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Land-use dominates climate controls on nitrogen and phosphorus export from managed and natural Nordic headwater catchments

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Abstract

Agricultural, forestry-impacted and natural catchments are all vectors of nutrient loading in the Nordic countries. Here, we present concentrations and fluxes of total nitrogen (totN) and phosphorus (totP) from 69 Nordic headwater catchments (Denmark: 12, Finland:18, Norway:17, Sweden:22) between 2000 and 2018. Catchments span the range of Nordic climatic and environmental conditions and include natural sites and sites impacted by agricultural and forest management. Concentrations and fluxes of totN and totP were highest in agricultural catchments, intermediate in forestry-impacted and lowest in natural catchments, and were positively related %agricultural land cover and summer temperature. Summer temperature may be a proxy for terrestrial productivity, while %agricultural land cover might be a proxy for catchment nutrient inputs. A regional trend analysis showed significant declines in N concentrations and export across agricultural (–15 μg totN L^{-1} $year^{-1}$) and natural (–0.4 μg NO_3-N L^{-1} $year^{-1}$) catchments, but individual sites displayed few long-term trends in concentrations (totN: 22%, totP: 25%) or export (totN: 6%, totP: 9%). Forestry-impacted sites had a significant decline in totP (–0.1 μg P L^{-1} $year^{-1}$). A small but significant increase in totP fluxes (+0.4 kg P

km^{-2} $year^{-1}$) from agricultural catchments was found, and countries showed contrasting patterns. Trends in annual concentrations and fluxes of totP and totN could not be explained in a straightforward way by changes in runoff or climate. Explanations for the totN decline include national mitigation measures in agriculture international policy to reduced air pollution and, possibly, large-scale increases in forest growth. Mitigation to reduce phosphorus appears to be more challenging than for nitrogen. If the green shift entails intensification of agricultural and forest production, new challenges for protection of water quality will emerge possible exacerbated by climate change. Further analysis of headwater totN and totP export should include seasonal trends, aquatic nutrient species and a focus on catchment nutrient inputs.

KEYWORDS

agriculture, bioeconomy, forest, forestry, long-term trend, mitigation, monitoring, stream

1 | INTRODUCTION

Reconciliation of increasing reliance on agricultural and forestry products with water quality protection is becoming more urgent under the green growth policies that are being developed in Europe. The so-called "green shift" towards a low-carbon and resource-efficient society to mitigate climate change is expected to require wood and crop-based biomass for replacement of fossil resources by renewable energy-sources (Scarlat, Dallemand, Monforti-Ferrario, & Nita, 2015).

During the green shift, production of meat and cereals in Europe are expected to intensify—partly as consequence of income growth—with adverse effects on water quality (Rosegrant, Ringler, Zhu, Tokgoz, & Bhandary, 2013). Agricultural production in Europe is one of the main pressures identified under the Water Framework Directive (WFD) that reduces ecological status of surface and coastal waters, by increasing runoff of nitrogen and phosphorus (EEA, 2019). The WFD is implemented nationally with approaches targeted at climate, soil, hydrological conditions and agricultural production (Ulen, Bechmann, Folster, Jarvie, & Tunney, 2007). The WFD aims for good ecological, chemical and hydromorphological status by the end of 2027. However, river basin management and restoration actions could be insufficient to reach the WFD aims within 2050. In particular, diffuse nutrient loadings should be reduced significantly (Hering et al., 2010; Räike, Taskinen, & Knuuttila, 2020). National agro-environmental legislation in the Nordic countries since 1990 has led to country-specific reductions of the N and P field balances (inputs from fertilizer substracted with removal by harvest) on agricultural land. As an example, Danish regulations decreased the N and P field balance with 45% and 76% during the period 1990–2015 (Blicher-Mathiesen et al., 2020) and in Finland with 35% and 60%, respectively, during the period 1995–2013 (Aakkula & Leppänen, 2014). These country-specific reductions in field nutrient balances are expected to affect nutrient runoff from agricultural catchments although the presence of legacy nutrient stores will confound such relationships (Tattari et al., 2017).

Forest management to increase biomass for bioenergy is another pressure with potential effects on nutrient loadings to surface waters (Kreutzweiser, Hazlett, & Gunn, 2008). Increased extraction of forest biomass and intensified forestry have trade-offs with ecosystem services like biodiversity (Eyvindson, Repo, & Monkkonen, 2018) and are likely to also impact water quality (Laudon et al., 2011). Local increases of nitrogen and phosphorus concentrations in forestry-impacted catchments is well-documented (De Wit et al., 2014; Lofgren, Ring, von Bromssen, Sorensen, & Hogbom, 2009) but its impact on a wider temporal and spatial scale is less clear (Sponseller et al., 2016). In areas with intensive peatland forestry like Finland, harvesting operations in forest on organic soils are of specific concern (Marttila et al., 2018; Nieminen et al., 2018). Furthermore, intensified forestry by conducting whole tree harvesting may require application of a fertilizer to avoid reduced forest growth (Akselsson, Westling, Sverdrup, & Gundersen, 2007; Merila et al., 2014). In freshwaters, phosphorus is usually considered to be the limiting nutrient (Schindler, 1977) although more recent studies suggest a role for nitrogen as well (Bergstrom & Jansson, 2006). In marine waters, production typically is limited by nitrogen (Howarth & Marino, 2006).

Agriculture and forestry are important pillars of Nordic societies. Forestry is of particular interest to Sweden and Finland, both of which have a high percentage of productive forest and a large forestry sector (Verkerk et al., 2019). Agriculture is a dominant land use in Denmark and across the Nordic region, nutrient loadings from agriculture is a large concern for eutrophication of freshwater and marine ecosystems (Frigstad et al., 2020; Karlson, Rosenberg, & Bonsdorff, 2002). These waters are valuable natural resources and essential for Nordic societies, economies and human wellbeing as they provide multiple ecosystem services (Kronvang et al., 2008; Marttila et al., 2020; Ulen et al., 2007).

A solid understanding of processes driving catchment export of nitrogen and phosphorus is thus at the basis of predictions of effects of the green shift on water quality. In addition to agricultural and forest management, catchment export of nitrogen and phosphorus can be accelerated by extreme hydrological events (Borgesen & Olesen, 2011; Mellander et al., 2018) despite the measures and efforts made particularly in agriculture to reduce loading. Hydrological conditions are changing in the Nordic region (Oygarden et al., 2014) and are expected to continue to do so in the future (Arheimer & Lindstrom, 2015; Huttunen et al., 2015). Nutrient-runoff mitigation measures at the catchment scale (e.g., buffer zones, artificial wetlands) in a changed climate (Carstensen et al., 2020; Haygarth et al., 2012; Laudon et al., 2016) may not be sufficient for adequate protection of water quality.

Small headwater catchments without point sources of nutrients provide an ideal framework to assess land use and climate impacts on nutrient export to surface waters, because nutrient retention in small streams is of lesser importance compared with larger river basins (Weigelhofer, Ramiao, Pitzl, Bondar-Kunze, & O'Keeffe, 2018). Nitrogen deposition from air pollution is a driver of changes in nitrogen runoff in such small catchments (Vuorenmaa et al., 2018), mitigated by international policy to reduce emissions of nitrogen to the atmosphere. Long-term changes in diffuse nutrient fluxes from managed catchments are strongly influenced by a complex combination of temporal and spatial factors, such as fluctuating climatic and hydrological conditions, land cover, soil characteristics, crop cycles and land-use practices in forestry and agriculture (Bechmann et al., 2014; Kyllmar, Carlsson, Gustafson, Ulen, & Johnsson, 2006; Tattari et al., 2017; Vuorenmaa, Rekolainen, Lepisto, Kenttamies, & Kauppila, 2002). Similarly, natural catchments are governed by climatic and hydrological conditions (Vuorenmaa et al., 2018) but lack the confounding influence of management. Such catchments provide a reference that enables distinguishing between interacting pressures, that is, intensified agricultural and forestry-related land use, and climate change (Skarbovik et al., 2020). Combined records from natural and managed catchments, from a wide range of environmental gradients and under common management regimes, are therefore potentially useful for assessing water quality responses to environmental change and evaluation of mitigation measures to protect water quality.

Here, we present concentrations and export of total nitrogen (totN) and total phosphorus (totP) and other species of nitrogen and phosphorus, in 69 Nordic headwater catchments for the period 2000–2018. The catchments are representative of Nordic natural (unmanaged) and agricultural landscapes and include also forestry-impacted sites. They cover a wide range of climate, soil type, runoff and management patterns. The primary focus is on totN and totP because all study sites have full records suitable for trend analysis (10 years or more) for these parameters. We test effects of land use, climate, runoff and land cover on spatial variation and temporal trends on concentrations and fluxes of totN and totP. Because nutrient runoff is potentially sensitive to differences in national mitigation measures (Hellsten et al., 2019; Kronvang et al., 2008; Ulen et al., 2007), we analyse for patterns by country in addition to examining patterns within land-use categories across the Nordic region.

2 | MATERIALS AND METHODS

Study sites and monitoring programs Data records on water chemistry, discharge and climate were compiled for 69 small catchments in Denmark (n = 12), Finland (n = 18), Norway (n = 17) and Sweden (n = 22) for the period 2000–2018 (Figure 1, Table 1). Detailed catchment-specific information is available (Table SI-1). All catchments are included in national monitoring programs, designed to assess long-term effects of air quality, agriculture and forestry on water quality. The monitoring programs follow standardized national or international procedures for sampling and chemical analysis, including QA-QC procedures. In all countries the analytical programs have changed over time, depending on funding and monitoring priorities. All sites have records of totN and totP for at least 10 full calendar years. Shorter time series were available for some variables (see Table SI-2). All catchments were attributed to a land-use category, that is, agriculture, forestry-impacted and natural.

