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Trends in concentrations and export of nitrogen in boreal forest streams

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Temporal trends in inorganic and organic nitrogen (N) export in the stream water between 1979 and 2006 were studied in eight forested headwater catchments in eastern Finland, where an increasing air-temperature trend and a decreasing N-deposition trend has been observed since the 1980s. The Seasonal Kendall test was conducted to study if the stream water N concentrations have changed concurrently and a mixed model regression analysis was used to study which catchment characteristics and hydrometeorological variables were related to the variation in stream water N. The annual concentrations of total organic N (TON) increased at two catchments and the concentrations of nitrate (NO_3-N) and ammonium (NH_4-N) decreased at three and four catchments, respectively. The main factor explaining variation in concentrations and export of N was percentage of peatlands in a catchment. The NH_4-N concentrations were also related to the N deposition, and the exports of NO_3, NH_4, and TON to precipitation. Quantitative changes in both the N concentrations and exports were small. The results suggested relatively small changes in the N concentrations and exports between 1979 and 2006, most probably because the effects of increased air and stream water temperatures largely have been concealed behind the concurrent decrease in N deposition.

Introduction

Excessive export of nitrogen (N) and phosphorus (P) from terrestrial ecosystems enhances eutrophication, which is a prominent environmental problem in inland and coastal waters (Vitousek *et al*. 1997, Bergström *et al*. 2005). In the boreal region, small lakes and streams are particularly sensitive to eutrophication (Schindler 1998). Export of N from forest ecosystems

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is largely controlled by hydrometeorological conditions, atmospheric N deposition, microbial N mineralization and immobilization, N uptake by vegetation and forest management practices (Wright *et al.* 2001, Goodale *et al.* 2002, Gundersen *et al.* 2006, Helliwell *et al.* 2007, Lepistö *et al.* 2008, Dise *et al.* 2009). Typical features for boreal forest soils are low temperatures and pH, leading to low net N mineralization and nitrification (Aber *et al.* 1989). Therefore, the soils are rich in total N, but the supply of plant available N is strongly limited. Consequently, the N cycle is nearly closed in boreal regions with low atmospheric N deposition, resulting in low export into stream waters (Tamm 1991).

In the boreal streams, most of the N is organic (TON), accounting for 50%–95% of the total N (Lepistö *et al.* 1995, Kortelainen *et al.* 1997) and more than 90% of the TON is dissolved (DON) (Mattsson *et al.* 2005, Kortelainen *et al.* 2006). Export of TON to stream waters is largely governed by the proportion of peat soils in the catchments (Pellerin *et al.* 2004, Mattsson *et al.* 2009). The inorganic N fractions, i.e. nitrate (NO_3-N) and ammonium (NH_4-N), are considered to play the most significant role in eutrophication such as promoting algal growth in waters (Andersson 1996), but also DON can be used by plankton (Berman and Bronk 2003, Brookshire *et al.* 2005).

In northern Europe, N deposition has been decreasing since the 1990s (e.g. Wright *et al.* 2001). This has been suggested to be the reason for the decline of dissolved inorganic N concentrations in streams and lakes (*see* Koopmans *et al.* 1995, Rekolainen *et al.* 2005), and the increase in concentrations of dissolved organic matter (TOC and TON) in lakes due to the recovery from acidification (Eikebrokk *et al.* 2004, Vuorenmaa *et al.* 2006), and rivers (Lepistö *et al.* 2008), but conclusive empirical evidence about this is still lacking. On the other hand, de Wit *et al.* (2007) reported not only a decrease but also increases in exports of inorganic N in boreal streams since the 1990s, suggesting that export of N has not been influenced by decreased deposition of N only, but other factors, such as changed hydrometeorological conditions (temperature, precipitation, etc.).

In the boreal region with a distinct winter period, about half of the annual runoff is produced during the spring snowmelt. However, the future winter and spring air temperatures and winter rainfall are predicted to increase, which may change the timing and magnitude of the runoff generation during the spring snowmelt (Bates *et al.* 2008). Concurrently, this may alter the nutrient export, particularly from peatlands with their considerable storages of C and N (Laudon *et al.* 2004, Petrone *et al.* 2007). Furthermore, the increasing temperatures and the increasing frequency of drought periods in summer may increase the mineralization and availability of ammonium (NH_4) and NO_3, which, if not immobilized by soil microbes or taken up by the vegetation, might leach to streams. However, the predictions on the effects of climate change on N export from forested areas are variable and controversial. Kallio *et al.* (1997), Groffman *et al.* (2001), Holmberg *et al.* (2006) and Weslien *et al.* (2009) suggested that a climate change may increase the overall N export, but Bouraoui *et al.* (2004) and de Wit *et al.* (2007) predicted that a change in the seasonal variation in the N export would be the most likely impact. In the study by Sarkkola *et al.* (2009) in eastern Finland, the increase in TOC export during the snow-free period during the latest decades was related to the concurrent increase in stream water temperatures. Since the exports of TOC and TON are strongly correlated (Lepistö *et al.* 2008), increasing temperatures may also increase the TON export.

In boreal forested headwater catchments, forest management operations are, besides the N deposition, the main anthropogenic sources of N in watercourses (e.g. Kortelainen *et al.* 1997). N exports are increased particularly by forest cuttings and soil preparation (Ahtiainen and Huttunen 1999, Laurén *et al.* 2005, Piirainen *et al.* 2007), drainage of peatlands for forestry (Ahtiainen and Huttunen 1999, Åström *et al.* 2002), and N fertilization (Lundin and Bergquist 1985). However, the effects of forest management practices on N leaching were generally of short duration, and stream water N concentrations returned to pre-operation levels within 10 years (e.g. Kubin 1998, Laurén *et al.* 2009).

In the present study, the trends in the export of inorganic and organic nitrogen (N) to stream waters during 1979–2006 were studied in 8 forested headwater catchments in eastern Finland where an increasing air-temperature trend and a decreasing N-deposition trend have been observed since the 1980s (Sarkkola *et al*. 2009). The main objectives were (1) to study if the annual and seasonal concentrations and exports of TON, NO_3-N, and NH_4-N changed concurrently with changed air temperature and N deposition trends, and (2) to identify which hydrometeorological and catchment variables could explain the variation in stream water N concentrations and exports. We hypothesized that because the TOC concentrations in stream water in eastern Finland have been increasing as a result of increasing water temperatures, the TON exports also show an increasing trend. Higher temperatures could also have increased the inorganic N exports, but we expect this effect to have been largely concealed behind the concurrent decrease in inorganic N deposition.

