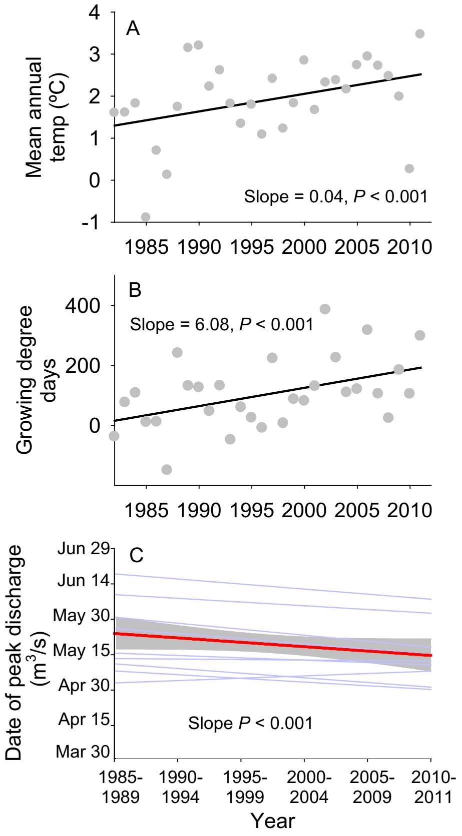

FIG. 5. Historic temperature related data in northern Sweden. Panels show (A) mean annual temperature from 1983 to 2011 calculated from 56 weather stations located throughout northern Sweden, (B) mean number of growing degree days above 10°C from 1 May to 30 September at the 56 weather stations from 1983 to 2011, (C) shift in the mean date of the peak discharge during spring flood from nine watersheds in northern Sweden between 1985 and 2011. Mean dates of peak discharge were calculated for each watershed from daily discharge (m^3/s) data for each of six yearly periods (1985–1989, 1990–1994, 1995–1999, 2000–2004, 2005–2009, and 2010–2011). Trends for each individual watershed, as well as the mean trend reflect means for each yearly period. The Rickleån Robertsfors watershed had only data from 1999 to 2011.

Plant and soil inventory data suggest marked changes in forest ecosystem structure in northern Sweden over the last three decades that are consistent with the idea that terrestrial processes are responsible for observed trends in hydrologic N export. The clearest of these changes is that tree stem volume, an index of forest biomass, has been increasing steadily since the implementation of stricter management guidelines that followed the passage of the 1948 Swedish Forestry Act (Nylund 2009). In support of our first hypothesis, the N accumulated in this forest biomass over time has greatly increased the strength of the terrestrial N sink over the last 50–100 yrs. Our estimate of annual N accumulation in tree biomass (4–5 kg·ha^{-1}·yr^{-1}) is similar to values reported for boreal forests in Finland (~7 kg·ha^{-1}·yr^{-1}; Korhonen et al. 2013), pine forests in Sweden (~4 kg·ha^{-1}·yr^{-1}; Ladanai et al. 2007), aggrading temporal forests in the eastern United States (3–8 kg·ha^{-1}·yr^{-1}; Aber et al. 2002), and could easily account for the observed declines in export from surface waters. Forest management practices likely play a large role in terrestrial N retention as the majority of the Swedish landscape is forested (Cory et al. 2013) and subject to management activities that are well known to exert strong influences on terrestrial nutrient cycles (Löfgren et al. 2009, Lucas et al. 2011). Timber extraction practices have changed in northern Sweden over the past sixty years as proportionally less land area has been clear-cut from 1978 to the present (Fig. 3). Contemporary forest management practices are designed to optimize biomass production, and thereby increase C and N storage, by shortening the time it takes for stands to establish after harvest, establishing stands of fast growing genotypes, establishing fast growing exotics, or by controlling disease (Laudon et al. 2011, Gundale et al. 2014). With higher stand-scale forest production rates in northern Sweden, indicated by an increasing standing stem volume and a decreased harvest rate, while average stand age has remained constant (~60 yr), it is likely that the current forest management strategy has increased N retention due to higher plant N demand (e.g.,Vitousek and Reiners 1975).