Discharge was measured daily, using an open channel stage-discharge relationship in Danish streams. V-notch profiles or crump weir with a stage-height relationship were used elsewhere. Water was predominantly sampled by grab sampling, except for agricultural catchments in Sweden and Norway where flow-proportional composite sampling was used (Deelstra et al., 2014; Kyllmar, Forsberg, Andersson, & Martensson, 2014b). Sampling frequency varied from weekly to every second week to monthly and was stable throughout the entire monitoring period, with some exceptions as described below. In agricultural catchments, field management is monitored on an annual basis, except in Finland where annual management data are not available from all sites (Tattari et al., 2017). In all catchments, point sources are of minimal importance for annual loading of nitrogen and phosphorus.

From Denmark, the five predominantly agricultural and seven catchments with natural vegetation belong to the National Monitoring and Assessment Program for the Aquatic and Terrestrial Environments (NOVANA) (Svendsen, Bijl, Boutrup, & Norup, 2005) and monitoring started in 1990. Water chemistry in catchments with natural vegetation was sampled every 2 weeks or every month until 2009, when the sampling programs were repeated every third year. Catchments with natural vegetation are mostly forested with extensive forest management in some while two contain <10% of extensively managed grassland areas (Supplementary Information) and are surrounded by agricultural areas of differing intensity. Farming practices in the five agricultural catchments include cereal production for intensive pig farming, intensive dairy farming areas with mixed fodder crops and cereal and sugar beet production on clay soil (Blicher-Mathiesen et al., 2020). All catchments are situated at low altitudes (8–110 m.a.s.l.). Catchment soils are mainly sand and sandy loams but one agricultural catchment is on clay soil (Svendsen et al., 2005). Subsurface drainage depends on soil texture (rare in sandy soils, common in fine-textured, especially clay soils).

For Finland, six agricultural catchments, eight forestry-impacted and four natural catchments are included (Seuna, 1983). Water discharge monitoring was initiated in 1957 and monitoring of water quality in 1962 (Vuorenmaa et al., 2002). Catchment boundaries are well-defined, and typically groundwater and surface water boundaries coincide. Most arable land in the agricultural catchments is located on graded soils, with high proportions of silt and clay except for one site (Haapajyrä) where acid sulphate soils dominate, related to post-ice age

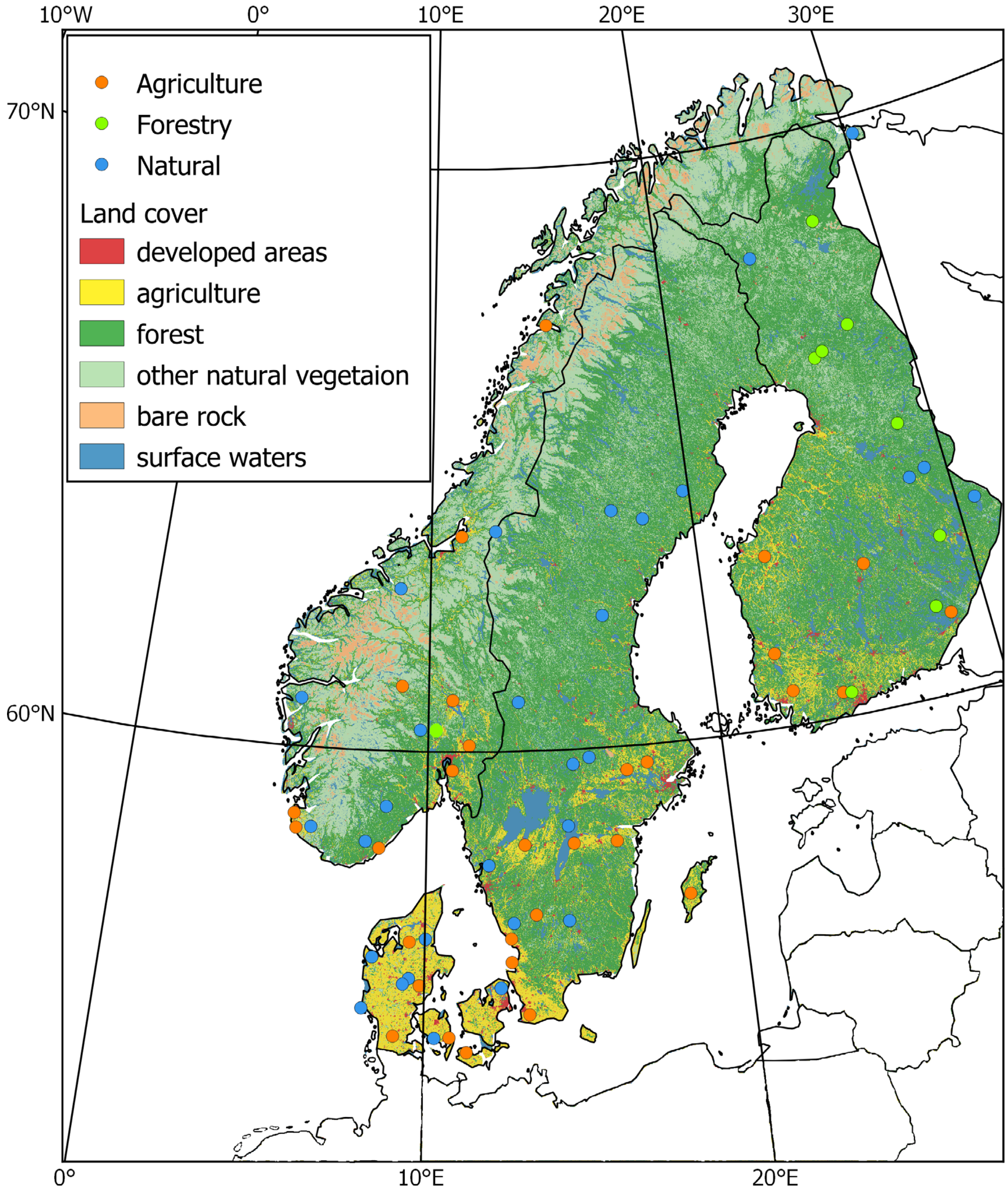

FIGURE 1 Map of study catchments in the Nordic countries. The land cover information bases on CORINE land cover information (https://land.copernicus.eu/pan-european/corine-land-cover). Colour coding refers to land use category

land uplift of marine sediments, which leach considerable amounts of sulphuric acids. Crop cultivation includes mostly cereals and root crops.

Eight small forestry-impacted catchments represent extensively managed forest land (Tattari et al., 2017; Vuorenmaa et al., 2002). The most important forestry practices (drainage, clear-cutting, soil scarification, fertilization) were undertaken in 1960–1990 (Kortelainen & Saukkonen, 1998). Soil types vary from mineral to peatland-dominated. Four Finnish natural catchments were included. These are dominated by coniferous forests and peatlands on moraine or organic soils (Ahtiainen & Huttunen, 1999; Lohila et al., 2015).

The nine Norwegian agricultural catchments are monitored under the Norwegian Agricultural Environmental Monitoring program (JOVA) (Bechmann et al., 2008). Monitoring at all but one catchment started between 1990 and 1995. The catchments represent the main farming systems in Norway; cereal production in the eastern and middle parts of the country, vegetable production in the south, intensive dairy farming in the west, and more extensive grass production in the southern mountains and in northern Norway (Wenng, Bechmann, Krogstad, & Skarbøvik, 2020). Soil texture varies from clay loam (dominated by surface runoff), loam and sand

TABLE 1 Key site characteristics, grouped by country and land use category, presented as median (minimum–maximum) values

		Denmark	Finland	Norway	Sweden	All
Agricultural	n	5	6	9	10	30
	Area (km^2)	7.5 (4.4–11)	5.7 (0.1–15)	3.1 (0.9–29)	7.8 (1.8–33)	
	Elevation (m.a.s.l)	30 (8–110)	48 (25–95)	119 (23–652)	42 (8–155)	
	Agriculture (% of area)	80 (64–87)	48 (17–100)	62 (35–88)	87 (58–93)	
	Forest (% of area)	7 (2–25)	44 (0–69)	29 (3–55)	4 (0–32)	
	Peatland (% of area)	2 (0–5)	9 (0–26)	1 (0–24)	0 (0–0)	
	MAT (°C)	10.2 (9.7–10.7)	6.2 (5.2–6.8)	6.9 (4.2–9.5)	8.5 (7.9–10.0)	
	MAP (mm)	773 (718–862)	626 (568–683)	948 (629–1,520)	619 (546–929)	
	MAQ (mm)	246 (146–550)	256 (239–343)	674 (287–1,067)	241 (150–467)	
	totN conc; flux	4,298; 1,151	2,694; 911	4,440; 3,000	4,419; 1,481	4,231; 1,450
	totP conc; flux	107; 28	90; 26	130; 151	172; 48	128; 44
Natural	n	7	4	7	12	30
	area (km^2)	4.6 (0.4–16)	1.2 (0.72–5)	2.6 (0.4–25)	1.2 (0.04–9)	
	Elevation (m.a.s.l)	53 (8–93)	183 (166–320)	521 (163–935)	322 (127–643)	
	Agriculture (% of area)	1 (0–10)	0 (0–0)	0 (0–0)	0 (0–0)	
	Forest (% of area)	90 (8–99)	72 (53–84)	20 (4–90)	81 (59–92)	
	Peatland (% of area)	0 (0–0)	28 (16–47)	6 (0–22)	14 (0–38)	
	MAT (°C)	10.0 (9.5–10.7)	3.7 (1.2–4.0)	5.1 (1.8–7.8)	5.2 (2.9–8.5)	
	MAP (mm)	824 (782–1,003)	691 (607–754)	1,203 (553–3,719)	760 (622–1,186)	
	MAQ (mm)	168 (98–218)	336 (289–376)	1,150 (436–2,299)	523 (315–946)	
	totN conc; flux	960; 117	240; 77	226; 240	318; 139	280; 139
	totP conc; flux	48; 10	7; 2	3; 4	7; 4	7; 4
Forestry	n		8	1		9
	area (km^2)		14.6 (0.7–56)	0.3		
	Elevation (m.a.s.l)		158 (41–270)	590		
	Agriculture (% of area)		0 (0–3)	0		
	Forest (% of area)		64 (38–98)	73		
	Peatland (% of area)		36 (2–59)	22		
	MAT (°C)		3.0 (0.9–6.7)	5.1		
	MAP (mm)		647 (588–754)	840		
	MAQ (mm)		389 (229–444)	877 (877–877)		
	totN conc; flux		584; 200	345; n.d.		555; 200
	totP conc; flux		22; 7	8; n.d.		20; 7

Note: Data for climate, discharge, concentrations and fluxes are given for the period 2000–2018. Concentrations and fluxes are medians of median annual values; units for totN and totP concentrations in $\mu g\ L^{-1}$; totN and totP fluxes in $kg\ km^{-2}\ year^{-1}$.