Material and methods

Study catchments

We studied the long-term N export from 8 boreal forested headwater catchments (Table 1) located in a region with similar bedrock and climatic conditions in eastern Finland. A detailed description of the catchments is given elsewhere (Finér *et al*. 1997, Ahtiainen and Huttunen 1999, Sarkkola *et al*. 2009), and only a brief outline of the catchments is presented here. The catchments vary in area from 29 to 494 ha, the proportion of peatland from 8% to 70%, and the mean slope from 0.3% to 1.5% (Table 1). The long-term (1979–2006) mean annual precipitation for the study area is 600 mm, 40% of which falls as snow. The temperature sum (threshold value +5 °C) is 1100 degree days.

The upland soils are weakly developed iron podzols transitioning to peaty podzols and then fibric histosols (peat) in the riparian areas. The parent material of the podzol soils is stony sandy till with a clay content of < 2%. The thickness of the peat layer is mostly (78% of peatland area) < 2 m.

The forests are dominated by Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*), and mixed with silver and downy birch (*Betula pendula* and *B. pubescens*). The peatland areas are mostly forested although stand densities vary considerably and treeless or sparsely stocked areas are common in the Suopuro and Korsukorpi catchments. The upland forests are mainly medium-rich *Vaccinium–Myrtillus* and poor *Empetrum–Myrtillus* types (Cajander 1949), and both minerotrophic and nutrient-poor ombrotrophic peatland types are found in each of the catchments.

Three of the catchments, Liuhapuro, Kangaslampi, and Porkkavaara, were unmanaged during the entire study and during a period of 10 years before starting the monitoring of N export. Five catchments were subjected to management practices, such as clear-cutting, site preparation, and ditch drainage. As in the earlier study by Sarkkola *et al*. (2009), water quality and runoff

Table 1. Characteristics of the study catchments. The mean N content of the ecosystem is the estimated N stored in the vegetation and soil.

Catchment	Area (ha)	Peatland proportion (% of tot. area)	Area of the fertile sites (% of tot. area)	Mean stand volume (m^3 ha^{-1})	Mean N content of the ecosystem (kg ha^{-1})
Kivipuro	54	32	74	88	4100
Liuhapuro	165	48	6	102	5400
Murtopuro	494	50	29	75	5200
Suopuro	113	70	22	30	17900
Välipuro	86	86	65	187	10500
Korsukorpi	69	56	32	61	10400
Porkkavaara	72	16	59	60	3100
Kangaslampi	29	9	20	132	3100

data for a 10-year period after the management practice was removed from the analysis to avoid the impact of management practices.

Runoff monitoring and stream water N analyses

Water level in the outlet stream was recorded continuously with a V-notch weir and daily runoff was calculated on the basis of the stage-discharge relationship. Stream water was sampled 1–5 times per month and more frequently during spring (up to 10 times per month). After 1995 the sampling during December–February was discontinued. In 2000, water samples from the Porkkavaara, Korsukorpi, and Kangaslampi catchments were gathered only in October and November. The stream water temperature was recorded for each water-sampling occasion (for the time series of mean annual runoff, *see* Fig. 1).

The water samples were analysed for total N (TN) and NH_4-N and NO_3-N using unfiltered samples according to National Board of Waters (1981). TN concentration in the water samples was determined colorimetrically after oxidization with $K_2S_2O_8$. NH_4-N was measured by a spectrophotometer, and NO_3-N by the cadmium method. Concentrations of TON were determined as the difference between TN and inorganic N. Since the year 1992, the total organic carbon (TOC) was determined using UV-persulphate oxidation followed by IR (infrared) gas measurements, and thereafter by using high-temperature oxidation.

Mean annual and seasonal N concentrations were calculated by averaging the measured values. In the seasonal data, spring is from March to May, summer from June to August, and autumn from September to November. Winter was excluded from the seasonal analysis due to low sampling frequency after 1995. The N export (kg ha^{-1}) was calculated as the product of the mean monthly N concentration and monthly runoff. In the calculation of the annual N exports, the missing monthly concentrations for December, January and February were interpolated using the measured values from November and March. The average concentrations and exports of inorganic and organic N by catchment are presented in Table 2.

Hydrometeorological and catchment characteristics

In order to analyse the factors affecting the variation in the stream water concentrations and exports of the N fractions, hydrometeorological variables and catchment specific characteristics were used as explanatory variables, which were compiled from different sources. Daily meteorological data were obtained from two weather stations located 10 and 15 km from the catchments. The six hydrometeorological variables used in the models were: annual precipitation (mm), snowfall (mm), snow depth (cm), mean annual air temperature (at 2-m height from the ground level, °C), stream water temperature (°C), and effective temperature sum with a threshold value of +5 °C. The bulk deposition of NO_3-N, NH_4-N, and TN were obtained from the station operated by the Finnish Environment Institute about 50 km northeast from the catchments. The deposition and air-temperature data were analysed in an earlier study, where a significant increase in air temperature and a decrease in N deposition were found (Sarkkola *et al.* 2009 and Fig. 1). Since the end of the 1980s, the TN deposition has decreased from about 8 kg ha^{-1} a^{-1} to 3 kg ha^{-1} a^{-1}.

The following characteristics of the eight catchments were obtained from the field inventories and measurements: stream length (m ha^{-1}), proportion of fertile and poor sites (%), proportion of peatland area (%), proportion of drained peatlands of the catchment area (%), tree-stand volume (m^3 ha^{-1}), proportion of tree species by tree-stand volume (Scots pine, Norway spruce, broadleaf trees, %), and total amount of organic matter in the trees, the peat, and the humus layer (kg ha^{-1}) and the total N content (kg ha^{-1}) in the trees, understorey vegetation and soil. A site was considered to be fertile if it was of the *Vaccinium myrtillus* type or more fertile site type according to the Finnish classifications for mineral-soil sites (Cajander 1949) and peatland sites (Vasander and Laine 2008). The total mass of organic matter in the peat layer, the humus layer, and the growing tree stands were estimated based on field inventories and GIS analysis described in detail by Sarkkola *et al.* (2009). The N stored in the trees and understorey vegetation was estimated using the N concentrations from the study by Finér