While active forest management has likely influenced the long-term changes described here, increased terrestrial sequestration of N, and the subsequent declines in hydrologic N export, may also be linked to changing climate conditions. In fact, long-term declines in inorganic N export from the Abiskojokk Röda Bron catchment, where there is no active forestry, suggest that land management is not a requirement to drive these long-term changes. Northern latitudes are thought to be particularly sensitive to changes in temperature (IPCC 2007), and observed increases in the seasonal amplitude of atmospheric CO_2 over the last sixty years suggest greater rates of C uptake during the growing season at high latitudes (Graven et al. 2013). Other studies have linked recent changes in temperature to longer growing seasons and overall greater rates of net primary production in northern forests (Lucht et al. 2002, Barichivich et al. 2013). Consistent with these observations, our data show an increase in mean annual temperature (by ~1°C), number of growing degree days, and a nine day shift toward earlier peak snowmelt discharge between 1983 and 2011. Together, these patterns indicate that climate-driven changes in plant growth and associated N demand, independent of the land management described previously, could lead to reduced hydrologic losses of this limiting nutrient. This contention is further supported by the observation that the greatest declines in the inorganic N export occurred during the growing season, consistent with a strengthening of the terrestrial N sink that is biologically mediated by increased plant uptake and/or soil retention.

In addition to increased N storage in tree biomass, our results also provide support for our second hypothesis, that reduced hydrologic losses of N in surface waters could be linked to increasing N accumulation and storage in soils. The soil N accumulation rate we estimated for northern Sweden from 1983 to 2009 was approximately 4–5 $kg \cdot ha^{-1} \cdot yr^{-1}$, which could also easily explain the decrease we observed in hydrologic N export. This accumulation rate is approximately two times greater than that reported for other parts of Sweden during this same period (Cory et al. 2013); however, the variance around our estimates suggest a high degree of uncertainty in this number. As suggested previously for trends in plant biomass accrual, the comparatively high rates of soil N accumulation in surface soils may reflect increases in forest productivity and corresponding increases in detrital production that may have resulted from climate change or forest management activities (e.g., Johnson and Curtis 2001). Soil C storage data support the conclusion that a biological driver underlies the observed increase in soil N. We did not observe a significant trend in the average C accumulation rate in the humus layer of soils of the predominantly managed forests in northern Sweden. But a comparison of our estimated C accumulation rate (approximately 80 $kg\ C \cdot ha^{-1} \cdot yr^{-1}$ over 30 yr) with estimates from work primarily focused in southern Sweden (120–250 $kg\ C \cdot ha^{-1} \cdot yr^{-1}$ over the last 40–80 yr; Ågren et al. 2007, Berg et al. 2009) suggests that forest productivity or other biological processes are strong influences on terrestrial N retention and are likely to be even stronger influences in the warmer and more productive southern range of the boreal forest.

It is likely that the forest management practices and climate trends described in this paper interact to shape the observed changes in terrestrial N demand, but our data do not allow us to isolate alternative drivers. Nonetheless, the results presented here illustrate substantial long-term increases in terrestrial N storage across the northern Swedish landscape which corresponds with decreasing hydrologic N exports. In fact, our estimates of the changing pools in plants and soils highlight an apparent growing imbalance in the forest N budget in this region. Assuming that annual N inputs to northern Swedish forests are the combination of atmospheric deposition (~2 $kg\ N \cdot ha^{-1} \cdot yr^{-1}$) and biological N_2-fixation in feather moss carpets (Lindo et al. 2013), our estimates of N accumulation rates suggest an imbalance exists between known N inputs and N accrual in vegetation and soils of ~5 $kg\ N \cdot ha^{-1} \cdot yr^{-1}$. Fertilizer addition to these watersheds do not occur on a large enough scale to account for this deficit (Lindkvist et al. 2011), which suggests either (1) a source of N_2-fixation previously unaccounted for these forest environments, such as free living heterotrophic soil N_2-fixers (Reed et al. 2011) or, more likely, (2) that we are missing the redistribution of N from deeper soil layers beyond our sampling depth. Field observations of a ^{15}N tracer study in a temperate forest suggested that redistribution of N from deeper soil layers could represent an important long-term source of N over forest stand development (Currie et al. 2004). Deep N redistribution could be particularly relevant to forests in Sweden where widespread ditching over the first two-thirds of the 20th century have resulted in many forests growing atop former mires and wetlands (Ecke 2009), areas that have accumulated large stocks of N at the depth of 5–6 m (Malmer and Holm 1984).