Abbreviations: MAP, mean annual precipitation; MAQ, mean annual discharge; MAT, mean annual temperature.

(subsurface drainage) to peat and sandy soils (subsurface drainage) (Bechmann, 2014).

The seven natural catchments in Norway are part of the national program for monitoring effects of long-range transported air pollution (Garmo & Skancke, 2018). Monitoring started during the early 1970s in four stations (de Wit, Hindar, & Hole, 2008), and around 1990 in three other stations. The catchments are all located on acid-sensitive bedrock and land cover varies from forest, forest interspersed with peatland, and predominantly tundra or bare rock. Catchments are hydrologically well-defined, with gneissic and granitic bedrock and soils developed on moraine/glacial till. The forestry site at Langtjern was established in 2008 (De Wit et al., 2014).

Eight of the 10 Swedish agricultural catchments are from the Swedish national agricultural monitoring program, and two are from a less intensively monitored regional program (Kyllmar et al., 2014a). The monitored catchments were established between 1988 and 1996. All catchments have a large proportion of agricultural land, most of which is tile drained. Catchments cover the main variations in climate and geological characteristics in Sweden and hence in agricultural production (Kyllmar et al., 2014b). In south-west Sweden, catchments have sandy loam soils and are characterized by intensive crop production including cereals, rape seed, potatoes and vegetables. In the southern inland highlands, where precipitation is higher and soils are coarse, grass and dairy production are typical. Swedish catchments with clay soils, mainly located in the central agricultural areas, are characterized by production of cereals and rape and a low number of animal units. In east Sweden, catchments with sandy loam soils have mixed crop production including irrigated areas with potatoes. In 2004, the sampling method changed from grab sampling to flow-proportional composite sampling and in this study only data from 2004 and onwards were used.

The 12 Swedish natural catchments were established to study long time trends in natural, undisturbed areas (Folster, Johnson, Futter, & Wilander, 2014) and consequently have not been impacted by forest management in the past four to five decades. Four catchments belong to the Integrated Monitoring of the Environmental Status in Swedish Forested Ecosystems (IM), established in the late 1980s and mid-1990s (Vuorenmaa et al., 2018). Six catchments are included the PMK5 long-term monitoring program (Folster & Wilander, 2002), one is part of the Krycklan Catchment Study (Laudon et al., 2013) and one is included in the Integrated Studies of the Effects of Liming Acidified Waters, ISELAW program (Appelberg, Lingdell, & Andren, 1995) where liming took place around 1990. Catchments are hydrologically well-defined and several of the catchments include a small lake or pond (0–13% water). All Swedish forested catchments are dominated by coniferous stands and the granitoid bedrock is covered with till-soils interspersed with mires and small lakes.

2.1 | Water chemical data and analytical methods

Common variables in all monitoring programs were totN and totP. In the natural catchments, monitoring programs also included nitrate (NO_3), ammonium (NH_4) (allowing computation of total organic N, TON) and total organic carbon (TOC). In Finnish catchments and some Danish natural catchments, dissolved reactive phosphorus (DRP) and suspended solids (SS) were included. In agricultural catchments, NO_3 was most often included in addition to DRP and SS. All monitoring programs used accredited laboratories and standardized analytical programs (Bechmann et al., 2008; Folster et al., 2014; Garmo & Skancke, 2018; Kortelainen et al., 2006; Kyllmar et al., 2014a; Pengerud et al., 2015).

2.2 | Calculation of fluxes

Element fluxes in catchments with grab sampling were calculated by linear interpolation of daily nutrient concentrations between sampling dates, multiplying daily concentrations with daily discharge to sum over a calendar year to give annual element export. In catchments with flow-proportional sampling, annual and monthly concentrations were calculated by summarizing daily fluxes over a month or a year and divided by total water discharge during the corresponding period. Daily nutrient fluxes were obtained by multiplying daily water discharge with concentrations in the corresponding weekly/ fortnightly water sample. Daily concentrations were set to be constant for the period between outtake of composite water samples, most often every second week (Wenng et al., 2020).

2.3 | Climate data

For each catchment, mean annual and summer (June, July, August) temperature and precipitation were derived from publicly available gridded datasets using area-weighted catchment averages. For Finland, Norway and Sweden, data were obtained from the Norwegian Meteorological Institute Nordic Gridded Climate Dataset, version 19.09, with a spatial resolution of 1×1 km (met.no). Data with a daily resolution, interpolated to a grid from observational data using Bayesian interpolation and scale-separation concepts, was used as input data. For Denmark, hourly timestep ERA5 data with a resolution of 0.25° (https://climate.copernicus.eu/climate-reanalysis) were used.

2.4 | Trend analysis

Annual median concentrations (totN, totP, NO_3, DRP and SS) were calculated using monthly median concentrations in all sites for the period 2000–2018 where at least 10 years of data were available. This guaranteed comparability as sampling method (grab or composite) and periods differed among countries and sites. Clear outlier data points outside normal variation (including extremes) were excluded from the median concentration calculations. Annual fluxes for totN and totP were calculated following standard national procedures. All statistical tests were run in R (version 3.5.0). For trend analyses, we used the Mann-Kendall trend test with R package

TTAinterfaceTrendAnalysis. We applied the Mann-Kendall trend test to yearly median concentrations and fluxes for the period 2000–2018 and used the Theil-Sen estimator to estimate temporal concentration trends. The Theil-Sen estimator can be applied when the data contains outliers or when data are missing (Bouza-Deaño, Ternero-Rodríguez, & Fernández-Espinosa, 2008). To transform absolute changes into relative changes, in percent per year, we multiplied Theil-Sen slopes by 100 and divided by the long-term median values. We used Regional Mann-Kendall analysis using R package rtk to study patterns in trends at country level (Norway, Finland, Sweden and Denmark) for concentrations (totN, totP, NO_3, DRP and SS) and fluxes (totP and totN).

2.5 | Statistical analysis

Partial least square regression (PLS) was used to evaluate how environmental variables explained the variance of median totN and totP concentrations as dependent variables. PLS is particularly useful, when a large set of explanatory variables is given and variables are collinear (Abdi, 2007; Wold, Sjostrom, & Eriksson, 2001). The explanatory variables used, in both the totN and totP model, were latitude, longitude, land cover categories (agriculture, forest, peatlands, shrublands and lakes), annual and summer mean temperature, annual runoff, annual precipitation and summer precipitation (Table SI-1). The strength of the PLS models was evaluated by the goodness of fit, r^2, and the goodness of prediction, Q^2, for each component. The importance of each explanatory variable was related to the variable influence on the projection (VIP). Variables with a VIP value above 1 are commonly identified as most influential when explain the variance of dependent variables. The PLS was run in R version 3.5.2 using the packages pls and plsdepot.

3 | RESULTS

3.1 | Catchment characteristics and mean water chemistry and nitrogen and phosphorus fluxes

The study sites included 30 agricultural catchments, 30 natural catchments and 9 forestry impacted catchments (8 in Finland, 1 in Norway), distributed throughout the Nordic countries (Figure 1, Table 1, Table SI-1). Catchment areas ranged between <0.5 km^2 to >25 km^2, with a tendency towards smaller-sized natural catchments. In Finland, peatlands covered on average 28% of the catchment area in natural and in forestry-impacted sites, considerably more than in Sweden (14%) and Norway (6%). Forest was the dominant land cover in natural catchments except in Norway, where mountains and shrublands dominated. The agricultural sites were generally located at lower elevations than the natural and forestry-impacted sites, except in Denmark. Agricultural sites had a warmer climate than natural and forestry-impacted sites, probably partly because of lower elevation and placement at lower latitudes with favourable climatic conditions for agriculture. East–west precipitation gradients are considerable in the Nordic countries, illustrated by the high precipitation received by the Norwegian catchments compared with Finland, while Denmark and Sweden were intermediate. Annual runoff was highest in Norway and lowest in Denmark. Low runoff in Denmark is notable because precipitation here was not markedly different from Finland and Sweden.

Median concentrations of totN and totP declined in the following order: agriculture (totN 4.2 mg L^{-1}; totP 0.14 mg L^{-1}) > forestry (totN 0.6 mg L^{-1}; totP 0.02 mg L^{-1}) > natural (totN 0.28 mg L^{-1}; totP 0.007 mg L^{-1}) (Table 1). Ranges in concentrations of totN and totP overlapped between countries, with a tendency towards highest totP in Sweden and Norway in the agricultural catchments and the lowest totP in natural Norwegian catchments (Figure 2). For totN, countries were more similar than for totP. In Denmark, ranges in totP and totN of agricultural and natural catchments were more similar than in other Nordic countries.

