

# ICP Waters Report 146/2021

## Effects of nitrogen on nutrient-limitation in oligotrophic northern surface waters



Photo: Jens Prohn

International Cooperative Programme on Assessment  
and Monitoring Effects of Air Pollution on Rivers and Lakes

Convention on Long-Range Transboundary Air Pollution



# REPORT

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<p>Summary</p> <p>Nitrogen deposition has a potentially eutrophying impact on oligotrophic freshwaters. Here we present a data analysis and a literature review with a joint focus on nitrogen (N) limitation in oligotrophic lakes and rivers. The data analysis consists of a Nordic lake dataset where relations between algal biomass (chlorophyll <i>a</i>), water chemistry and N deposition are investigated, and an analysis of element stoichiometry (nitrogen : phosphorus ratios) in Norwegian natural rivers. The literature review includes international publications on N limitation from a wide variation of monitoring and experimental studies. The data analysis and literature both support that N limitation exists in surface waters that receive low N deposition and that elevated N deposition can shift an ecosystem from N to P limitation. The results are used to review and revise ranges of critical loads of nitrogen for surface waters, as a contribution to the ongoing review and revision of empirical critical loads under the Convention on Long-range Transboundary Air Pollution.</p>
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INTERNATIONAL COOPERATIVE PROGRAMME ON  
ASSESSMENT AND MONITORING EFFECTS OF AIR  
POLLUTION ON RIVERS AND LAKES

**Effects of nitrogen on nutrient-limitation in  
oligotrophic northern surface waters**

Prepared at the ICP Waters Programme Centre  
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# Preface

The International Cooperative Programme on Assessment and Monitoring of the Effects of Air Pollution on Rivers and Lakes (ICP Waters) was established under the Executive Body of the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) in July 1985. Since then, ICP Waters has been an important contributor to document the effects of implementing the Protocols under the Convention. ICP Waters has prepared numerous assessments, reports and publications that address the effects of long-range transported air pollution.

ICP Waters and its Programme Centre is chaired and hosted by the Norwegian Institute for Water Research (NIVA), respectively. A programme subcentre is established at NORCE in Bergen. ICP Waters is supported financially by the Norwegian Ministry of Climate and Environment and the Trust Fund of the UNECE LRTAP Convention.

The main aim of the ICP Waters programme is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution, in particular acidification, on surface waters. More than 20 countries in Europe and North America participate on a regular basis.

ICP Waters contributes to joint assessments under the Convention. Currently, empirical critical loads for nitrogen are being reviewed and revised under the responsibility of the Centre for Coordination of Effects under the Task Force on Modelling and Mapping. This report contains an analysis of Nordic lake dataset, a Norwegian river dataset and a literature review focusing on effects of nutrient nitrogen on oligotrophic freshwaters that are impacted by atmospheric nitrogen deposition. The lake dataset contains water chemistry and biological data from lakes in Norway, Sweden and Finland and was compiled in 2017 with the aim to compare national classification of ecological status according to the Water Framework Directive. The compilation of the dataset was financed by the Norwegian Environment Agency and the Swedish Agency of Marine and Water Management. The river data derive from a Norwegian monitoring programme for “reference rivers”, funded by the Norwegian Environment Agency. The literature review is an update of ICP Waters report 101/2010.

Jan-Erik Thrane was primarily responsible for Chapter 2, while Heleen de Wit was responsible for Chapter 3. Kari Austnes advised on use of data and interpretation of results. We wish to thank those who helped with the data compilation, in particular Simon Hallstan (Swedish University of Agricultural Sciences, SLU). We would also like to thank Gaute Velle (NORCE), Jens Fölster (SLU), Alina R. Zurnachyan (Ministry of Environment, Armenia) and Dag O. Hessen (University of Oslo), who gave valuable inputs to the draft version of this report.

Oslo, December 2021

Heleen de Wit  
Chair of ICP Waters

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## Summary

Recent studies suggest that nitrogen deposition can impact ecosystems in natural, oligotrophic surface waters. Critical loads for nitrogen can act as a tool to support effect-based European policies for the abatement of air pollution. Under the Convention on Long-range Transboundary Air Pollution (CLRTAP), empirical critical loads for nitrogen are currently being revised. The previous revision was done in 2011, and new data and literature are now available.

Here, we present a two-fold contribution to the ongoing revision of critical loads of nitrogen, for surface waters. The first contribution consists of an analysis of a Nordic lake dataset (water chemistry and phytoplankton) and a dataset on water chemistry from Norwegian rivers. The second contribution is an update of a literature review presented in an ICP Waters report in 2010.

We used an existing database for the period 2006 to 2017 from 296 natural lakes in Sweden, Norway and Finland to investigate relations between chlorophyll *a* (*chl<sub>a</sub>*; a proxy for algal biomass), water chemistry (total phosphorus (TP), total nitrogen (TN), dissolved inorganic nitrogen (DIN)), and nitrogen (N) deposition. We found that the amount of *chl<sub>a</sub>* overall correlated better with TP than with TN, but that TN explained more of the variation in *chl<sub>a</sub>* than TP at low TN:TP and DIN:TP ratios. This indicates that N limitation becomes progressively more important as the availability of N relative to P decreases. Using median values from 18 geographical regions, we found that the amount of *chl<sub>a</sub>* per unit of TP increased with N deposition up to deposition levels of circa 4 kg N ha<sup>-1</sup> yr<sup>-1</sup> and then levelled off. Even though the *chl<sub>a</sub>*:TP was highly variable within regions, the pattern illustrates that N limitation is common in lakes below these deposition levels and that the lakes shift to P limitation at higher N deposition.