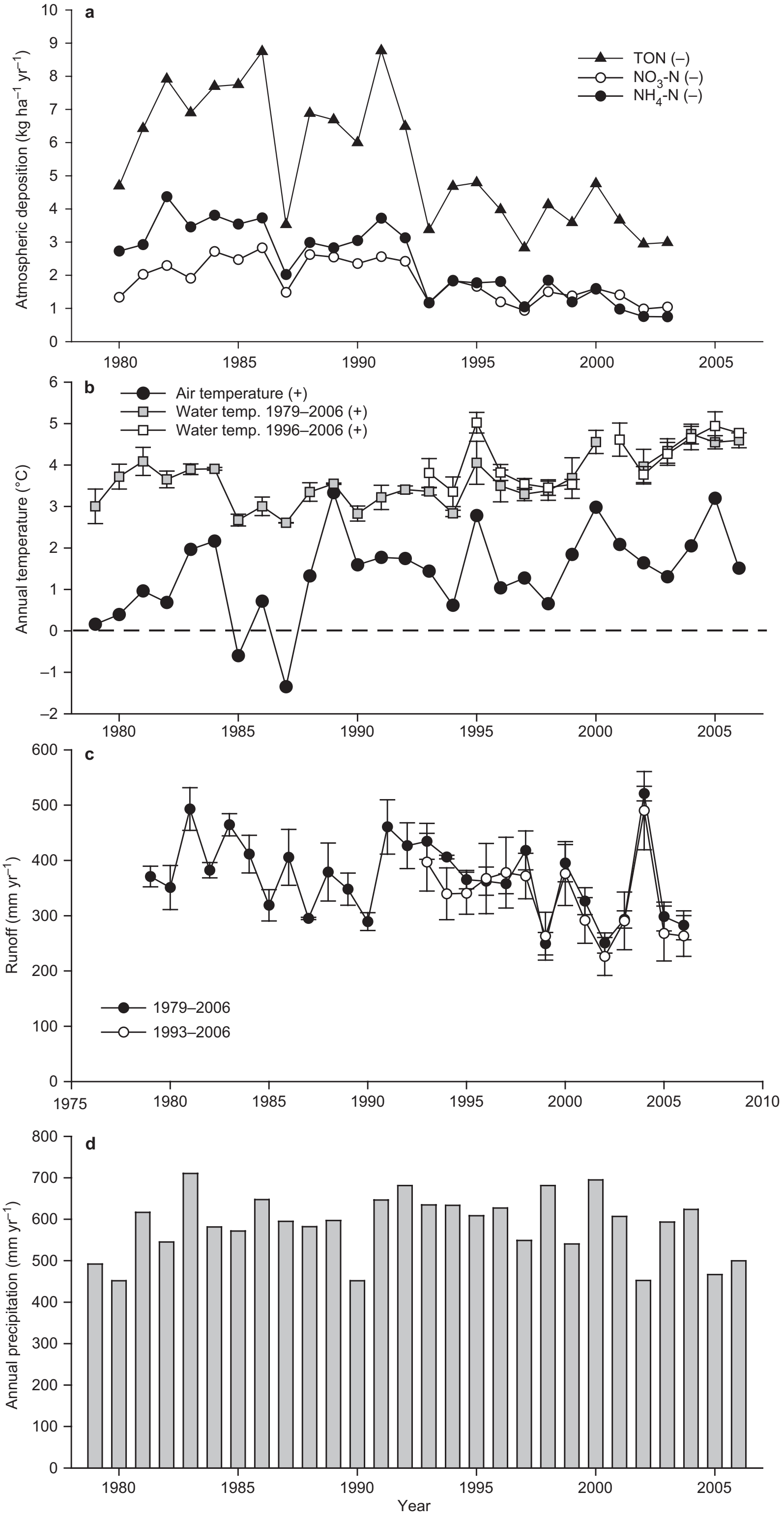

Fig. 1. (**a**) Mean annual nitrogen deposition (direction of the statistically significant trends are in the parentheses), (**b**) mean air temperature and mean water temperature in the streams of the study catchments, (**c**) mean annual runoff from the study catchments, and (**d**) annual precipitation in eastern Finland during the monitoring period 1979–2006 (*see* also Sarkkola *et al.* 2009). The series 1979–2006 contains the temperature and runoff data of the catchments Liuhapuro, Välipuro, Murtopuro, Suopuro and Kivipuro; the series 1993–2006 contains the temperature data of catchment Porkkasalo, Kangaslampi, and Korsukorpi.

Table 2. Mean annual runoff, stream water nitrogen concentrations and exports (TON = total organic nitrogen, TN = total nitrogen, NO_3-N = nitrate nitrogen, and NH_4-N = ammonium nitrogen), and ratio of total organic carbon (TOC) and TON (C/N ratio) in the stream water in the each of the studied 8 catchments during the monitoring years.

Catchment	Years of monitoring	Runoff (mm)	NH_4-N (μg l^{-1})	NO_3-N (μg l^{-1})	TON (μg l^{-1})	TN (μg l^{-1})	C/N ratio	NH_4-N (kg ha^{-1})	NO_3-N (kg ha^{-1})	TON (kg ha^{-1})
Kivipuro	1979–1982, 1996–2006	316.6	7.4	10.3	533.6	548.3	55.7	0.020	0.018	1.647
Liuhapuro	1980–2006	381.4	8.8	16.6	486.3	527.9	56.4	0.026	0.025	1.973
Murtopuro	1979–1982, 1996–2006	412.8	12.0	17.6	388.2	487.2	63.3	0.033	0.059	2.096
Suopuro	1979–1982, 1994–2006	370.2	19.8	9.5	368.3	430.8	65.9	0.030	0.032	1.346
Välipuro	1979–2006	344.1	13.2	19.1	487.4	562.5	65.5	0.028	0.022	1.775
Korsukorpi	1992–2006	400.5	7.5	13.4	298.2	406.4	85.0	0.017	0.030	1.259
Porkkavaara	1996–2006	369.0	3.1	5.7	171.6	217.9	61.4	0.010	0.014	0.729
Kangaslampi	1996–2006	242.6	3.4	4.0	261.5	268.9	51.9	0.008	0.009	0.576

et al. (2003). The N stored in the surface peat layers (0–60 cm) was estimated using the values reported by Finér *et al*. (1997), and for the deeper peat layers (> 60 cm), assuming a bulk density of 120 kg m^{-3} and an N concentration of 1.5% (Laiho and Laine 1994).

Statistical analyses

Time-series analysis

Linear trends in annual and seasonal time series were calculated using the non-parametric Seasonal Kendall test (Gilbert 1996). The slopes of the trends express the median change in the annual and seasonal time series and were calculated using the Sen slope (S) estimation method. A trend was considered significant at $p < 0.05$. The studied time series included concentration and export of TON, NO_3-N, and NH_4-N, as well as the ratio of TOC to TON concentration (C/N ratio). Spearman correlations (r_S) were calculated in order to identify the catchment characteristics that affected the magnitude of the trends (S value) in the concentrations of N fractions.