In all land-use categories, NH_4 made up a very small part of totN concentrations (<5%) (Table 2). By contrast, NO_3 was the dominant fraction of totN in agricultural catchments (55–85%) while in natural and forestry-impacted catchments, concentrations of NO_3 were between 1% and 17% of totN, with Denmark as a notable exception (59%). In natural and forestry-impacted catchments, the organic fraction dominated totN (77–95%). In agricultural catchments, DRP was between 24 and 38% of totP and was significantly correlated with totP (data not shown, r^2 = 0.54, p < .0001). In natural catchments, totP was significantly correlated with TOC (data not shown, r^2 = 0.76, p < .0001) suggesting that most totP was organic P. In Danish natural sites, totP concentrations were positively correlated with SS (p < .05, data not shown) and negatively with TOC (p = .053), suggesting that totP here was particle-bound rather than of organic origin. Where DRP was measured in natural catchments (14 sites), it made up 18–42% of totP, which indicates that a considerable part of totP was bio-available. The DRP fraction is traditionally thought to identify inorganic reactive fractions of P but may also include labile organic fractions (e.g., Haygarth & Sharpley, 2000). SS were correlated with totP in agricultural catchments (data not shown, r^2 = 0.38, p < .001), indicating that a considerable part of totP was particle-bound.

Median annual fluxes of totN and totP (kg km^{-2} $year^{-1}$) declined in the order agriculture (totN 1,450; totP 44) > forestry (totN 200; totP 7) > natural (totN 139; totP 4) (Table 1). Norway had the highest totN fluxes in agricultural and natural catchments, probably because of higher runoff (Figure 3). The same pattern emerged for totP fluxes in agricultural catchments, but the Danish natural catchments exported more totP than the other natural catchments despite lower runoff.

3.2 | Spatial patterns in concentrations and fluxes

We explored spatial patterns in totN and totP concentrations and fluxes in relation to land use climate and runoff. Flow-weighted and arithmetically averaged concentrations revealed similar relationships

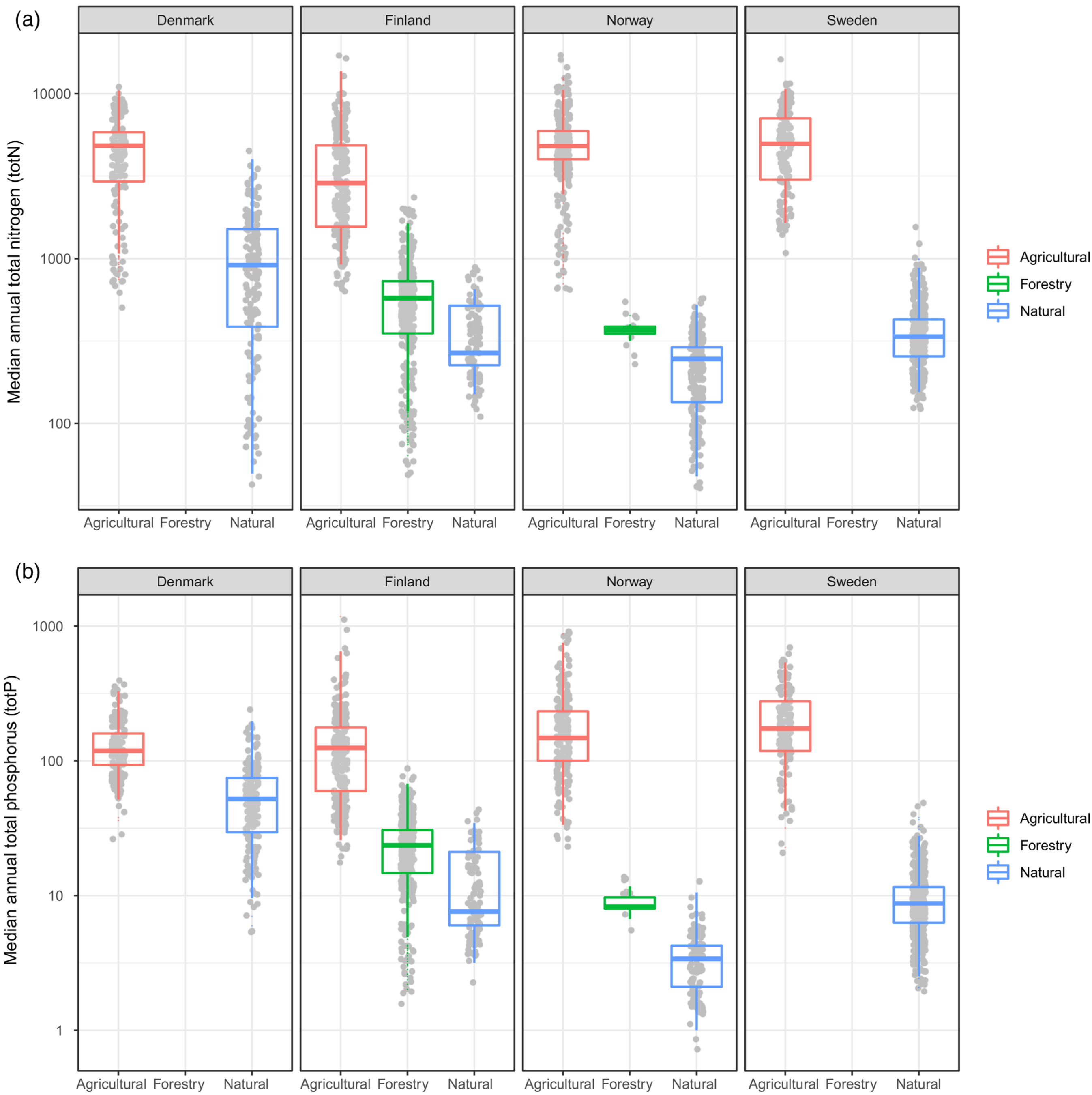

FIGURE 2 Concentration ranges for the grouped sites, for median totN and totP (μg^{-1}) calculated for annual averages for agricultural (n = 30), forestry (n = 9) and natural (n = 30) sites for the period 2000–2018

(data not shown) and we focus here on results for arithmetically averaged concentrations and fluxes.

The PLS models with annual median totN and totP concentrations as dependent variables revealed similar relationships. Only the results for totN are given here (Figure 4) while totP results are given in SI (Figure SI-1). The first two components explained 71% of the variance of totN concentrations between catchments (r^2) and the prediction ability (Q^2) was 66%. The catchment scores for each component (Figure 4a) clearly distinguished agricultural sites from natural and forestry-impacted sites. The Danish natural sites (labelled specifically because of deviations from other natural sites, see paragraph 3.1) also clustered together. The PLS model for totN included five explanatory variables with a VIP value above 1; the proportion of agricultural land cover, summer temperature and annual temperature, all of which were positively related, and forest cover and latitude, which were negatively related to totN (Figure 4b). This suggests that totN and totP concentrations were largely driven by a land-use gradient, from high percentage agriculture to high percentage forest. This land-use gradient is to a large extent also a climate gradient, with highest nutrient concentrations associated with warmer regions located in the

TABLE 2 Mean concentrations of totP, totN, TOC and suspended solids (SS), and mean fractions (in mass/mass) for DRP to totP, SS to totP, TON to totN, NO_3 to totN and NH4 tot totN, grouped by land use category and country (DEN, Denmark; FIN, Finland; NOR, Norway; SWE, Sweden), calculated from median concentrations for each site

			totP	DRP:TotP	SS:TotP	TotN	TON:TotN	NO_3:totN	NH_4:totN	TOC	SS
Category	Country	n	μg L^{-1}	g g^{-1}	g g^{-1}	μg L^{-1}	g g^{-1}	g g^{-1}	g g^{-1}	mg C L^{-1}	mg L^{-1}
AGR	DEN	5	107	0.38	0.15	4,298	0.54	0.81	0.02	n.d.	14
	FIN	6	90	0.24	0.28	2,694	0.29	0.55	0.04	13	17
	NOR	9	130	0.36	0.18	4,440	n.d.	0.77	n.d.	n.d.	11
	SWE	10	172	0.31	0.29	4,419	n.d.	0.85	0.00	9	38
FOR	FIN	8	22	0.25	0.21	584	0.77	0.14	0.02	16	4
	NOR	1	8	n.d.	n.d.	345	0.92	0.02	0.05	17	n.d.
NAT	DEN	7	48	0.40	0.10	960	0.36	0.59	0.03	15	4
	FIN	4	7	0.18	0.09	240	0.95	0.01	0.01	11	1
	NOR	7	3	n.d.	n.d.	226	0.79	0.17	0.03	3	n.d.
	SWE	12	7	0.42	0.15	318	0.90	0.05	0.03	9	1

Note: Note that full records were not available for each variable (Supplementary information, Table 2).
Abbreviations: AGR, agricultural; FOR, forestry-impacted; n, number of sites; NAT, natural; n.d., no data.

southern part of the Nordic countries. By contrast, the natural land cover types, that is, forests, peatlands, lakes and shrublands were associated with lower nutrient concentrations and were grouped together with precipitation and runoff, demonstrating that natural and more nutrient-poor catchments are located in colder and wetter regions of the Nordic countries. Thus, climate and land use are confounded which implies that identification of separate impacts of these two factors is challenging.

The PLS analysis and the correlation matrix (Figure SI-2) were consistent in identifying percentage agriculture as the most important driver of totN and totP concentrations, while %forest had the opposite effect. However, %agriculture and %forest were strongly negatively correlated. In addition, the PLS identified positive relationships between nutrient concentrations and (annual and summer) temperature and negative relationships with runoff. Using a simple linear regression analysis, we tested the explanatory power of single drivers for variation in totN and totP concentrations and fluxes within each land use category (Figure 5). Significant ($p < .05$) relationships between nutrient concentrations and summer temperature were found for the natural (totN: r^2 0.50; totP: r^2 0.50) and forestry-impacted catchments (totN: r^2 0.79; totP: r^2 0.50) (Figure 5a,b), but not for agricultural catchments. In the natural catchments, summer temperature was positively correlated with log-transformed concentrations of TON and TOC (data not shown, TON: $r^2 = 0.48$, $p < .0001$; TOC: $r^2 = 0.43$, $p < .0001$).