Regarding regional and seasonal patterns in nutrient limitation and stoichiometry in rivers, DIN:TP ratios and DIN concentrations indicated that N limitation is common in northern Norway. Most of rivers in the two northern ecoregions (N deposition 1.3 to 1.5 kg N ha<sup>-1</sup> year<sup>-1</sup>) were N limited for at least one month during the growing season from May to September based on stoichiometric ratios. About one third of the northern rivers were N limited in ≥ four of the growing season months. The rivers in central and eastern Norway (N deposition 2.9 to 3.2 kg N ha<sup>-1</sup> year<sup>-1</sup>) were a mix of both N and P limited, while the rivers in southern and western Norway (N deposition > 6.0 kg N ha<sup>-1</sup> year<sup>-1</sup>) were generally P limited. Low values of DIN:TP were primarily driven by retention of DIN by vegetation and soils in catchments, which resulted in DIN concentrations well below 10 µg/L from June to September in most rivers in the two northern ecoregions. In the ecoregion South, where N deposition was higher, DIN concentrations were above 30 µg/L in most rivers during the growing season.

The literature review presented evidence of nutrient limitation from various types of studies, including paleolimnological, whole-lake experiments, gradient studies and mesocosm- and in-situ bioassays. Many studies published post-2010 accept the mechanism of possible nitrogen limitation in oligotrophic surface waters. Evidence of effects of enhanced N availability is most abundant for phytoplankton, from studies in Europe and North America. Clear-water lakes appear to be more sensitive to N deposition than humic lakes, because of the generally lower N-retention capacity of their catchments (a function of vegetation and soil carbon pools) and because increased DOM contributes to light-limitation. Concentrations of DOM are positively associated with similar factors that impact N-retention capacity, i.e., soil and vegetation cover. Other organism groups (benthic algae, macrophytes) are also assumed to be sensitive to N. However, multiple factors control productivity and critical loads of N deposition are difficult to quantify.

The overall conclusions from this report are that there is now stronger agreement in the literature than in 2010 that N can be a limiting nutrient in oligotrophic surface waters, and that N limitation is common especially in northern, low N deposition, areas.

We propose, based on the data analysis presented in this report and on the literature review, a critical load (CL) range for nitrogen as a nutrient of 2-6 kg N ha<sup>-1</sup> year<sup>-1</sup>, where clear-water sub-Arctic and alpine lakes have a CL range of 2-4 kg N ha<sup>-1</sup> year<sup>-1</sup> and boreal (non-dystrophic) lakes have a CL range of 3-6 kg N ha<sup>-1</sup> year<sup>-1</sup>. Dystrophic, humic lakes in catchments with forests, wetlands and well-developed forest soils are less sensitive to N deposition because more N is retained in the catchment, while high DOM limits the eutrophying effects because of light limitation. Hence, their CL range is proposed to be 5-10 kg N ha<sup>-1</sup> year<sup>-1</sup>.

# 1 Introduction: Biological effects of nitrogen oligotrophic surface waters

The growth and biomass of autotrophs in freshwater, and especially phytoplankton in lakes, has generally been viewed as limited by phosphorus (P) and less by nitrogen (N) (Schindler, 1977; Schindler et al., 2008; Sterner, 2008). However, this view has been challenged in the last couple of decades. de Wit and Lindholm (2010) reviewed literature on eutrophying effects of N availability and N deposition on oligotrophic freshwater ecosystems. Much of the same literature is reviewed in (Keck and Lepori, 2012). The review summarized peer-reviewed studies that demonstrated that increased availability of N, related to atmospheric N deposition in nutrient-poor temperate, boreal and arctic lakes affect freshwater biology. The literature was grouped after type of evidence, i.e. on algal productivity from paleolimnological studies, whole-lake experiments, regional lake surveys and experimental nutrient additions (mesocosm studies and bioassays).

After a century of elevated anthropogenic N deposition (Holtgrieve et al., 2011), policies and measures for reducing N deposition have led to a significant decrease in N deposition over the last two decades (Torseth et al., 2012; Engardt et al., 2017). The concept of critical loads (CLs) is an important policy tool under the Convention on Long-range Transboundary Air Pollution (CLRTAP). A CL is defined as “a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Nilsson and Grennfelt, 1988). As stated by Forsius et al. (2021), “the CL concept has been used in policy negotiations by both the EU Commission and the Convention on Long-range Transboundary Air Pollution (CLRTAP), in which European maps of CLs and critical levels have been used to optimise emission reductions by connecting costs for reductions measures, emission scenarios, deposition and air quality modelling and CLs”. Critical loads can be modelled or based on empirical evidence, e.g. from N addition experiments. Ecosystem effects of anthropogenic emissions of N to the atmosphere, and associated inputs of DIN to terrestrial and aquatic ecosystems have been documented extensively (e.g. Dirnböck et al. (2014); Meunier et al. (2016)), but biological responses to reductions in N deposition are challenging to predict (Payne et al., 2017; Gilliam et al., 2019). As policy aims for emission reduction should be based on biological effects under CLRTAP, a continued evaluation of ecosystem responses to deposition impacts should be emphasized (Forsius et al., 2021).