Models for annual and seasonal N concentration and export

A mixed-model regression analysis was used to identify the variables that controlled the stream water TON, NH_4-N and NO_3-N concentration and export and C/N ratio. The mixed model approach was used in the model construction in order to account for autocorrelation between repeated measurements (*see* also Sarkkola *et al*. 2009). We identified two hierarchical levels of variation in the datasets: (1) between catchments, and (2) within the time series from a catchment. The mixed model had the following form:

$$y_{ij} = \alpha_{ij} + \beta_1 x_{1ij} + \beta_2 x_{2ij} + ... + \beta_n x_{nij} + u_j + e_{ij} \quad (1)$$

where y_{ij} is the response variable (TON, NH_4-N or NO_3-N (μg l^{-1} or kg ha^{-1}) and C/N ratio) for the period i in the catchment j. x_{1ij} … x_{nij} are the explanatory (fixed) variables, α_{ij} is the intercept,

$\beta_1 \ldots \beta_n$ are the parameters of the explanatory variables, u_j is the random effect of the catchment j, e_{ij} is the random error accounting for the within-catchment variation between successive seasonal or annual observations, and n is the number of explanatory variables. The variables u_j and e_{ij} are assumed to be uncorrelated white noise and follow $\sim N(0, \sigma^2)$. The explanatory variables were included in the model as such or their relationship against the nonlinear dependent variable was linearized with logarithmic transformations.

Standard errors of the parameter estimates were used to determine the significance of the parameters. A parameter estimate was judged significant when its absolute value was larger than the estimate of the standard error. The value of –2(log-likelihood) was used to compare the overall goodness-of-fit of the models of increasing number of explanatory variables. The building of the mixed model was conducted in the following way: Firstly, an initial likelihood measure was derived from the model including only the intercept. Thereafter, explanatory variables were one by one added to the model based on the premise that the addition of a variable must maximise the decrease of the likelihood measure. Based on the decrease of the likelihood measure for each explanatory variable, the importance of the variable in explaining concentrations and exports of TON, NO_3-N and NH_4-N, and the CN ratio was ranked. Variables were added to the model until there was no significant improvement in the likelihood measure, or one or more of the explanatory variables becomes non-significant, which is an indication of model over-parameterisation. In order to assure that the order of the added variable did not affect its significance, model versions with different order of the explanatory variables were constructed. The models were estimated with the restricted iterative generalized least-square (RIGLS) method by using MLwiN 2.0 software (Rasbash *et al.* 2001). To evaluate the reliability and accuracy of the model, the systematic error (B) and the relative systematic error (B_r) were calculated for each season as follows:

$$B = \frac{1}{m}\sum_{j=1}^{m}\left[\frac{1}{n_j}\sum_{i=1}^{n_j}\left(y_{ij} - \hat{y}_{ij}\right)\right] \quad (2)$$

$$B_r = \frac{1}{m}\sum_{j=1}^{m}\left[\frac{1}{n_j}\sum_{i=1}^{n_j}\left(\frac{y_{ij} - \hat{y}_{ij}}{y_{ij}}\right)\right] \quad (3)$$

where m is the number of catchments, n_j is the number of observations in catchment j, and $\hat{y}_{ij}$ the predicted value of the response variable at time period i in catchment j.

Results

Trends in stream water N concentrations and export

The mean annual TON concentration averaged over all catchments was 374.4 μg l^{-1} and those of NO_3-N and NH_4-N 12.4 μg l^{-1} and 9.4 μg l^{-1}, respectively (Table 2). The mean annual TON concentrations had increased in two catchments during the monitoring period, and the annual concentrations of NH_4-N decreased in four catchments and the NO_3-N concentrations in three catchments (Table 3 and Figs. 2–4). The mean annual exports of TON, NO_3-N and NH_4-N averaged over all eight catchments were 0.02, 0.03 and 1.4 kg ha^{-1}, respectively (Table 2).

The annual NH_4-N export decreased in three catchments during the study period whereas the TON and NO_3-N exports changed significantly only in one catchment (Table 3 and Fig. 5). The mean annual C/N ratio in stream water was 63 and it increased significantly in three catchments during the study period (Table 3 and Fig. 6). The concentrations of TON were lower in spring than in summer and autumn, but those of NO_3-N and NH_4-N were higher in spring than in the other seasons (Figs. 2–4). There were no clear seasonal differences in the number of significant trends in the concentrations and exports (Table 4).

Factors related to N concentration and export

The trends in stream water TON, NO_3-N and NH_4-N concentrations in the study catchments were correlated with the catchments characteristic only in two cases. The decreasing trend in the NH_4-N concentrations was significantly steeper in the catchments with high peatland percentage