There were no significant correlations between annual mean runoff and fluxes of totN and totP (SI Figure 2), which surprised us because discharge is used to calculate element fluxes. Possibly, differences in seasonality of runoff and nutrient concentrations are stronger controls of the total annual fluxes than the annual runoff itself. In managed catchments, fertilizer application and remineralization of crop residues are important causes of seasonal variation in nutrient concentrations. Within each land-use category, positive relationships between runoff and fluxes of totN ($r^2 = 0.26$) and totP ($r^2 = 0.59$) were found for agricultural catchments and for fluxes of totN in natural catchments ($r^2 = 0.35$) (Figure 5c,d).

Natural catchments had less variation in soil type than agricultural catchments (SI Table 1), that is, they were dominated by moraine soils while the agricultural sites had clay, loam, peat and sandy soils. Grouping of sites by management and soil type did not result in a better explanation of factors driving N and P export (data not shown).

3.3 | Trends in concentrations and fluxes

We used annual mean flow-weighted concentrations and annual medians of measured concentrations as basis for trend estimates and found no significant differences using a pair-wise comparison (data not shown). Consistent with the other results reported, we continue with reporting trends based on the median concentrations. Absolute and relative trends in both fluxes and concentrations of totP and totN showed considerable variation between land use categories and countries (Figure 6), without the emergence of clear spatial patterns. Out of 69 sites, 22 and 25% had significant change in totN and totP concentrations, respectively. For totN concentrations, negative significant trends exceeded positive trends ($p < .05$, 4 positive, 11 negative). In addition, for totP, negative trends were more frequent than positive trends ($p < .05$, 6 positive, 11 negative). Fewer sites displayed significant changes in fluxes (totN: $p < .05$, 6%; totP: $p < .05$, 9%) and runoff ($p < .05$, 9%) The forestry-impacted sites had a relatively high number of significant trends (three and four out of nine sites had significant change in totN and totP concentrations, respectively), and the directional change of the significant totN and totP trends was consistent in each site.

Patterns of changes emerged when testing concentrations and fluxes of totN, totP, NO_3, and DRP for each land use category in a

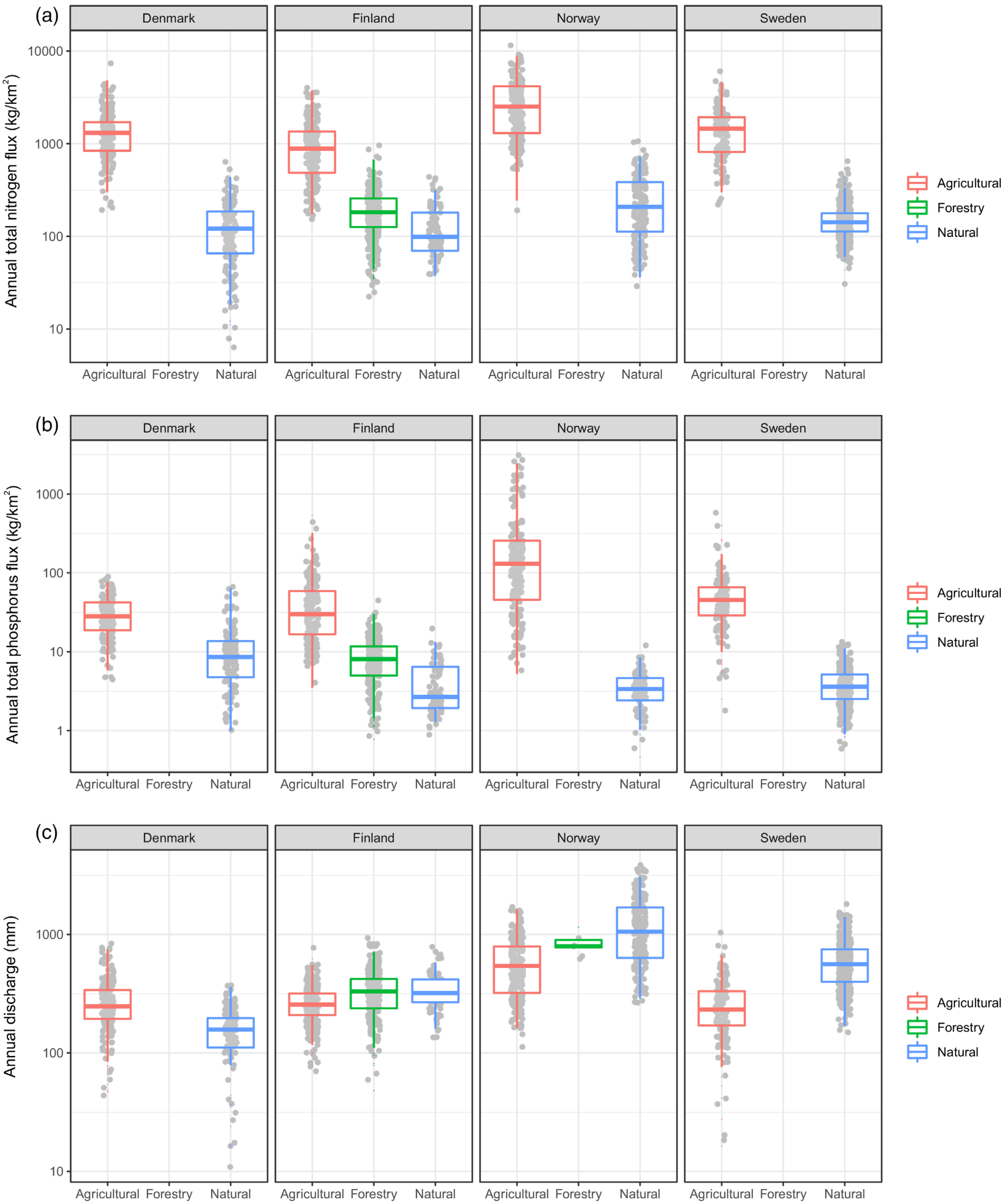

FIGURE 3 Flux and annual discharge ranges for the grouped sites, for median annual totN and totP flux (kg km^{-2}) from agricultural (n = 30), forestry (n = 9) and natural (n = 30) sites for the period 2000–2018

regional Mann-Kendall test (Table 3). The sen-slopes in Table 3 are in absolute units, in contrast to the trends in Figure 6. In agricultural sites across the Nordic countries, there was a significant ($p < .01$) downward trend in concentrations of totN (−15 μg L N $^{-1}$ year^{-1}) and a weakly significant downward trend in NO_3 ($p < .1$) (−9 μg L N $^{-1}$ year^{-1}). The natural sites show significant downward trends in

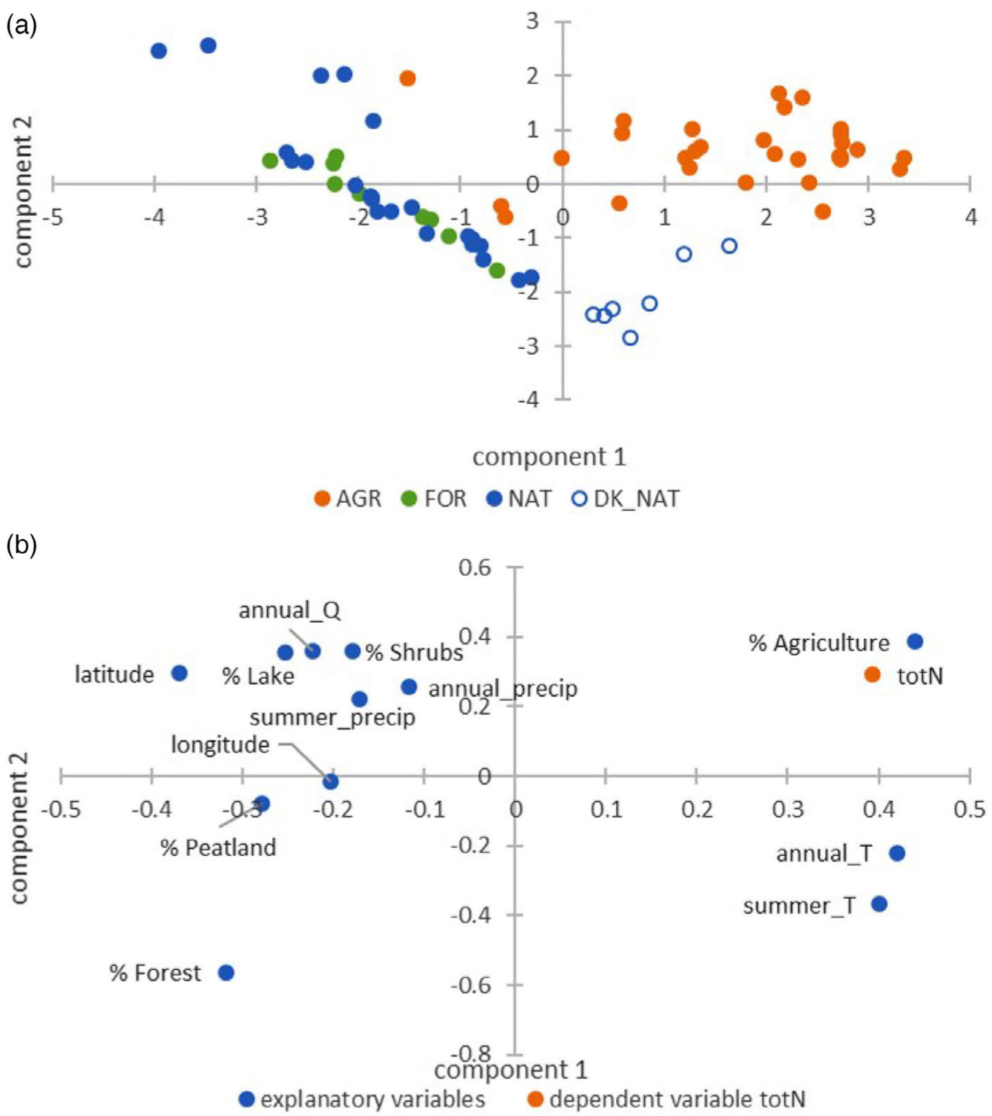

FIGURE 4 (a) Scoring of each single observation (catchments) for the two first components; (b) Loading of the explanatory variables and the dependent variable totN concentration for the first two components

concentrations of NO_3, totP and DRP. The sen-slope for NO_3 (−0.42 μg N L^{-1} $year^{-1}$) was similar to the sen-slope in totN (−0.45 μg N L^{-1} $year^{-1}$) for natural sites. Results from agricultural and natural sites suggest that most of the changes in totN were related to changes in NO_3 (Table 3).