The latest review and revision on empirical critical loads for the eutrophying effects of nitrogen was published in 2011 (Bobbink and Hettelingh, 2011). The review by de Wit and Lindholm (2010) fed into the chapter on inland surface water habitats in this report. Currently the empirical critical loads for nitrogen are being revised (Bobbink et al., in preparation). The aim of this report is to provide background for this ongoing revision. The contribution is two-fold. The first contribution (chapter 2) is updated information on effects of N on surface waters through an analysis of a Nordic lake dataset (water chemistry and biology) and of water chemistry of Norwegian rivers. The second contribution (chapter 3) is an update of the literature review by de Wit and Lindholm (2010), with a similar structure, but reviewing literature that was published from 2010 and onwards. Finally, updated empirical critical loads for nitrogen for surface waters are proposed.



## 2 Nutrient limitation in oligotrophic surface waters in relation to nitrogen deposition in the Nordic countries

### 2.1 Introduction

Nitrogen (N) has traditionally been considered less important than phosphorus (P) as a limiting element for the growth and biomass of autotrophs in freshwater, and especially for phytoplankton in lakes (Schindler, 1977; Sterner, 2008). Indeed, P is generally responsible for eutrophication of lakes (Schindler et al., 2008) and also shows a stronger correlation with phytoplankton biomass than N does across broad geographical scales (Dillon and Rigler, 1974; Phillips et al., 2008). A close relationship between total P and phytoplankton biomass is, however, expected, since a large fraction of the total P-pool in lakes usually is associated with algal cells (Lewis Jr and Wurtsbaugh, 2008). Hence, a strong correlation between the total P and biomass does not necessarily imply that P is a causally limiting factor. Reviewing a large body of experimental N and P addition studies, Elser et al. (2007) showed that simultaneous addition of both N and P gives higher growth responses than single N- or P additions for both lake phytoplankton and stream phyto-benthos (algae living on the surface of bottom substrates). The positive synergistic effect of N and P can likely be explained by the fact that addition of one element quickly shifts the autotrophs into limitation by the other element (Elser et al., 2007). This suggests that many natural (unpolluted) aquatic systems probably are close to the limit between N and P limitation, and ultimately, that both N and P can be limiting depending on the balance between supply and autotrophic demand (Lewis Jr and Wurtsbaugh, 2008).

Most lakes in the boreal and alpine parts of Europe are found in relatively natural catchments with little local influence from settlements or agriculture. Still, atmospheric deposition of long range transported pollution may significantly influence the water chemistry of these lakes, including the supply of dissolved inorganic nitrogen (DIN; Stoddard, 1994). In fact, there are indications that the increased atmospheric N deposition due to anthropogenic activity over the last century, and especially during the last 50-60 years (Stoddard et al., 2001; Holtgrieve et al., 2011), has shifted lakes in many areas from a natural state of N limitation to P limitation (Bergström and Jansson, 2006; Elser et al., 2009). In regions of low N deposition, e.g., in Northern Sweden and high mountains in the US, N limitation is common in lakes (Bergström and Jansson, 2006; Bergström, 2010; Isles et al., 2020) and streams (Myrstener et al., 2018). Additionally, atmospheric supply of inorganic N has contributed to increased phytoplankton biomass in unproductive lakes in these areas (Bergström and Jansson, 2006).

The likelihood of N vs. P limitation may be predicted from the nutrient stoichiometry, and a low ratio of DIN to total P (DIN:TP) is a good predictor of N limitation both in lakes (Bergström and Jansson, 2006; Bergström, 2010) and in coastal systems (Ptacnik et al., 2010). Concentrations of inorganic N in natural, oligotrophic surface waters depend on interactions between deposition, climate and catchment characteristics (Aber et al., 1989; Stoddard, 1994; Dise and Wright, 1995; Aber et al., 1998; Wright et al., 2001; Goodale et al., 2005; Wright et al., 2006; Austnes et al., in preparation) and are currently declining at many sites due to decreasing N deposition (Garmo et al., 2020). TP in unpolluted northern lakes is largely linked to DOM, and surface waters are currently browning (Monteith et al., 2007; de Wit et al., 2016). These developments are likely to result in lower DIN:TP ratios, especially in northern areas (Isles et al., 2018). As a result, increasing prevalence of N limitation may be expected,

and especially in surface waters where the N:P ratio in the water is close to the tipping point between N and P limitation.

In this analysis, we use an existing, large monitoring dataset of phytoplankton and chl $a$  from Fennoscandian lakes (Finland, Norway and Sweden) and a dataset of high temporal resolution on water chemistry from Norwegian rivers to assess 1) the effects of long-range transported N on phytoplankton biomass and patterns of nutrient limitation in lakes, and 2) seasonal and regional patterns in N:P stoichiometry and the potential for N limitation in rivers. Our analysis is guided by the primary focus of ICP Waters on natural ecosystems. We therefore focus on catchments with a minimum of direct human activities, such as settlements, agriculture or other anthropogenic factors that potentially impact local runoff of N and P to surface waters. Any ecosystem responses to N should therefore, in theory, be associated with atmospheric deposition. However, such responses can also be confounded by other factors, such as climate. We use the datasets to address the following main questions:

- 1) How do phytoplankton biomass (measured as chlorophyll  $a$  (chl $a$ )) in natural oligotrophic Fennoscandian lakes relate to concentrations of N and P?
- 2) Can we detect changes in the relationship between chl $a$  and N and P depending on the ambient N:P stoichiometry?
- 3) Does the amount of chl $a$  per unit P increase with DIN or N deposition, indicating that atmospheric deposition of N causes increased primary production in natural lakes?
- 4) How does the DIN:TP ratios in natural Norwegian rivers vary through the year, and how common (based on N:P stoichiometry) is N limitation relative to P limitation in different ecoregions in Norway?
- 5) Which variables (catchment, deposition, climate) can explain the variation in DIN concentrations and DIN:TP ratios in surface waters?