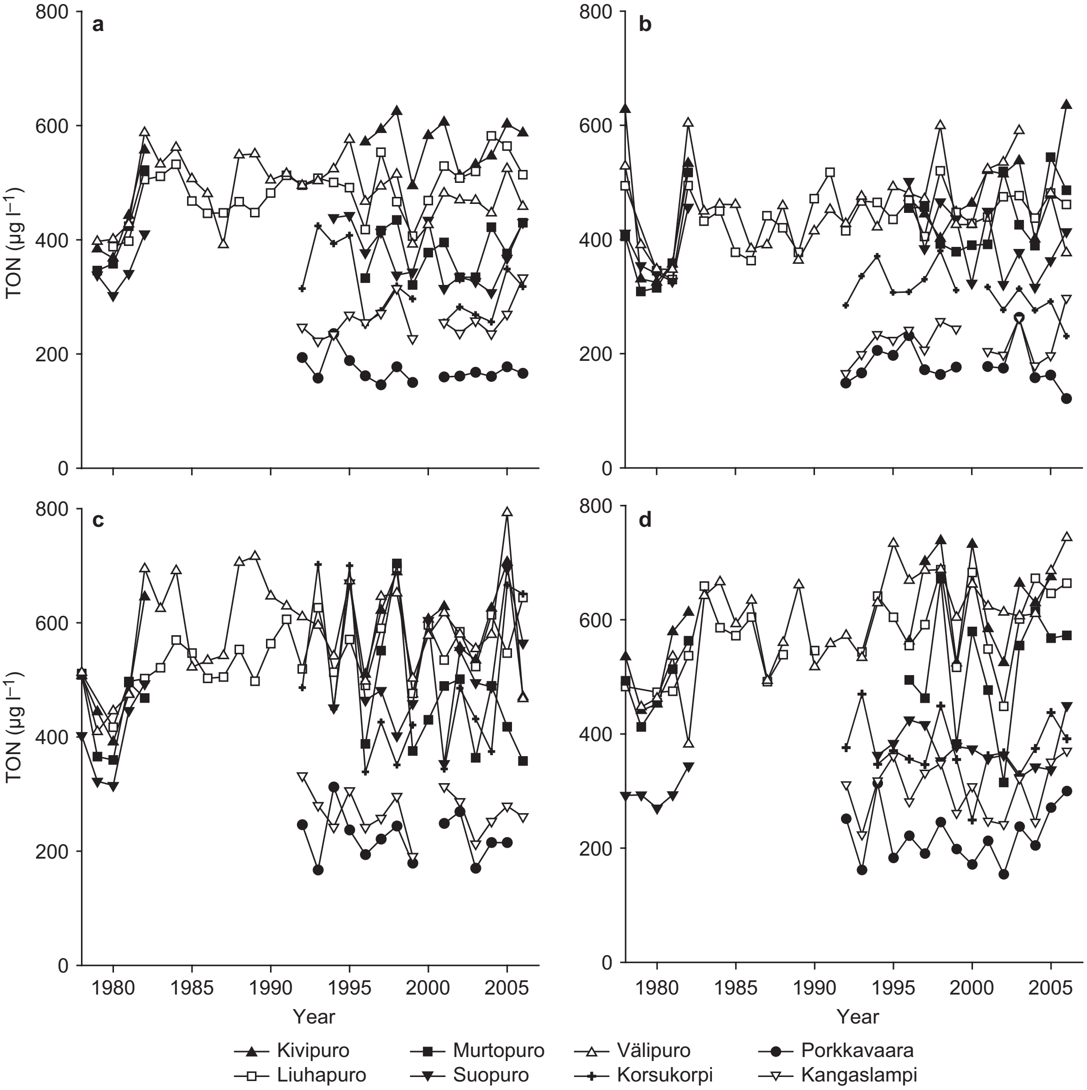

Fig. 2. Time-series for the (**a**) annual, (**b**) spring, (**c**) summer, and (**d**) autumn concentrations of stream water total organic nitrogen (TON) in the study catchments in 1979–2006. The non-continuous series mean that the years affected by forest management operations were removed from the data. Statistically significant trends are presented in Tables 3 and 4.

Table 3. Directions of statistically significant ($p < 0.05$) trends in the annual concentrations and export of NH_4-N, NO_3-N, TON and the C/N ratio in stream water in the studied catchments.

Catchment	NH_4-N		NO_3-N		TON		C/N ratio
	Concentration	Export	Concentration	Export	Concentration	Export	
Kivipuro	–				+		
Liuhapuro	–	–			+	–	
Murtopuro			–				
Suopuro	–	–	–				
Välipuro				+			+
Korsukorpi	–	–					+
Porkkavaara							+
Kangaslampi			–				

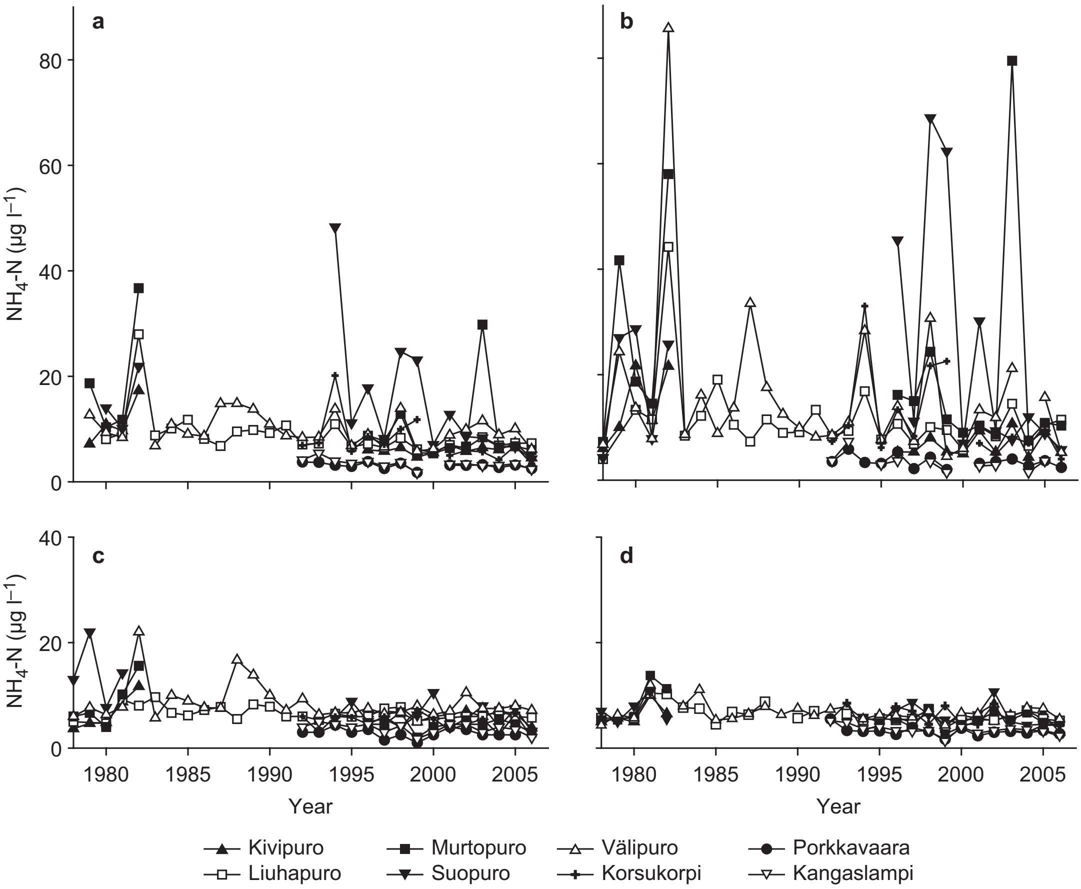

Fig. 3. Time-series for the (**a**) annual, (**b**) spring, (**c**) summer, and (**d**) autumn concentrations of stream water ammonium nitrogen (NH_4-N) in the study catchments in years 1979–2006. For further explanations, *see* Fig. 2.

as compared with the ones with low peatland percentage (r_S = –0.647, p = 0.04), and the increasing trend in the C/N ratio was steeper in catchments with high tree stand biomass as compared with those with low tree stand biomass (r_S = 0.667, p = 0.03).

According to the mixed model regression analysis the peatland proportion explained most of the variation in annual and seasonal TON, NH_4-N and NO_3-N concentrations and exports (Table 5). The stream water TON concentrations were also positively related to the water temperature in autumn, and the C/N ratio to the temperature sum and the N stored in soil and vegetation in the catchments. The variation in the annual stream water NO_3-N concentration was positively related to the total N deposition, and the variation in the annual and seasonal stream water NH_4-N concentrations and exports to the NH_4-N deposition. The exports of TON, NO_3-N and NH_4-N were also positively related to the amount of precipitation. Other studied variables (snow depth, snow fall, the area of the catchment, the fertility of site) were only occasionally significant in explaining the annual and seasonal N concentration and export.