In agricultural sites, significant downward trends in concentrations of P-species in Denmark (−0.3 μm L^{-1} $year^{-1}$ DRP) contrasted with significant upward trends in Norway and Sweden (+1.2 and +0.6 μm L^{-1} $year^{-1}$ DRP, respectively), resulting in a lack of change for all agricultural sites combined. Trends in N-species concentrations were mostly downward across all countries. In the natural sites, only Sweden had a significant downward change in totN concentrations.

With regard to fluxes, the regional M-K results (Table 4) indicated a significant decline in export of NO_3, at rates of −6.7 and −0.25 kg N km^{-2} $year^{-1}$, for agricultural and natural catchments, respectively, consistent with the trend in NO_3 concentrations but not with the trend in totN concentrations. Export of totP showed a strong positive trend ($p < .05$, 0.4 kg P km^{-2} $year^{-1}$) for agricultural sites despite the variations in trends strength between countries, and the contrasting results for totP concentrations. For all natural sites combined, the export of DRP declined significantly ($p < .05$, −0.02 kg P km^{-2} $year^{-1}$).

Thus, the regional M-K results indicated downward trends in the Nordic countries in N-species across land use categories, downward trends in concentrations of P-species in natural and forestry-impacted sites, and increases in totP export from agricultural catchments.

In a correlation matrix, we tested if the trends in concentrations and fluxes of N and P species could be related to trends in climate (temperature, precipitation) and hydrology (discharge) within each land use category but except for forestry-impacted sites, few significant correlations were found (Figure SI-3). In the forestry-impacted sites (n = 8), trends in runoff correlated negatively with trends in nutrient species, but its significance largely depended on one site and we do not regard this result as particularly robust. Thus, regional patterns of change in totN and totP concentrations for 2000 to 2018 could not be explained with simple relationships with climate and runoff.

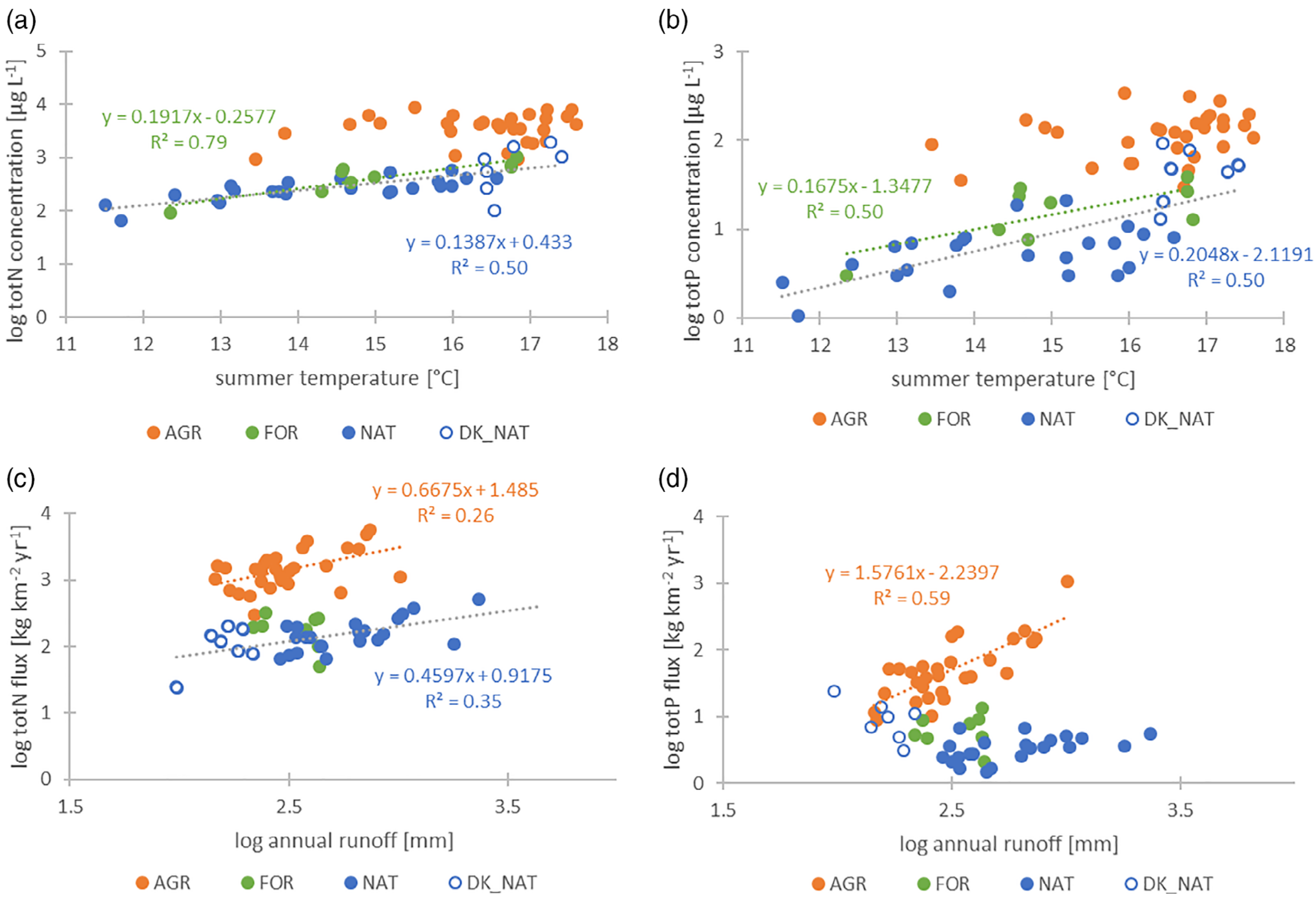

FIGURE 5 Concentrations of totN and totP (log-transformed) versus summer T (panel a, b); fluxes of totN and totP (log-transformed) versus annual runoff (log transformed) (panel c, d). Only significant regression lines for each land use category (significant slopes, $p < .05$) are shown. AGR, agricultural catchments; FOR, forestry-impacted catchments; NAT, natural catchments; DK_NAT, natural catchments in Denmark (included in the NAT regression equations)

4 | DISCUSSION

Long-term water quality and hydrological records from headwater Nordic catchments spanning large environmental and climate gradients offer a unique possibility to evaluate combined effects of land use, land cover and climate on surface water quality in managed and unmanaged landscapes. Most often, long-term assessments focus on a dominant land use, either agricultural (Blann, Anderson, Sands, & Vondracek, 2009; Pengerud et al., 2015), forestry-impacted (Kreutzweiser et al., 2008) or natural catchments (Vuorenmaa et al., 2018). Significant proportions of nutrient loadings from Nordic countries to marine recipients originate from agriculture, natural and semi-natural ecosystems (HELCOM, 2018; Lepisto, Granlund, Kortelainen, & Raike, 2006) suggesting that changes in nutrient runoff from both managed and unmanaged ecosystems is pertinent to the ecological status of receiving waters.

We found that long-term averaged nutrient concentrations and export were an order of magnitude higher from agricultural catchments compared with forestry-impacted and natural catchments, and that forestry-impacted catchment delivered significantly more nutrients than natural catchments. The high nutrient export from agricultural catchments is primarily driven by long-term surpluses of N and P, as indicated by statistics on gross nutrient balances (calculated from inputs of manure and fertilizer and removal from harvest) for agricultural land which vary roughly between 30 and 120 kg ha year^{-1} for N, and 0 to 12 kg ha year^{-1} for P in the Nordic countries after 2000 (Eurostat, 2020). In natural catchments, the main source of nutrient loading is atmospheric deposition, typically between 1 and 10 kg N ha^{-1} year^{-1} (Vuorenmaa et al., 2017) and usually retained for 90% within the catchment (Vuorenmaa et al., 2017; Watmough et al., 2005). In some agricultural catchments, losses of nitrogen may be close to a steady-state between inputs and outputs (Basu, Thompson, & Rao, 2011; Thompson, Basu, Lascurain, Aubeneau, & Rao, 2011). However, catchment characteristics such as tile drainage, topography, texture and mitigation measures to reduce nutrient runoff will control the fate of the nutrient excess, that is, runoff, groundwater or soil storage (Hellsten et al., 2019; Kronvang et al., 2005a; Kronvang, Vagstad, Behrendt, Bogestrand, & Larsen, 2007). Forest management typically consists of a mosaic of numerous treatments (harvesting, drainage, fertilization, soil tillage) with considerable