If the potential imbalance between supply and accrual is not attenuated by a previously unaccounted for source, our data suggest that N limitation may be incrementally increasing across the region, as predicted by the progressive N limitation hypothesis (Hungate et al. 2003, Luo et al. 2004). As forest productivity is enhanced by silvicultural practices and climate change (i.e., CO_2 increases, climate warming, and longer growing seasons), tree demand for soil N would inevitably increase (Hungate et al. 2003). Higher inorganic N uptake by trees could by itself reduce soil inorganic N concentrations, and thereby account for declining inorganic N stream and river exports. Removal of plant-bound N at final felling could also contribute to the long-term decline in hydrologic inorganic N exports; however, net inputs (i.e., deposition-leaching) during a rotation period are thought to replace the N removed at harvesting of boreal stands (Merila et al. 2014).

Trees may also indirectly affect soil inorganic N concentrations through their effects on soil microbial communities (Alberton et al. 2007). For instance, a high demand for N in N-limited trees leads to greater investments of C into roots and ectomycorrhizal (ECM) associations (Högberg et al. 2008, Lucas and Casper 2008). This could in turn reduce soil inorganic N concentrations by enhancing the microbial N sink (Näsholm et al. 2013), and the build-up of slowly turning over organic N and C pools produced from fungal necromass (Högberg et al. 2011). Although it is possible that increases in water use efficiency could attenuate the strength of the N sink because decreased transpiration could result in decreased N uptake, the terrestrial N sink we report is likely to become further strengthened as the concentration of CO_2 increases in the atmosphere (Näsholm et al. 2013). Enhancements in forest productivity could thus reduce soil inorganic N concentrations, increase N retention in forests, and reduce exports to streams and rivers.

In conclusion, our results show that over a broad geographical scale (i.e., northern Sweden) long-term stream and river inorganic N export has consistently declined over the past three decades. This pattern is likely influenced by a large-scale tightening of the N cycle in an already N-limited environment, in response to increasing terrestrial demands on N resources. These findings highlight the potential for forest management and climate change to exert controls over the storage and retention of N in the landscape – and its subsequent export at ecologically relevant levels to hydrologic environments. Such a relationship could be particularly important for regions where mitigating N exports to vulnerable freshwater and marine habitats is a management priority. In contrast, in high latitude regions not subject to elevated rates of anthropogenic loading, this tightening of the terrestrial N cycle could have negative consequences for freshwater communities and food webs. For example, primary production in lakes in boreal and sub-arctic Sweden is also limited by N availability, particularly during the mid-to-late summer (Bergström et al. 2013). The declines in hydrologic export observed in this study, especially during the growing season, may exacerbate this limitation, with consequences for the productivity of higher trophic levels in aquatic environments. The long-term trends described here have potentially far reaching implications for timber production and water quality in northern regions and underscore the need to better understand the capacity of forest ecosystems to retain nutrients in response to both climate change and management.

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Literature Cited

Aber, J. D., K. J. Nadelhoffer, P. Steudler, and J. M. Melillo. 1989. Nitrogen saturation in northern forest ecosystems. BioScience 39:378–386.

Aber, J. D., et al. 1998. Nitrogen saturation in temperate forest ecosystems: hypothesis revisited. BioScience 48:921–934.

Aber, J. D., S. V. Ollinger, C. T. Driscoll, G. E. Likens, R. T. Holmes, R. J. Freuder, and C. L. Goodale. 2002. Inorganic nitrogen losses from a forested ecosystem in response to physical, chemical, biotic, and climatic perturbations. Ecosystems 5:648–658.

Ågren, G., R. Hyvönen, and T. Nilsson. 2007. Are Swedish forest soils sinks or sources for CO_2–model analyses based on forest inventory data? Biogeochemistry 82:217–227.

Alberton, O., T. W. Kuyper, and A. Gorissen. 2007. Competition for nitrogen between *Pinus sylvestris* and ectomycorrhizal fungi generates potential for negative feedback under elevated CO_2. Plant and Soil 296:159–172.

Argerich, A., et al. 2013. Trends in stream nitrogen concentrations for forested reference catchments across the USA. Environmental Research Letters 8:014039.

Barichivich, J., K. R. Briffa, R. B. Myneni, T. J. Osborn, T. M. Melvin, P. Ciais, S. L. Piao, and C. Tucker. 2013. Large-scale variations in the vegetation growing season and annual cycle of atmospheric CO_2 at high northern latitudes from 1950 to 2011. Global Change Biology 19:3167–3183.

Berg, B., M. B. Johansson, A. Nilsson, P. Gundersen, and L. Norell. 2009. Sequestration of carbon in the humus layer of Swedish forests: direct measurements. Canadian Journal of Forest Research-Revue Canadienne De Recherche Forestiere 39:962–975.