The constructed concentration models explained 37%–72% of the total variance of the TON, 30%–62% of the NH_4-N, and 11%–50% of the NO_3-N (Table 5).The corresponding values for the export models were 63%–71% for TON, 33%–42% for NO_3-N, and 52%–60% for NH_4-N. These proportion estimates indicate that a large part of the total variation in the modeled variables remained unexplained, especially in N concentrations. The examination of the residuals did not

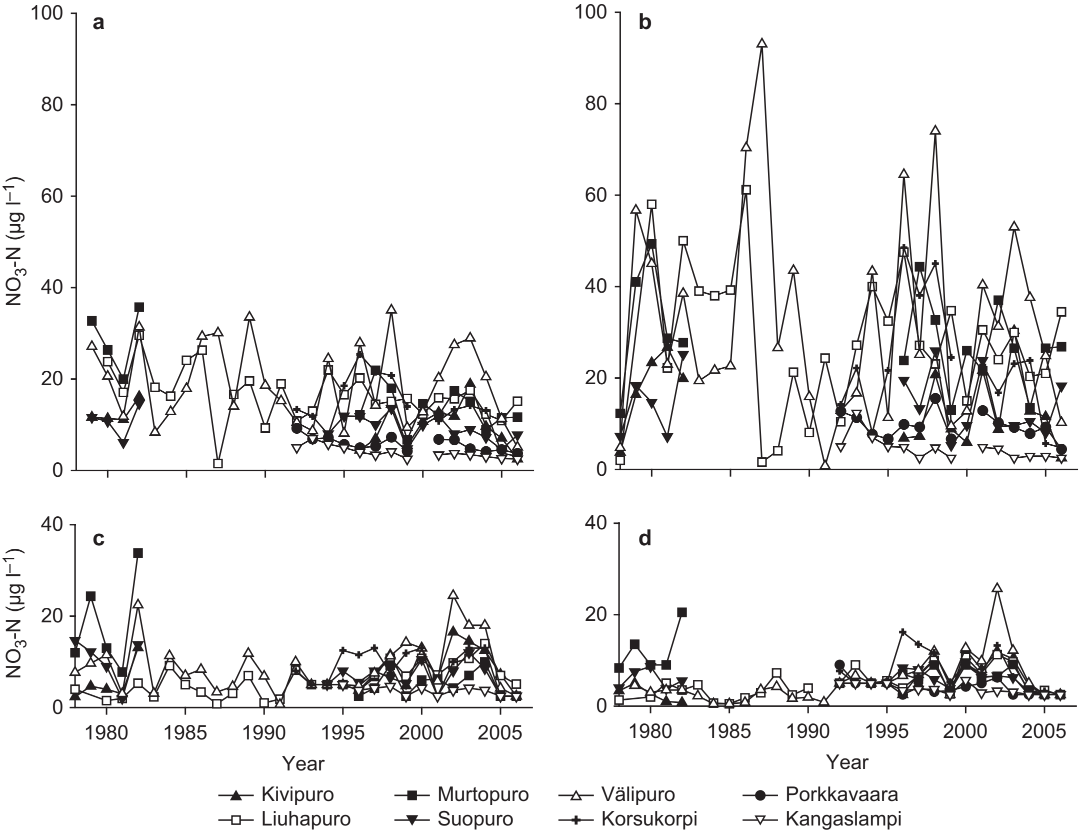

Fig. 4. Time-series of the (**a**) annual, (**b**) spring, (**c**) summer, and (**d**) autumn concentrations of stream water nitrate nitrogen (NO_3-N) in the study catchments in years 1979–2006. For further explanation, *see* Fig. 1.

reveal any systematic errors in the model predictions. However, the bias of the models varied much and was in some cases considerable.

Discussion

We analyzed the long-term trends in the stream water concentrations and export of TON and inorganic N in eight boreal forested catchments. The results did not indicate any change, or only a slight decrease in the concentrations of NO_3-N and NH_4-N. The TON concentrations (ca. 97% of the TN) did not show any change in six of the catchments and an increase in two of the catchments during the monitored period of the last 15–28 years. Even though, the lengths of the time series were not equal, it did not significantly affect the final results, because most of the temporal changes had occurred after the mid-1990s. The increase in the TON concentration during the last 10–15 years was reported earlier (e.g. Lepistö *et al*. 2008), whereas a large number of studies reported increased concentrations of organic carbon (Worrall *et al*. 2004, Eikebrokk *et al*. 2004, Vuorenmaa *et al*. 2006, Monteith *et al*. 2007). The increase in the mean annual TON concentration in the present study was less evident than the simultaneous increase in the TOC in the same catchments (Sarkkola *et al*. 2009: fig. 5). Earlier studies from unmanaged boreal forested catchments show a close correlation between stream water TOC and TON (e.g. Mattsson *et al*. 2003, Kortelainen *et al*. 2006). The weak correlation between TOC and TON in our study catchments indicates that the increasing trend in TOC observed in the earlier study (Sarkkola *et al*. 2009) was caused by the increased leaching of relatively undecomposed organic matter rather than highly decom-