## 2.2 Materials and methods

### 2.2.1 Datasets

#### Lake data

To assess the effects of N on phytoplankton in lakes, we used a Nordic database that has been compiled from monitoring programs in Norway, Sweden and Finland (Fölster et al., 2021; we only used data from lakes, but the database also has data on rivers). The full lake dataset contains data on water chemistry, chlorophyll  $a$  (chl $a$ ) and phytoplankton biovolume from ca. 1975 to 2017. We decided to focus on data from the last 15 years, since i) this period contains data from a high number of lakes from all 3 countries, and ii) N deposition in the Fennoscandia has been relatively stable (although slightly declining) over this period (Aas et al., 2017; Andersson et al., 2018). Moreover, the data from Finland in the database was already aggregated post 2006. Hence, we used data from 2006 to 2017 for the analyses. The database contains more information on chl $a$  than phytoplankton biovolume, and we therefore used chl $a$  as the parameter representing phytoplankton biomass. The correlation between log(chl $a$ ) and log(phytoplankton biovolume) was high ( $R^2 = 0.79$ ,  $n = 3672$ ). N deposition data as estimated by the EMEP<sup>1</sup> model was used (sum of wet and dry deposition of oxidized and reduced N). The model is a chemical transport model for estimating air concentrations and deposition based on emissions and meteorological data (Simpson et al., 2012; Tsyro et al., 2020). Data are available on a 0.1 x 0.1 long-lat grid ([https://emep.int/mscw/mscw\\_moddata.html](https://emep.int/mscw/mscw_moddata.html)), and each lake station was

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<sup>1</sup> [Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe](https://emep.int/mscw/mscw_moddata.html)

assigned the deposition of the grid cell in which it is located. We calculated the mean N deposition for 2006 to 2017 (same period as water chemistry data; see above) for all lakes. Catchment cover (% forest, agriculture and urban or artificial surfaces) was obtained from the Corine database (European Environment Agency<sup>2</sup>).

The original lake dataset is heterogeneous with respect to sampling frequency (the number of years and dates per year), sampling depths, parameters, and the number of sampling stations per lake. Further, the full dataset contains lakes along the whole spectrum of catchments from no human activities to catchments strongly impacted by agriculture and pollution sources related to high population density (sewage, developed areas, etc). Since we were only interested in catchments with natural land cover, we excluded lakes with > 1% agriculture or urban catchment cover. We further aggregated the data as follows: 1) We only kept samples with data on chl<sub>a</sub>, total phosphorus (TP), total nitrogen (TN) and dissolved inorganic N (DIN; NO<sub>3</sub>-N + NH<sub>4</sub>-N). Values that were recorded as below the detection limit were set to half the detection limit. 2) To ensure that we only analyzed data from the epilimnion, we only included samples taken at 5 m water depth or shallower. If there was more than one sample from 0-5 m at a given sampling date (e.g., 0.5, 2, and 5 m), we calculated a mean value from the depths. Over 95% of the Norwegian chl<sub>a</sub> data and 70 % of the Swedish chl<sub>a</sub> data were from 0-5 m. Data from Finland was already aggregated for the epilimnion prior to data submission, or from composite samples from the epilimnion. 3) The epilimnion data was further subset to the “algal growing season”, which we defined as 1<sup>st</sup> of June to 30<sup>th</sup> of September. We are aware that the extent of the growing season varies with latitude and altitude, and hence, in northern high-altitude lakes, the growing season may be considerably shorter. Still, as the data stem from monitoring programs, we assume that most samples containing data on chl<sub>a</sub> were taken during the local growing season. Only stations with ≥ 2 sampling dates within this period were included. 4) Based on the growing season data, we calculated yearly median values for each sampling station (some lakes had more than one station). Then, a mean for each station was calculated for all years after year 2006 (for Finland, averages for 2006-2012 was used because there were no data after 2012). Finally, to obtain one value per lake, we calculated the mean for all stations within each lake. Most lakes had only one sampling station. Six outlier lakes with mean TP > 50 µg/L were removed.

The aggregated Nordic lake dataset contained mean summer epilimnion values of chl<sub>a</sub>, TP, TN and DIN (NO<sub>3</sub>-N+NH<sub>4</sub>-N) for 2006-2017 from 296 natural lakes (181 in Finland, 89 in Sweden and 26 in Norway; Figure 1). Most of the lakes were oligo-mesotrophic, with chl<sub>a</sub> concentrations between 2-8 µg/L and TP concentrations between 6-18 µg/L (Table 1). The levels of DIN ranged between 6 and 21 µg N/L for most lakes (Table 1).

**Table 1.** Summary statistics for chl<sub>a</sub>, nutrients and nutrient ratios from the 296 natural lakes. TP = total phosphorus, TN = total nitrogen and DIN = dissolved inorganic nitrogen. All variables have units of µg/L; ratios are by weight.