Bergström, A. K., C. Faithfull, D. Karlsson, and J. Karlsson. 2013. Nitrogen deposition and warming: effects on phytoplankton nutrient limitation in subarctic lakes. Global Change Biology 19:2557–2568.

Bernal, S., L. O. Hedin, G. E. Likens, S. Gerber, and D. C. Buso. 2012. Complex response of the forest nitrogen cycle to climate change. Proceedings of the National Academy of Sciences USA 109:3406–3411.

Bernhardt, E. S., et al. 2005. Can't see the forest for the stream? In-stream processing and terrestrial nitrogen exports. BioScience 55:219–230.

Binkley, D., and P. Högberg. 1997. Does atmospheric deposition of nitrogen threaten Swedish forests? Forest Ecology and Management 92:119–152.

Cory, N., P. Nilsson, J. Fridman, and G. Kempe. 2013. An update on the Swedish forests from the National Forest Inventory (in Swedish with English figure captions). Swedish University of Agricultural Sciences, Umeå, Sweden.

Craft, C. B., E. D. Seneca, and S. W. Broome. 1991. Loss on ignition and Kjeldahl digestion for estimating organic carbon and total nitrogen in estuarine marsh soils: calibration with dry combustion. Estuaries 14:175–179.

Currie, W. S., K. J. Nadelhoffer, and J. D. Aber. 2004. Redistributions of ^{15}N highlight turnover and replenishment of mineral soil organic N as a long-term control on forest C balance. Forest Ecology and Management 196:109–127.

Driscoll, C. T., et al. 2003. Nitrogen pollution in the northeastern United States: sources, effects, and management options. BioScience 53:357–374.

Ecke, F. 2009. Drainage ditching at the catchment scale affects water quality and macrophyte occurrence in Swedish lakes. Freshwater Biology 54:119–126.

Egnell, G., H. Laudon, and O. Rosvall. 2011. The role of forest biomass in future energy use: potential and constraints. Forests 2:578–589.

Falkowski, P., et al. 2000. The global carbon cycle: a test of our knowledge of Earth as a system. Science 290:291–296.

Falkowski, P. G., T. Fenchel, and E. F. Delong. 2008. The microbial engines that drive Earth's biogeochemical cycles. Science 320:1034–1039.

Finzi, A. C., J. J. Cole, S. C. Doney, E. A. Holland, and R. B. Jackson. 2011. Research frontiers in the analysis of coupled biogeochemical cycles. Frontiers in Ecology and the Environment 9:74–80.

Fridman, J., S. Holm, M. Nilsson, P. Nilsson, A. H. Ringvall, and G. Ståhl. 2014. Adapting National Forest Inventories to changing requirements: the case of the Swedish National Forest Inventory at the turn of the 20th century. Silva Fennica 48:1095.

Futter, M. N., J. Klaminder, R. W. Lucas, H. Laudon, and S. J. Köhler. 2012. Uncertainty in silicate mineral weathering rate estimates: source partitioning and policy implications. Environmental Research Letters 7:024025.

Galloway, J. N., A. R. Townsend, J. W. Erisman, M. Bekunda, Z. C. Cai, J. R. Freney, L. A. Martinelli, S. P. Seitzinger, and M. A. Sutton. 2008. Transformation of the nitrogen cycle: recent trends, questions, and potential solutions. Science 320:889–892.

Goodale, C. L., J. D. Aber, and P. M. Vitousek. 2003. An unexpected nitrate decline in New Hampshire streams. Ecosystems 6:75–86.

Goodale, C. L., J. D. Aber, P. M. Vitousek, and W. H. McDowell. 2005. Long-term decreases in stream nitrate: successional causes unlikely; possible links to DOC? Ecosystems 8:334–337.

Graven, H. D., et al. 2013. Enhanced seasonal exchange of CO_2 by northern ecosystems since 1960. Science 341:1085–1089.

Gundale, M. J., T. H. Deluca, and A. Nordin. 2011. Bryophytes attenuate anthropogenic nitrogen inputs in boreal forests. Global Change Biology 17:2743–2753.

Gundale, M. J., P. Kardol, M.-C. Nilsson, U. Nilsson, R. W. Lucas, and D. A. Wardle. 2014. Interactions with soil biota shift from negative to positive when a tree species is moved outside its native range. New Phytologist 202:415–421.