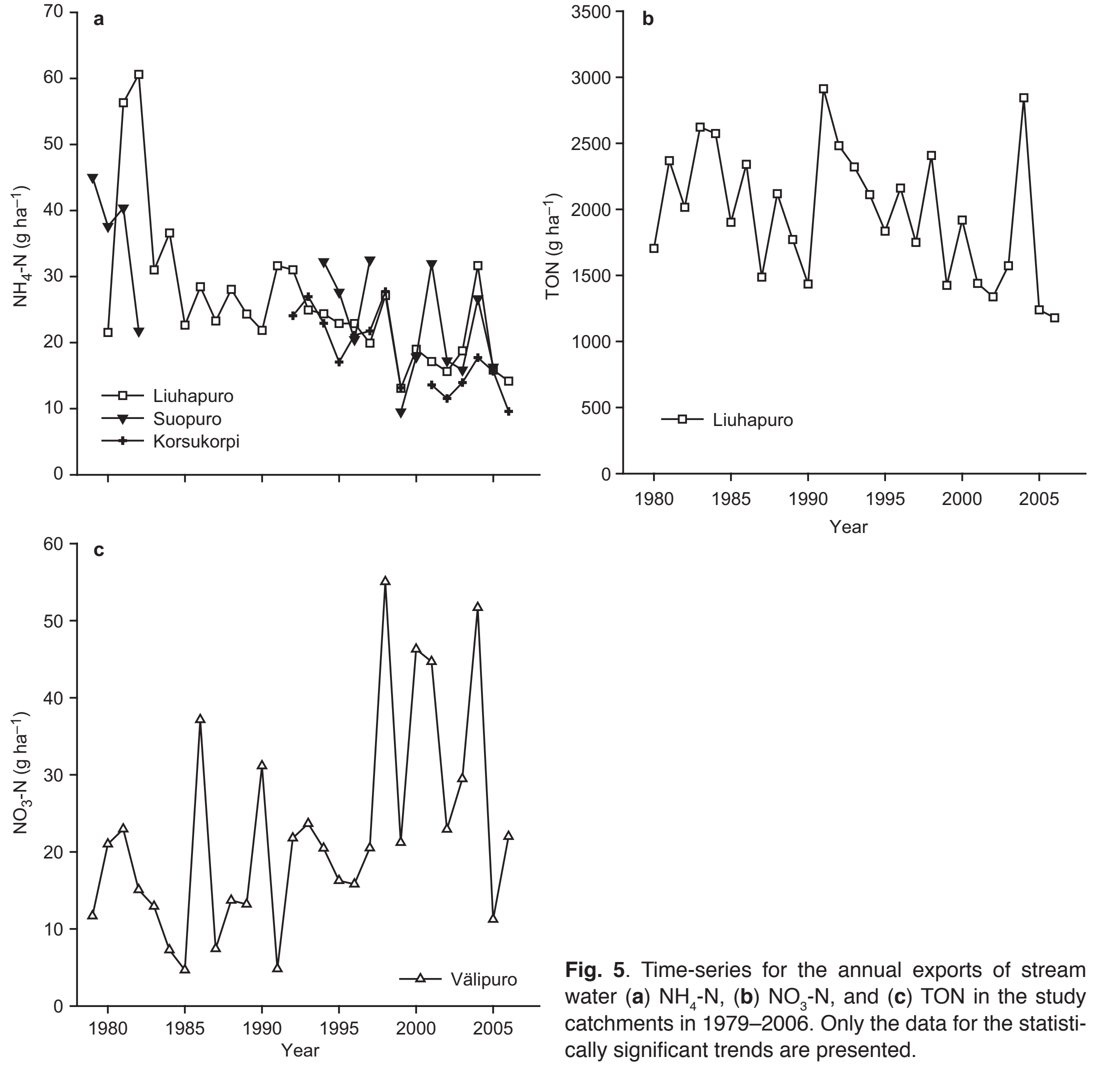

Fig. 5. Time-series for the annual exports of stream water (**a**) NH_4-N, (**b**) NO_3-N, and (**c**) TON in the study catchments in 1979–2006. Only the data for the statistically significant trends are presented.

posed material. A large number of studies have shown that the N concentration in organic matter increases along with an increased decomposition (e.g. Laiho and Laine 1994).

The mean annual air temperature increased by about 1.2 °C during the monitoring period (1979–2006), and significant seasonal trends were found in summer and autumn temperatures (Sarkkola *et al*. 2009: fig. 2). The respective temporal increases also occurred in the water temperatures (Fig. 1). The increased temperature may have increased the N mineralization from the soil and the export of inorganic N into the stream water. Since the N leaching did not increase, this may indicate a greater uptake and

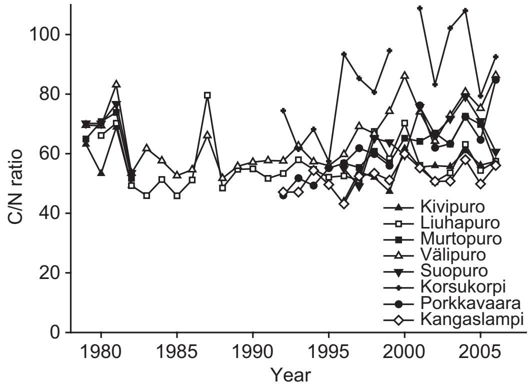

Fig. 6. Time series for the annual stream water C/N ratio in the catchments in 1979–2006. Statistically significant trends are listed in Table 2.

Table 4. Directions of statistically significant ($p < 0.05$) trends in the seasonal (spring, summer and autumn) concentrations and export of NH_4-N, NO_3-N, TON and the C/N ratio in the stream water in the studied catchments.

Catchment	NH_4-N						NO_3-N						TON						C/N ratio		
	Concentration			Export			Concentration			Export			Concentration			Export					
	Spr	Sum	Aut	Spr	Sum	Aut	Spr	Sum	Aut	Spr	Sum	Aut	Spr	Sum	Aut	Spr	Sum	Aut	Spr	Sum	Aut
Kivipuro													+						+		
Liuhapuro	–			–		–		+						+	+	–		–			
Murtopuro	–					–	–	–	–		–		+							–	
Suopuro		–	–																		
Välipuro									+			+							+		
Korsukorpi																				+	
Porkkavaara																			+		+
Kangaslampi	–						–	–	–												+

accumulation of N by the vegetation under the warmer summer conditions and the extended growth periods (Bauer *et al*. 2004). Melillo *et al*. (2002) studied the effect of increased air temperature on N mineralization in N-limited forest soils under laboratory conditions and found that soil warming increased the vegetative N uptake more than net N mineralization from the soil. However, the poor correlation between increased air and stream water temperatures and inorganic N export may have mostly resulted from the fact that the effects of temperature have been concealed behind the concurrent decrease in the N deposition. According to the mixed-model analysis, the variation in the annual and seasonal stream water NH_4-N concentrations and exports was significantly related to the level of NH_4-N deposition, and the variation in the annual stream water NO_3-N concentration, to the total N deposition. Our findings are in accordance with those of Rekolainen *et al*. (2005) who reported a decrease in the inorganic N concentrations in Finnish oligotrophic lakes during the period 1980–2003 concurrently with the decrease in N deposition. Vuorenmaa *et al*. (2002) analyzed the TN export from boreal headwater catchments during the 1980s and 1990s and explained the observed decreasing trends by the decrease in TN deposition. Aber *et al*. (2003) studied the soil N chemistry in forested areas in northeastern USA and found a positive relationship between NO_3-N export and N deposition. In our study areas, the TN deposition had decreased from the top values of about 8 kg ha^{-1} a^{-1} to 3 kg ha^{-1} a^{-1} since the end of the 1980s.

The relatively low concentrations of NH_4-N and NO_3-N and the decreasing trends of them in our study catchments indicate a low risk for N saturation. This is also supported by the fact that the N deposition in the catchments was higher than the export into the stream waters. The soil C/N ratio for the study catchments varied between 30 and 79. This was higher than the < 25–30 ratio level, regarded as the critical limit for the occurrence of net nitrification (Willard *et al*. 1997, Gundersen *et al*. 1998, Kjonaas and Wright 1998, Dise *et al*. 2009) and an increasing risk of NO_3-N leaching (Lowett *et al*. 2002). The stream water C/N ratios were also high (52–85) and increased during the monitoring

Table 5. Parameters and performance values of the mixed models constructed for the mean annual and seasonal concentrations and exports of TON, NO_3-N, NH_4-N, and the CN ratio in stream water in the studied catchments. Depending on the model, the hydrometeorological variables are either the annual or seasonal averages. The parameter values of the fixed parts of the models (α, β, in Eq. 1) are significant at $p < 0.05$. For random part, the non-significant parameters (u, e) are indicated with ^{ns}.