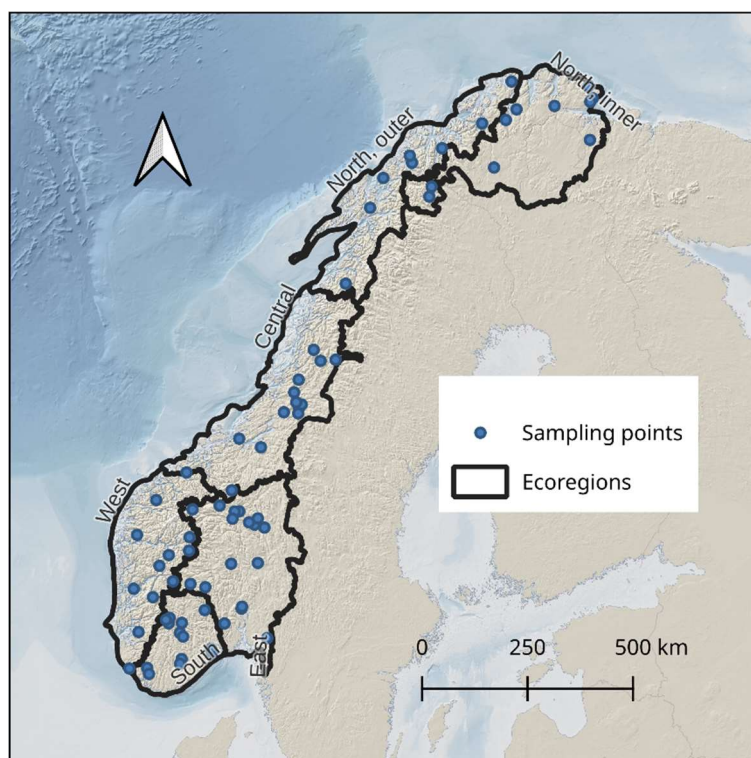
Parameter	min	Q1	median	Q3	max
Chl <sub>a</sub>	0.26	1.9	3.5	7.9	44
TP	1	5.5	9.8	18	46
TN	87	255	324	414	830
DIN	2.9	5.9	10	21.3	261
TN:TP	12	23	31	45	296
DIN:TP	0.16	0.46	1.1	2.9	206

<sup>2</sup> <https://www.eea.europa.eu/data-and-maps/data/clc-2000-raster-4>

### River data

To address seasonal and regional patterns in N:P stoichiometry and evaluate the likelihood of N limitation of autotrophic growth in rivers, we used a dataset on water chemistry from 74 rivers in Norway. All rivers were monitored through a Norwegian monitoring program for “reference rivers” (Thrane et al., 2020; Sandin et al., 2021) and were selected for the program because of the low level of local pressures. The fraction of agricultural land was < 0.5 % in 84 % of the catchments, and below 2 % in all other catchments. These rivers are thus suitable for studying potential effects of nitrogen deposition. The rivers are located in six different ecoregions defined as Southern Norway (South), Western Norway (West), Eastern Norway (East), Central Norway (Central), Outer parts of Northern Norway (North, outer) and Inner parts of Northern Norway (North, inner; Figure 1).

Monthly samples for analysis of water chemistry (12 samples per year) were taken for two years in each river. Half of the rivers were sampled in 2017 and 2019, and the other half in 2018 and 2020. For analyses of water chemistry, we used monthly mean values for the two years. All samples were analyzed for a wide range of chemical parameters at an accredited laboratory using standard methods (see Thrane et al., 2020; Sandin et al., 2021). DIN was calculated as the sum of  $\text{NO}_3^-$ -N and  $\text{NH}_4^+$ -N. Catchment cover (fraction of agriculture, forest, mountains, bogs (wetland) and lakes) and climate parameters (average summer (May-September) and winter (October-April) temperature and precipitation), as well as catchment area, elevation and mean river flow, were estimated for each catchment using the NEVINA database (<https://nevina.nve.no/>) from the Norwegian Water Resource and Energy Department. Data on N deposition was obtained from the Norwegian Institute for Air Research (NILU) as means of total deposition (sum of wet and dry deposition of oxidized and reduced N) for 2012-2016 (Aas et al., 2017)<sup>3</sup>.



**Figure 1.** The location of the 74 rivers and the six ecoregions. Each point marks the location where river water samples were taken.

<sup>3</sup> Calculated using the observational based method and available at a  $0.25^\circ \times 0.125^\circ$  long-lat grid.

### 2.2.1.1 Data analysis

#### Relationships between chl $a$ and N, P and N deposition in natural Fennoscandian lakes

To assess the relationships between chl $a$ , N, P and N deposition, we did four main analytical steps: First, we ran linear regressions of log(chl $a$ ) vs. log(TP), log(TN) and log(DIN) for the whole dataset of natural lakes (n=296). This was done to test the overall correlations between chl $a$  and nutrients. Based on stoichiometry, however, we expect N to be limiting when the ratio of bioavailable N:P is below the N:P ratio that the algae require for growth (Klausmeier et al., 2008). Conversely, P should be limiting when the ratio of bioavailable N:P is above the algal N:P demand. To test this, we ran linear regressions to assess how log(TP) and log(TN) explain variation in log(chl $a$ ) for groups of lakes with different TN:TP and DIN:TP ratios. We divided the dataset into groups of low TN:TP (0-20) and DIN:TP (<1); medium TN:TP (20-50) and DIN:TP (1-2); and high TN:TP (> 50) and DIN:TP (> 2; all ratios by weight). We hypothesized that N will correlate better with chl $a$  in the groups of lakes with low TN:TP and DIN:TP, while P will correlate better in the groups of high TN:TP and DIN:TP. For all linear regressions, we used log-transformed data to homogenize variance and obtain approximately normally distributed residuals. Assumptions of linearity, homoscedasticity and normality of residuals was also tested using diagnostic plots (residuals vs. fitted values, qq-plots etc.). Regressions were judged as significant if  $p$  was < 0.05.