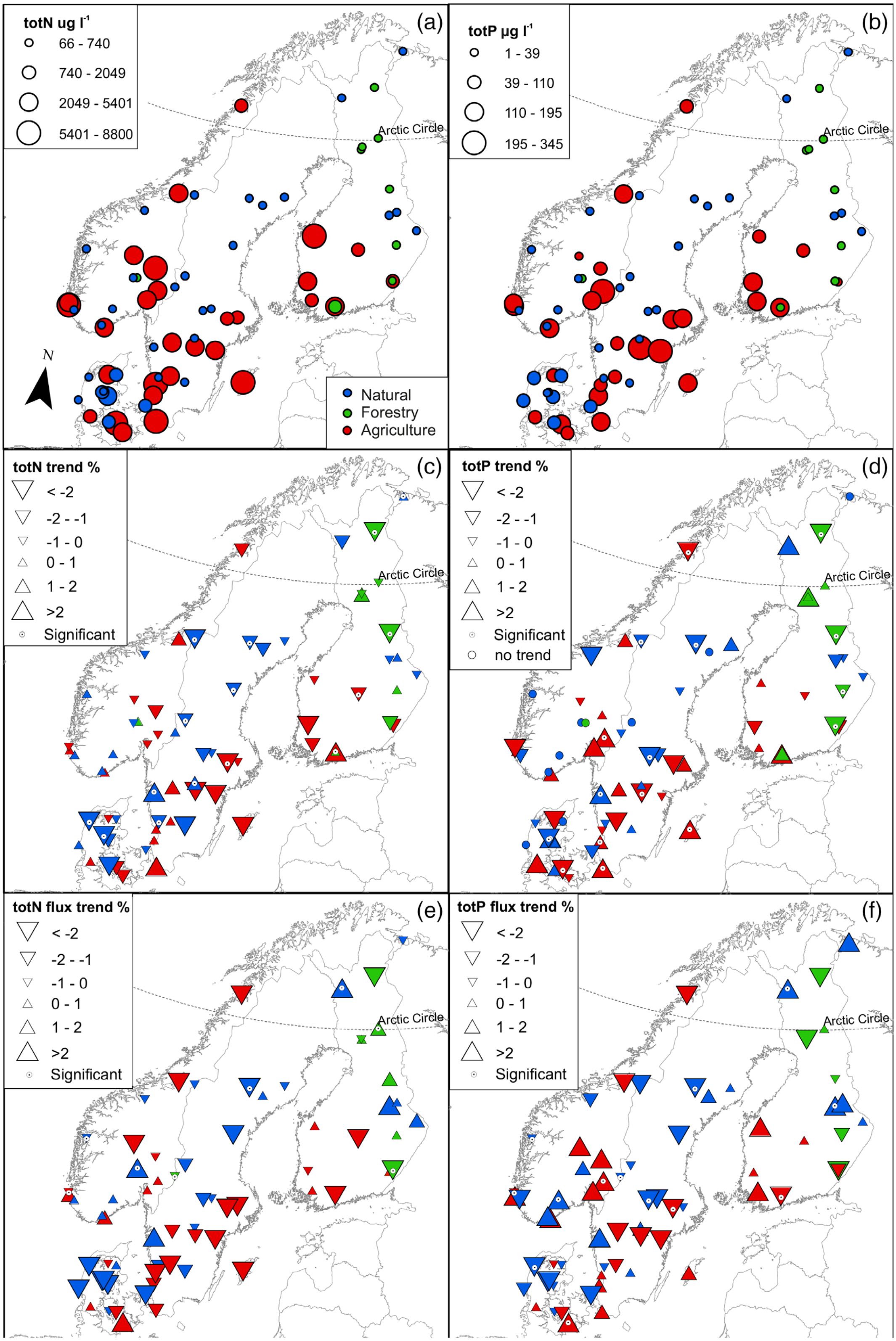

FIGURE 6 Map showing annual median concentrations of (a) totN and (b) totP, (c) trend of annual median totN concentration (% $year^{-1}$), (d) trend of annual median totP concentration (% $year^{-1}$), (e) trend of annual flux for totN (% $year^{-1}$) and (f) trend of annual flux for totP (% $year^{-1}$) for the period 2000–2018

TABLE 3 Results of regional Mann-Kendal test for concentrations of totN, NO_3 (in μg N L^{-1} $year^{-1}$), totP and DRP (in μg P L^{-1} $year^{-1}$), grouped by agricultural (AGR), Forestry-impacted (FOR), Natural NAT) sites and country

		Denmark slope	p	Finland slope	p	Norway slope	p	Sweden slope	p	All slope	p
AGR	totN	−8.9	n.s.	−22.6	*	−24.0	*	−7.1	n.s.	−15.2	**
	NO_3	−4.4	n.s.	−25.0	*	−13.9	n.s.	31.3	n.s.	−9.5	n.s.
	totP	−0.8	**	−0.3	n.s.	1.2	n.s.	1.6	n.s.	0.1	n.s.
	DRP	−0.3	***	−0.2	n.s.	0.6	*	1.2	**	0.0	n.s.
NAT	totN	−2.7	n.s.	−0.4	n.s.	0.5	n.s.	−1.8	**	−0.45	n.s.
	NO_3	−2.7	*	0.0	n.s.	−0.4	**	−0.6	***	−0.42	***
	totP	0.3	n.s.	−0.1	*	0.0	n.s.	−0.02	n.s.	−0.01	*
	DRP	−0.05	n.s.	n.d.		n.d.		−0.10	***	−0.03	***
FOR	totN	n.d.		−1.7	n.s.	n.d.		n.d.			
	NO_3	n.d.		0.3	n.s.	n.d.		n.d.			
	totP	n.d.		−0.1	*	n.d.		n.d.			
	DRP	n.d.		n.d.		n.d.		n.d.			

Note: Significance levels: *<.05, **<.01, ***<.001, n.s., p > .05. n.d., no data.

TABLE 4 Results of regional Mann-Kendal test for fluxes of totN, NO_3 (in kg N km^{-2} $year^{-1}$), totP and DRP in kg P km^{-2} $year^{-1}$), grouped by agricultural (AGR), Forestry-impacted (FOR), Natural NAT) site and country

		Denmark slope	p	Finland slope	p	Norway slope	p	Sweden slope	p	All slope	p
AGR	totN	0.34	n.s.	0.35	n.s.	−5.15	n.s.	−27.6	*	−8.46	n.s.
	NO_3	−7.21	n.s.	−3.23	n.s.	−2.73	n.s.	−18.1	n.s.	−6.72	*
	totP	0.13	n.s.	0.52	*	2.33	***	−0.66	n.s.	0.44	**
	DRP	0.01	n.s.	0.021	n.s.	0.507	**	−0.17	n.s.	0.055	n.s.
NAT	totN	−0.67	n.s.	1.08	n.s.	2.42	**	−0.51	n.s.	0.18	n.s.
	NO_3	−0.99	*	−0.018	n.s.	−0.17	n.s.	−0.29	***	−0.25	***
	totP	−0.16	n.s.	0.035	n.s.	0.055	*	0.00	n.s.	0.007	n.s.
	DRP	−0.02	n.s.	−0.0032	n.s.			−0.038	**	−0.022	*
FOR	totN			1.44	n.s.					1.44	n.s.
	NO_3			0.045	n.s.					0.045	n.s.
	totP			−0.044	n.s.					−0.044	n.s.
	DRP			n.d.						n.d.	

Note: Significance levels: *<.05, **<.01, ***<.001, n.s., p > .05. n.d., no data.

temporal and spatial variations (Ahtiainen & Huttunen, 1999; Kreutzweiser et al., 2008; Tattari et al., 2017). In forestry-impacted catchments, elevated nutrient runoff compared with natural catchments can be related both to application of fertilizer and to mobilization of soil nutrient stores.

When considering all land use categories simultaneously, we found that spatial variations in site-specific totN and totP concentrations were largely driven by a land-use gradient which partly overlapped with a climate gradient, demonstrating that climate and land use are confounded. The highest nutrient concentrations were associated with warmer, agricultural regions located in the south. Natural land cover types (i.e., forests, peatlands, lakes and shrublands) with lower nutrient concentrations were associated with cooler and wetter conditions. Agricultural land cover was the single-most powerful explanation for describing spatial variation in nutrient concentrations and can be considered as a proxy for gross nutrient balances as discussed earlier. Consistent with our study, totN and totP concentrations in Norwegian lakes, including natural and agriculturally impacted systems, were found to be positively related to terrestrial productivity and negatively to runoff (Hessen, Andersen, Larsen, Skjelkvale, & de Wit, 2009). Hessen and co-authors also highlighted nitrogen deposition as a strong driver of aquatic concentrations of N-species. This factor is likely to be most easily detectable in catchments with low nitrogen retention capacity, that is, with little soil and vegetation cover (Kaste, Austnes, & de Wit, 2020).

The positive correlations between spatial variation in totN concentrations and summer temperature in the natural and forestry-impacted catchments were mirrored by positive correlations between TON and summer temperature, and TOC and summer temperature. This suggests these correlations are a demonstration of the strong links between the element cycles of nitrogen and carbon in forested ecosystems (Mattsson, Kortelainen, & Raike, 2005). Surface water concentrations of dissolved organic matter are highest in carbon-rich catchments (Sobek, Tranvik, Prairie, Kortelainen, & Cole, 2007), which are found in Nordic regions with higher average temperatures (Callesen et al., 2003). Temperature, particularly during summer season, can be interpreted as a proxy for terrestrial productivity at least in climate where moisture is mostly not a limiting factor (Piao et al., 2011). By contrast, significant relationships between nutrient concentrations and summer temperature were not found in agricultural catchments, suggesting that combined effects of crop, management practices and soil type are stronger controls on element cycling than temperature in these systems (Bechmann et al., 2008).