	α	ln(peatland) (%)	Precip. (mm)	Snow depth (cm)	Snow fall (mm)	ln(stream water temp.) (°C)	Mean stand vol. (m^3 ha^{-1})	TN depos. kg ha^{-1}	NH_4 depos. kg ha^{-1}	ln(area) (ha)	Temp. sum (dd)	Fertile sites (dummy)	ln(N-stores) (kg ha^{-1})	u	e	Bias	Bias%	Percentage of the tot. variance eplained
ln(TON) (μg l^{-1})																		
Annual	4.7933	0.3275			0.0009									0.1031	0.0158	3.77	–11.05	72
Spring	3.9022	0.4953	0.0011											0.0545	0.0197	7.30	–2.86	68
Summer	4.6015	0.4040												0.0400	0.0316	8.22	–4.54	58
Autumn	4.4651	0.3614				0.1609								0.1049	0.0242	15.95	–5.85	37
ln(TON) (kg ha^{-1})																		
Annual	4.2724	0.5301	0.0022	0.0083							–0.0005			0.0721	0.0755	–22.77	–18.45	71
Spring	4.5471	0.4989	0.0034				–0.0027							0.0880	0.0423	24.68	–6.52	63
Summer	1.6953	0.4650	0.0099									–0.2399		0.0098	0.2198	25.80	–13.50	64
Autumn	1.6423	0.5737	0.0096					0.0003						0.0578	0.2020	21.58	–13.70	63
ln(NO_3) (μg l^{-1})																		
Annual	–0.2527	0.6493						0.00006						0.0495^{ns}	0.1821	0.21	–23.00	50
Spring	0.1702	0.6227	0.0027											0.0967^{ns}	0.5211	1.59	–82.15	30
Summer	0.6387	0.3004												0.0156^{ns}	0.4378	1.39	–26.82	9
Autumn	2.3315		–0.0034								–0.0027	0.2749		0.0156^{ns}	0.3700	0.23	–55.10	11
ln(NO_3) (kg ha^{-1})																		
Annual	–1.6575	0.3594	0.0025							0.3934				0.0204^{ns}	0.3645	0.35	–39.07	42
Spring	–1.600	0.2474	0.0048	0.0196						0.445				0.0084^{ns}	0.5672	–0.07	–89.54	33
Summer	–1.3769		0.0100											0.0951^{ns}	0.5432	0.66	–39.26	33
Autumn	0.9114		0.0062				–0.0054				–0.0030			0.2091^{ns}	0.6372	0.53	–59.14	12
ln(NH_4) (μg l^{-1})																		
Annual	–0.6526	0.6233							0.0002					0.0239^{ns}	0.1521	0.49	–19.72	63
Spring	–0.9677	0.8008							0.0003^{a}					0.0145^{ns}	0.3105	–1.07	–33.30	61
Summer	0.4178	0.3313							0.0003					0.0422^{ns}	0.1260	0.74	–7.45	34
Autumn	0.6361	0.3435	–0.0029						0.0006					0.0025^{ns}	0.1886	1.92	–6.14	31
ln(NH_4) (kg ha^{-1})																		
Annual	–0.8517	0.6466	0.0016						0.0002					0.0310^{ns}	0.1844	1.00	–15.18	54
Spring	–0.5663	0.4929							0.0003^{a}	0.1964				0.0082^{ns}	0.2031	–5.91	–76.78	52
Summer	–2.2307	0.3557	0.0084						0.0005					0.0091^{ns}	0.2587	0.55	–11.22	60
Autumn	–2.1539	0.5012	0.0074						0.0009					0.0000^{ns}	0.3045	2.65	30.46	59
C/N ratio																		
Annual	–53.1550										0.0210		10.1342	60.0279^{ns}	83.0507	–0.81	–3.79	21

[a] total winter and spring deposition.

period, which could also be used as an indicator of a poor nitrification rate. These high C/N ratios may also explain the relatively low NO_3-N concentrations in the catchments. The negative relationships between precipitation and the autumn concentrations of NO_3-N (Table 5) may have been explained by the fact that nitrification is low under water saturated conditions.

The peatland proportion was the most important factor for the variation in the TON, NO_3-N and NH_4-N concentrations and exports into the stream water (Table 5). These results are in accordance with those of Kortelainen *et al.* (1997, 2006), who found a positive relationship between peatland coverage and the concentrations and exports of TON and NH_4-N. On the other hand, Kortelainen *et al.* (2006) found no significant relationship between the peatland proportion and the exports of NO_3-N from boreal headwater catchments with undisturbed, pristine peatlands. Pristine peatlands tend to increase the water retention time, which may favor gaseous N emissions through denitrification and retard NO_3 production through nitrification (Saunders and Kalff 2005 and the references therein).

In this study, there was a negative correlation between the mean tree stand volume in the catchments and the spring TON and autumn NO_3-N exports in the stream water. The tree stand may decrease the N export indirectly through a decreased runoff caused by tree stand evapotranspiration, and directly by an increased N uptake through the N demand by the trees. Kortelainen *et al.* (2006) found a weak relationship between tree stand volume and stream water N concentrations, while Mattsson *et al.* (2003) showed a strong positive correlation between stand volume, the proportion of Norway spruce and stream water N concentrations and exports. The relatively poor correlation between tree stand volume and N export in our study catchments may be a consequence of slow stand growth and production compared with more favorable climatic conditions and more fertile sites, where the role of the tree stand in N cycling may be more important (*see* Reynolds *et al.* 1994).

The annual and seasonal runoffs as well as the precipitation in the study catchments showed a slight decreasing trend during the monitoring period, but because of high temporal variability, these trends were not statistically significant (Sarkkola *et al.* 2009). Since the N export is derived as the product of the N concentration in the stream water and the runoff, precipitation as the trigger of runoff may become the main hydrometeorological driver in controlling the variation in the N exports (Table 5, *see* also Clair *et al.* 1994). Precipitation was found to be the dominant factor for the N exports during summer and autumn, whereas in spring the peatland proportion explained most of the variation in N exports in boreal headwater catchments (Kortelainen *et al.* 2006).

Conclusions

The results from the long-term monitoring of eight boreal forested catchments in an area charcterized by an increasing trend in air temperature and a decreasing trend in N deposition indicate that there are no general trends in the concentrations and exports of TON, NH_4-N and NO_3-N in stream water. Only in a few catchments the TON concentrations followed the observed increasing trends in stream water TOC concentrations, suggesting an effect of increasing temperature. In some of the catchments the NH_4-N and NO_3-N concentrations had a decreasing trend, suggesting an effect of the decrease in N deposition. However, the temporal changes in N concentrations and exports were small, most probably because the effects of the increased air and stream water temperatures have been largely concealed behind the concurrent decrease in the N deposition. The proportion of peatland within the catchment was the main explaining factor for the variation in the concentrations and export of inorganic and organic N-fractions between the catchments. The results indicate that deposition and hydrology rather than global warming largely determine stream water N concentrations and exports. Since the temperatures continue to increase, it is important to continue the monitoring in order to detect changes in N leaching related to climate change.

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