A challenge when analyzing relationships between chl $a$ , TN and TP using monitoring data is that TN and TP are highly collinear (the  $R^2$  for log(TN) vs. log(TP) was 0.67), meaning that we cannot estimate the independent “effects” of the predictors in regression analyses (i.e., some of the variation explained by TN may result from the correlation between TN and TP, and vice versa). To reduce the collinearity between TN and TP, and thereby obtain more independent estimates of the effects of TN and TP, we did a stratified subsampling of the dataset. This was done by dividing the lakes into “low” (below median) and “high” (above median) levels of TN and TP, and assigning each lake into one of the four following groups: “low TP/low TN” (n=121), “low TP, high TN” (n=27), “high TN, high TP” (n=121), and “low TN/high TP” (n=27). In an iterative procedure (1000 iterations), we randomly selected 15 lakes in each group and analyzed log(chl $a$ ) as a function of log(TP) and log(TN) in each subset of 60 lakes. The median  $R^2$  and  $p$ -value for log(TP) and log(TN) was extracted from the 1000 iterations and used as indicators of the relative importance of TP and TN for explaining variation in chl $a$  in the dataset.

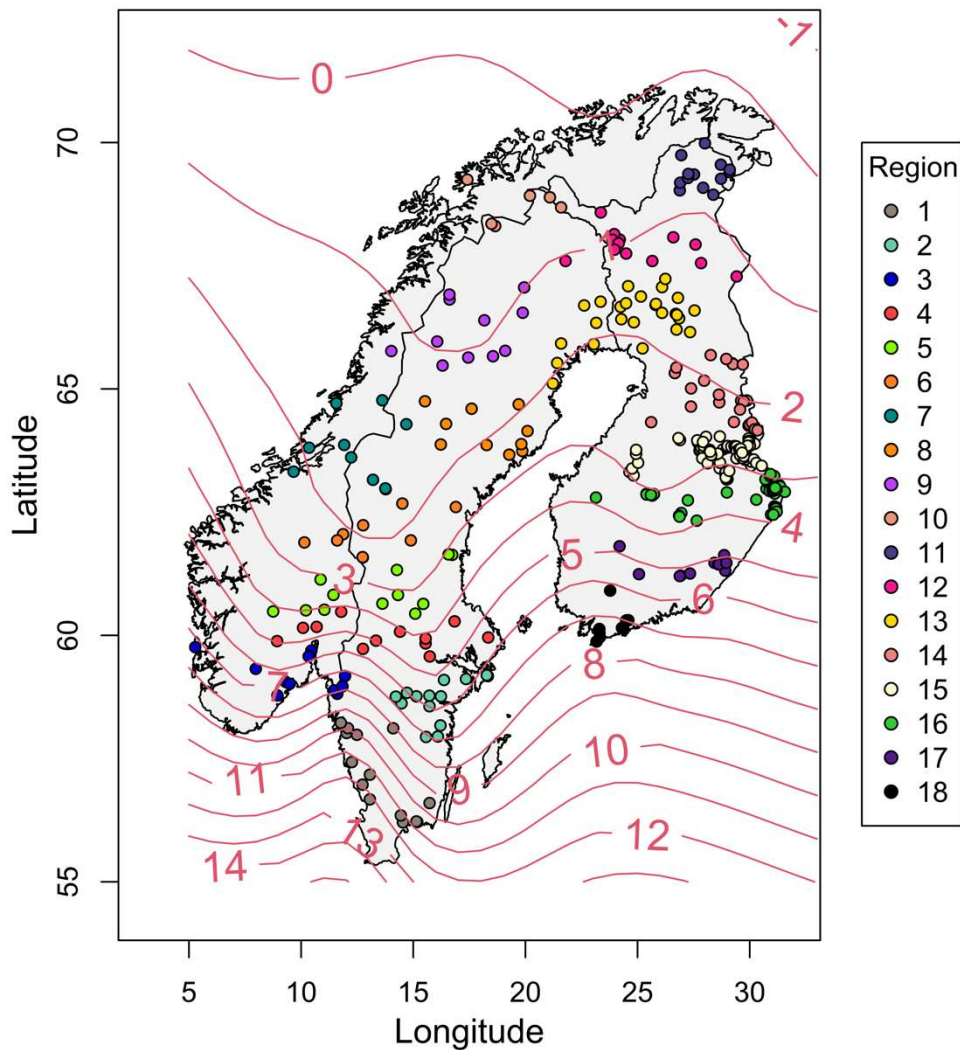
As a fourth step, we wanted to test whether we could detect a positive effect of N deposition on the amount of chl $a$  through increased input of DIN to the lakes. As stated by Bergström (2010), N-limited lakes should respond with an increase in the amount of chl $a$  per unit TP (chl $a$  : TP) upon an increase in N availability. If lakes are P limited, however, no increase in chl $a$  : TP is expected, since chl $a$  and TP will increase in proportion. We followed the approach in Bergström et al. (2005) and distributed the Norwegian, Swedish and Finnish lakes to 18 regions based on levels of N deposition and similarity in climate and geography. For each region, we calculated median values and standard deviations for N deposition, chl $a$  : TP and DIN. We plotted the regional median chl $a$  : TP as a function of regional median N deposition to test whether chl $a$  : TP increased with regional N deposition.