Högberg, P., et al. 2008. High temporal resolution tracing of photosynthate carbon from the tree canopy to forest soil microorganisms. New Phytologist 177:220–228.

Högberg, P., C. Johannisson, S. Yarwood, I. Callesen, T. Näsholm, D. D. Myrold, and M. N. Högberg. 2011. Recovery of ectomycorrhiza after 'nitrogen saturation' of a conifer forest. New Phytologist 189:515–525.

Hungate, B. A., J. S. Dukes, M. R. Shaw, Y. Q. Luo, and C. B. Field. 2003. Nitrogen and climate change. Science 302:1512–1513.

Huntington, T. G. 2005. Can nitrogen sequestration explain the unexpected nitrate decline in New Hampshire streams? Ecosystems 8:331–333.

IPCC 2007. Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change *in* [S. Solomon, S. D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor, and H. L. Miller, (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York. NY, USA, 996 pp.

Johnson, D. W., and P. S. Curtis. 2001. Effects of forest management on soil C and N storage: meta analysis. Forest Ecology and Management 140:227–238.

Johnson, D. W., and J. Turner. 2014. Nitrogen budgets of forest ecosystems: a review. Forest Ecology and Management 318:370–379.

Korhonen, J. F. J., et al. 2013. Nitrogen balance of a boreal Scots pine forest. Biogeosciences 10:1083–1095.

Kothawala, D. N., S. A. Watmough, M. N. Futter, L. M. Zhang, and P. J. Dillon. 2011. Stream nitrate responds rapidly to decreasing nitrate deposition. Ecosystems 14:274–286.

Ladanai, S., G. I. Agren, R. Hyvonen, and H. Lundkvist. 2007. Nitrogen budgets for Scots pine and Norway spruce ecosystems 12 and 7 years after the end of long-term fertilisation. Forest Ecology and Management 238: 130–140.

Laudon, H., R. A. Sponseller, R. W. Lucas, M. N. Futter, G. Egnell, K. Bishop, A. Ågren, E. Ring, and P. Högberg. 2011. Consequences of more intensive forestry for the sustainable management of forest soils and waters. Forests 2:243–260.

Lindkvist, A., O. Kardell, and C. Nordlund. 2011. Intensive forestry as progress or decay? An analysis of the debate about forest fertilization in Sweden, 1960–2010. Forests 2:112–146.

Lindo, Z., M. C. Nilsson, and M. J. Gundale. 2013. Bryophyte-cyanobacteria associations as regulators of the northern latitude carbon balance in response to global change. Global Change Biology 19:2022–2035.

Löfgren, S., E. Ring, C. von Brömssen, R. Sørensen, and L. Högbom. 2009. Short-term effects of clear-cutting on the water chemistry of two boreal streams in northern Sweden: a paired catchment study. Ambio 38:347–356.

Lucas, R. W., and B. B. Casper. 2008. Ectomycorrhizal community and ecosystem functioning following simulated atmospheric N deposition. Soil Biology and Biochemistry 40:1662–1669.

Lucas, R. W., J. Klaminder, M. N. Futter, K. H. Bishop, G. Egnell, H. Laudon, and P. Högberg. 2011. A meta-analysis of the effects of nitrogen additions on base cations: implications for plants, soils, and streams. Forest Ecology and Management 262:95–104.

Lucas, R. W., R. A. Sponseller, and H. Laudon. 2013. Controls over base cation concentrations in stream and river waters: a long-term analysis on the role of deposition and climate. Ecosystems 16:707–721.

Lucas, R. W., H. Holmström, and T. Lämås. 2014. Intensive forest harvesting and pools of base cations in forest ecosystems: a modeling study using the Heureka decision support system. Forest Ecology and Management 325:26–36.

Lucht, W., I. C. Prentice, R. B. Myneni, S. Sitch, P. Friedlingstein, W. Cramer, P. Bousquet, W. Buermann, and B. Smith. 2002. Climatic control of the high-latitude vegetation greening trend and Pinatubo effect. Science 296:1687–1689.

Luo, Y., et al. 2004. Progressive nitrogen limitation of ecosystem responses to rising atmospheric carbon dioxide. BioScience 54:731–739.

Malmer, N., and E. Holm. 1984. Variation in the C/N quotient of peat in relation to decomposition rate and age-determination with ^{210}Pb. Oikos 43:171–182.

Marklund, L. G. 1988. Biomass function for pine, spruce and birch in Sweden. Department of Forest Survey, Swedish University of Agricultural Sciences, Umeå, Sweden.