Danish natural catchments had three to five times higher nutrient concentrations than other Nordic natural catchments, which could be indicative of profound differences in natural reference conditions (Skarbovik et al., 2020). The EU WFD (EC, 2000) defines reference conditions as “water bodies with no, or only very minor, anthropogenic alterations compared with conditions normally associated with undisturbed conditions”. However, legacies of former land use (Hamilton, 2012) combined with lateral groundwater flow (Brunke & Gonser, 1997) in flat landscapes complicate interpretations of catchment effects on streamwaters. Geology, soils, climate and land use history in Denmark contrast with other Nordic countries (Emanuelsson, 2009). Deeper, sandy soils which predominate in the flat Danish landscape are probably associated with a relatively larger proportion of precipitation feeding groundwater, while the Fennoscandian shield is characterized by thinner soils, greater relief and more superficial hydrological pathways. Most loamy and clayey agricultural soils in Denmark are tile-drained which directs part of the precipitation surplus directly to surface waters (Møller, Børgesen, Bach, Iversen, & Moeslund, 2018). Skarbovik et al. (2020) also suggest that bank erosion may be more important in Danish than in other Nordic streams. In Norway and Sweden, agricultural soils are often tile-drained, especially clayey soils with low hydraulic conductivity that are more exposed to the risk of surface runoff. Steeper slopes in Norway also contributes to higher erosion and P export (Bechmann et al., 2008). In addition, higher temperatures and longer growing seasons make evaporative losses relatively more important in the hydrological cycle in southern parts of the Nordic countries (Kortelainen, Saukkonen, & Mattsson, 1997). The different composition of totN and totP species in agricultural and natural sites suggests a wide variation in susceptibility to hydrological and management impacts on their transport and leaching, for example, diffuse and particulate transport, and transport of nutrients in inorganic versus organic forms. Variation in soil type and texture was higher in agricultural catchments, with clay, loam, peat and sandy soils than in (semi-) natural catchments which were dominated by moraine soils.

We found a general decline in concentrations and fluxes of total nitrogen and NO_3 from agricultural and natural catchments in the Nordic region as a whole, for the period 2000–2018. These downward trends for agricultural catchments agree with the downward trends found in the gross N budget for agricultural land during the period 2000–2016, which is highest for Sweden (−27%) and lowest for Norway (−3%), with Denmark (−15%) and Finland (−14%) being in between (Eurostat, 2020). However, the reductions in both gross N budget and stream concentrations and fluxes of N were substantially higher during the 1990s in Denmark, Finland and Sweden (Hellsten et al., 2019; Windolf, Blicher-Mathiesen, Carstensen, & Kronvang, 2012). The reduction in gross N budget was driven by intensive mitigation campaigns to minimize N losses, especially in Denmark (Hansen, Thorling, Schullehner, Termansen, & Dalgaard, 2017; Kronvang et al., 2005b) and Sweden, where catch crop and spring ploughing were implemented (Folster et al., 2014). The totN load to 10 estuaries (catchments covering 35% of the Danish land area) decreased by 39% during the period 1990–2009 (Windolf et al., 2012) following mandatory national regulations on agricultural production (Kronvang et al., 2005b). Agricultural extensification in two Norwegian sites reduced totN export here, although there was little national focus on mitigation measures to reduce nitrogen losses (Hellsten et al., 2019). Earlier studies demonstrated a predominance of declines in nitrogen export from agricultural headwater catchments in Denmark and Sweden, but lack of change in Norway (Kyllmar et al., 2014a; Stålnacke et al., 2014). Changes in fertilizer application explained downward totN trends in Finnish agricultural catchments while upward trends were related to crop distribution (Tattari et al., 2017).

Declines in NO_3 concentrations in natural catchments dominated over change in totN. In natural catchments, most totN consists of organic N and the dynamics of organic N are closely linked to those of DOM, which is usually elevated during the summer and autumn (Lepisto, Kortelainen, & Mattsson, 2008) whereas NO_3 is highest during the dormant season (de Wit et al., 2008) and likely to be more strongly linked to atmospheric deposition (Vuorenmaa et al., 2018). The widespread decline in annual totN concentrations in agricultural and natural sites could not be explained by simple relationships with climate or runoff. Spatial variation in totN export was strongly related to annual runoff in agricultural catchments, however, and implies that increases in runoff could lead to increased element export (e.g., Oygarden et al., 2014). It is likely that investigations of climatic and hydrological impacts on water quality and element export would benefit from a focus on seasonal trends and/or from including a longer time period (de Wit et al., 2008; Jeppesen et al., 2009; Jeppesen et al., 2011; Wenng et al., 2020).

Given the profound contrasts in N concentrations between land use categories, and the lack of correlations between trends in N species and trends in climatic and hydrological variables, it is likely that the decline in N species is the concerted effect of various interplaying factors, including environmental policy. Other studies document long-term declines in reactive nitrogen from unmanaged and managed Nordic landscapes (Garmo et al., 2014; Rekolainen, Mitikka, Vuorenmaa, & Johansson, 2005) which are likely caused by changes in N deposition and climate-mediated changes in hydrology (de Wit et al., 2008; Kaste et al., 2020; Vuorenmaa et al., 2018). The decline in N deposition itself is related to reduced emissions to the atmosphere (Grennfelt et al., 2020). Lucas et al. (2016) suggested that increased forest growth was responsible for declining NO_3 concentrations in rivers draining natural and forestry-impacted landscapes in Northern Sweden, an area that receives relatively low levels of N deposition. Substantial increases in forest standing stock are common in Finland, Sweden and Norway (de Wit, Austnes, Hylen, & Dalsgaard, 2015; Luyssaert et al., 2010) suggesting that increased nitrogen uptake by forests may affect NO_3 leaching from forested catchments elsewhere in the Nordic region. In Chesapeake Bay, USA, a watershed with considerably higher proportion of agriculture and developed areas than the Nordic countries, declines in atmospheric N deposition were found to be the second-most important factor responsible for the decrease in N export to the bay (Ator, Garcia, Schwarz, Blomquist, & Sekellick, 2019).

Contrary to Lucas et al. (2016), Räike et al. (2020) suggest that forestry practices in Northern Finland have increased N runoff to the Baltic through increasing the area of ditched peatlands to promote forest growth, which has led to increases in organic N (Tattari et al., 2017). Forestry-impacted catchments exported on average over 40% more N than natural catchments, which suggests that forest management could substantially increase levels of totN in surface waters. Forest management-induced increases in N export from, for example, clear-cutting, soil preparation and ditching are well-documented (De Wit et al., 2014; Kreutzweiser et al., 2008; Nieminen et al., 2018), but the duration of increased nutrient export following disturbance is usually short relative to the forest rotation cycle (Sponseller et al., 2016). Furthermore, appropriate, context-sensitive (Ring et al., 2017) forest management can safeguard water quality (Sundnes et al., 2020; Sponseller et al., 2016).

Patterns of long-term change in totP concentrations and export were more varied temporally and spatially than for nitrogen; Denmark and Finland had significant declines while Sweden and Norway had significant increases in DRP, resulting in an overall neutral trend for all agricultural catchments (Kronvang, Tornbjerg, Hoffmann, Poulsen, & Windolf, 2016). To what extent climate can explain the patterns of long-term change in P export is unclear. Management of agricultural P-losses is complicated by increased precipitation intensity with subsequent increased erosion (Farkas, Beldring, Bechmann, & Deelstra, 2013) combined with a strong legacy of soil-P in the fields (Sharpley et al., 2013). Effects of mitigation of phosphorus sources might thus take several decades to document in monitoring programs (Bol et al., 2018; Mellander et al., 2018). In Norway, agricultural mitigation measures focused primarily on phosphorus, however, measures implemented to reduce totP export were changed in 2013 from general measures to measures only for high risk areas and increasing focus on production (Bechmann, Greipsland, & Falk Øgaard, 2019). Removal of subsidies to abstain from autumn tillage, followed by an increase in autumn ploughing and erosion appears to be an explanation for increases in totP export in two Norwegian catchments (Bechmann et al., 2020). Hardly any sign of significant change was found in the forestry-impacted catchments except for reduced P concentrations likely related to reductions in P fertilizing (Tattari et al., 2017). There was a small overall decline in totP for natural catchments, primarily related Finland and Sweden. Huser, Futter, Wang, and Folster (2018) suggest that the totP-decline in Swedish lakes could be a combined effect of climate change and increased uptake of phosphorus by forests, similar to the explanation provided for declining NO_3 runoff in northern Swedish rivers (Lucas et al., 2016). Increased forest growth may explain trends in Finnish natural catchments while the lack of change in totP in Norwegian natural catchments could be related to a lower proportion of forest in these catchments. The significant decline in P export from Finnish forestry-impacted catchments was, however, attributed to reduced fertilizer use.

The lack of widespread reductions in totP export is substantiated by long-term records from rivers in Finland (Räike et al., 2020) and Norway (Skarbovik, Stalnacke, Kaste, & Austnes, 2014), and from marine recipients (Frigstad et al., 2020; Kuss et al., 2020). Our study implies that mitigation of P is more challenging than for N, because of more variation in sources (Kronvang et al., 2007), complexity of hydrological pathways (Mellander et al., 2018) and delays in responses to mitigation as a consequence of legacy pools of phosphorus in soils (Bol et al., 2018; Jeppesen et al., 2009) and lake sediments (Couture et al., 2018).

Nutrient export from agricultural, forestry-impacted and natural ecosystems, in declining order of importance, is a strong control of freshwater and marine ecological status in the Nordic countries. Mitigation measure have been effective especially for reduction of nitrogen runoff, but the effect of mitigation can be counteracted by climate change (Crossman et al., 2013). If the green shift will be associated with intensification of agricultural and forest production and increased use of fertilizer, this will pose new challenges for protection of water quality (Marttila et al., 2020). Long-term monitoring records of small headwaters under varied combinations of land use, climate and land cover are valuable and necessary for assessing combined effects of stressors on water quality and nutrient cycling and retention at the landscape level. We recommend sustained funding of long-term monitoring of managed and unmanaged, natural catchments. Further analysis should consider (a) further inclusion of nutrient species (e.g., nitrate, particulate P, organic forms, and so forth) for investigation of possible contrasting responses to climate, runoff and mitigation and their impacts on aquatic ecology, (b) examine long-term patterns in seasonal variation, (c) incorporate information about nutrient inputs (including atmospheric deposition), and agricultural and forestry management.

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DATA AVAILABILITY STATEMENT

The raw data are openly available in a public repository that does not issue DOIs. The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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SUPPORTING INFORMATION

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