The choice of regions was done as follows: First, using the mgcv-library in R, we fitted a two-dimensional generalized additive model (GAM) of N deposition as a function of latitude and longitude, and plotted the predicted N deposition on a map (Figure 2). A GAM uses splines to fit the response variable as smooth function of the predictor variables and is therefore well suited for modelling non-linear relationships (Wood, 2017). Then, we assigned the lakes to regions of similar N deposition (largely decided by latitude) and geographical position. The geographical borders were generally taken as east/west of the Baltic Sea, and east/west of the watershed line between Norway and Sweden (see

Figure A. 1 and Table A. 1 for an overview of the different regions, number of lakes per region, and summary statistics for environmental variables).

**Analysis of seasonal patterns in N:P stoichiometry and the likelihood of N vs. P limitation in rivers.**

To assess the likelihood of N vs. P limitation of river autotrophs, we analyzed seasonal variation in DIN:TP ratios and DIN concentrations in the 74 rivers. For a given month in the growing season (May-September), N limitation was judged as likely if the DIN:TP was below a theoretical threshold for N vs. P limitation (estimated using bioassays by Bergström (2010)) *and* the concentrations of DIN was < 10 µg/L (Myrstener et al., 2018; Myrstener, 2020). We also evaluated the likelihood and prevalence of N limitation for the different ecoregions. Seasonal trends for DIN:TP in the different ecoregions were estimated by pooling all rivers per ecoregion and then fitting  $\log_{10}(\text{DIN:TP})$  as a function of day of year using a GAM. We also explored relationships between catchment and climate variables, and patterns in nutrient stoichiometry and limitation using principal component analysis (PCA). The following variables were included in the PCA: fraction of forest, mountain, lakes and bogs in the catchment, catchment area (km<sup>2</sup>), median catchment elevation (masl), mean river flow (m<sup>3</sup>/s), summer precipitation (mm), mean summer and winter temperature (°C), and N deposition (kg/ha/year). Due to the different scales of the variables, all variables were scaled to unit variance before analysis. Water chemical variables (median growing season concentrations for DIN, TP, DIN:TP, TN, TN:TP, PO<sub>4</sub>, total organic carbon (TOC) and Ca) were included as passive variables, i.e., they were fitted onto the ordination using the `envfit()`-function in the *vegan* library (Oksanen et al., 2019) in R.