Mazhitova, G. 2006. Soils of the boreal forest. Pages 183–187 *in* W. Chesworth, editor. Encyclopedia of soil science. Springer, Berlin, Germany.

McElroy, M. B., S. C. Wofsy, and Y. L. Yung. 1977. Nitrogen cycle: perterbations due to man and their impact on atmospheric N_2O and O_3. Philosophical Transactions of the Royal Society B 277:159–181.

McLauchlan, K. K., J. M. Craine, W. W. Oswald, P. R. Leavitt, and G. E. Likens. 2007. Changes in nitrogen cycling during the past century in a northern hardwood forest. Proceedings of the National Academy of Sciences USA 104:7466–7470.

McMaster, G. S., and W. Wilhelm. 1997. Growing degree-days: one equation, two interpretations. Agricultural and Forest Meteorology 87:291–300.

Merila, P., K. Mustajarvi, H. S. Helmisaari, S. Hilli, A. J. Lindroos, T. M. Nieminen, P. Nojd, P. Rautio, M. Salemaa, and L. Ukonmaanaho. 2014. Above- and below-ground N stocks in coniferous boreal forests in Finland: implications for sustainability of more intensive biomass utilization. Forest Ecology and Management 311:17–28.

Näsholm, T., P. Högberg, O. Franklin, D. Metcalfe, S. G. Keel, C. Campbell, V. Hurry, S. Linder, and M. N. Högberg. 2013. Are ectomycorrhizal fungi alleviating or aggravating nitrogen limitation of tree growth in boreal forests? New Phytologist 198:214–221.

Nylund, J.-E. 2009. Forestry legislation in Sweden. Report No. 14. The Swedish University of Agricultural Sciences, Uppsala, Sweden.

Ortiz, C. A., J. Liski, A. I. Gärdenäs, A. Lehtonen, M. Lundblad, J. Stendahl, G. I. Ågren, and E. Karltun. 2013. Soil organic carbon stock changes in Swedish forest soils: a comparison of uncertainties and their sources through a national inventory and two simulation models. Ecological Modelling 251:221–231.

R Development Core Team. 2011. R: a language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. www.r-project.org/

Reed, S. C., C. C. Cleveland and A. R. Townsend. 2011. Functional ecology of free-living nitrogen fixation: a contemporary perspective. Pages 489–512 *in* D. J. Futuyma, H. B. Shaffer, and D. Simberloff, editors. Annual review of ecology, evolution, and systematics. Volume 42. Annual Reviews, Palo Alto, California, USA.

Rodhe, H., and M. J. Rood. 1986. Temporal evolution of nitrogen-compounds in Swedish precipitation since 1955. Nature 321:762–764.

Schimel, J. P., and J. Bennett. 2004. Nitrogen mineralization: challenges of a changing paradigm. Ecology 85:591–602.

Stewart, W. D. P., G. P. Fitzgerald, and R. H. Burris. 1967. In situ studis of N_2 fixation using acetylene reduction technique. Proceedings of the National Academy of Sciences USA 58:2071–2078.

Stoddard, J. L., et al. 1999. Regional trends in aquatic recovery from acidification in North America and Europe. Nature 401:575–578.

Strömqvist, J., B. Arheimer, J. Dahne, C. Donnelly, and G. Lindstrom. 2012. Water and nutrient predictions in ungauged basins: set-up and evaluation of a model at the national scale. Hydrological Sciences Journal-Journal Des Sciences Hydrologiques 57:229–247.

Virtanen, A. I. 1945. Symbiotic nitrogen fixation. Nature 155:747–748.

Vitousek, P. M., and W. A. Reiners. 1975. Ecosystem succession and nutrient retention: hypothesis. BioScience 25:376–381.

Wijler, J., and C. C. Delwiche. 1954. Investigations on the denitrifying process in soil. Plant and Soil 5:155–169.

SUPPORTING INFORMATION

Additional supporting information may be found in the online version of this article at http://onlinelibrary.wiley.com/doi/10.1890/14-2413.1/suppinfo

DATA AVAILABILITY

Data associated with this paper have been deposited as follows:
Hydrological data, http://vattenweb.smhi.se/station/;
Concentration data, http://webstar.vatten.slu.se/db.html;
Tree volume data, http://www.slu.se/en/webbtjanster-miljoanalys/forest-statistics/skogsdata/;
Soil data, http://www.slu.se/markinventeringen;
Weather data, http://www.smhi.se/klimatdata.