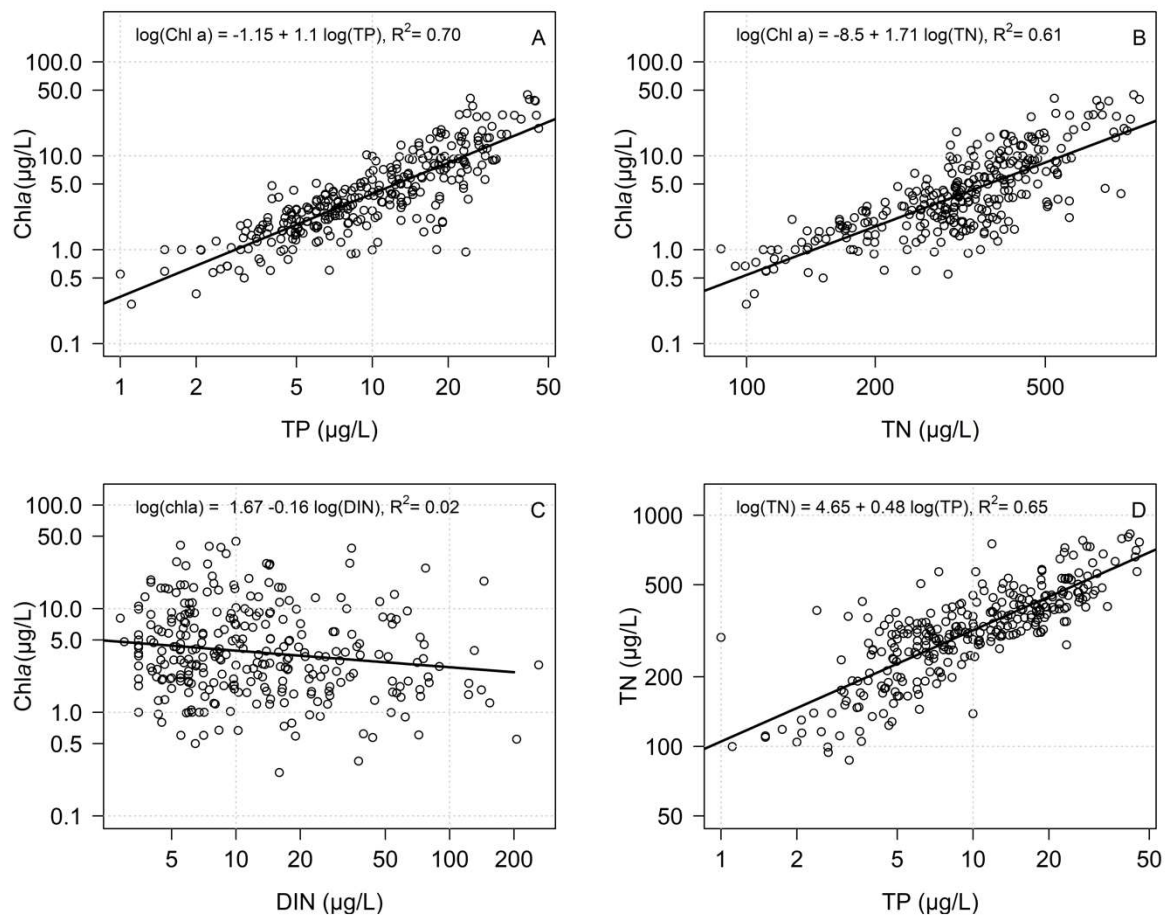


**Figure 2.** Overview of the 296 natural lakes with predicted N deposition ( $\text{kg N ha}^{-1} \text{yr}^{-1}$ ) from a GAM added as isolines. The lakes were distributed to 18 regions marked with different colors (see also Figure A. 1 and Table A. 1).

## 2.3 Results and discussion

### 2.3.1 Relationships between *chl<sub>a</sub>*, nitrogen and phosphorus in the Nordic lakes

Overall, there was a strong relationship between  $\log(\text{chl}_a)$  and  $\log(\text{TP})$  ( $R^2 = 0.70$ ,  $p < 0.0001$ ,  $n = 296$ ; Figure 3A) and a weaker, but still relatively strong, relationship between  $\log(\text{chl}_a)$  and  $\log(\text{TN})$  ( $R^2 = 0.61$ ,  $p < 0.0001$ ,  $n = 296$ ; Figure 3B). There was a significant ( $p = 0.009$ ), though weak, negative relationship between  $\log(\text{chl}_a)$  and  $\log(\text{DIN})$  (Figure 3C), probably reflecting algal uptake of DIN in lakes with high *chl<sub>a</sub>* concentrations. *Chl<sub>a</sub>* is better correlated with TP than TN in the full dataset, indicating that P is the main driver of variation in *chl<sub>a</sub>* across the whole range of Nordic lakes. However, a better correlation with TP than TN may be expected, since a significant fraction of TP in lakes is associated with algal particles (Lewis Jr and Wurtsbaugh, 2008). Hence, the strong overall correlation only indicates a tight coupling between algal biomass and TP, and not necessarily that P is limiting growth or biomass development.



**Figure 3.** The relationship between A) chl $\alpha$  and TP, B) chl $\alpha$  and TN, C) chl $\alpha$  and DIN, and TN and TP in lakes ( $n=296$ ). For A), B) and D),  $p$  is  $< 0.0001$ ; for C),  $p = 0.009$ . Note that axes are log-transformed in all plots.

The correlation between  $\log(\text{TP})$  and  $\log(\text{TN})$  was high ( $R^2 = 0.65$ ,  $p < 0.0001$ ,  $n = 296$ ; Figure 3D). In boreal oligotrophic catchments, N and P are strongly controlled by the concentration of dissolved organic matter, as a large fraction of both elements can be organically bound. We found that both elements were highly correlated with TOC, but with a stronger correlation between  $\log(\text{TN})$  and  $\log(\text{TOC})$  ( $R^2 = 0.71$ ,  $p < 0.0001$ ,  $n = 115$ ) than between  $\log(\text{TP})$  and  $\log(\text{TOC})$  ( $R^2 = 0.52$ ,  $p < 0.0001$ ,  $n=115$ ). This indicates that N was more strongly related to dissolved organic matter than P, which, judging from the high correlation with chl $\alpha$ , most likely has a larger fraction of the total nutrient pool bound in phytoplankton biomass.

The tight coupling between TP and TN makes it difficult to separate the independent “effects” of TP and TN on chl $\alpha$  in linear regressions. To reduce the collinearity between TP and TN, we did a stratified subsampling of the dataset by selecting subsets of lakes where TN and TP were more orthogonal (see methods, chapter 2.2.1.1). Compared to the relationship between  $\log(\text{TN})$  and  $\log(\text{TP})$  in the full dataset ( $R^2 = 0.65$ ), the mean  $R^2$  of the same relationship in the stratified subsampling was  $0.36 (\pm 0.08 = 1 \text{ SD})$ ; Table 2). Even though TN and TP were still correlated, the subsampling managed to reduce the collinearity, which should make the “effects” of TN and TP on chl $\alpha$  more independent in the regressions based on the subsets.



Regressing  $\log(\text{chl}a)$  as a function of both  $\log(\text{TP})$  and  $\log(\text{TN})$  in the subsets revealed a relatively higher  $R^2$  for  $\log(\text{TP})$  than  $\log(\text{TN})$  compared to the regressions on the full dataset. In the subset-analysis, the  $R^2$  for  $\log(\text{TP})$  (0.58) was 40% higher than the  $R^2$  for  $\log(\text{TN})$  (0.41) (Table 2), while the relative difference in  $R^2$  for the full dataset was  $(0.70/0.61) - 1 = 15\%$ . This indicates that some of the variation explained by TN in the full dataset in fact was due to collinearity with TP. Overall, these regressions show that TP is the main driver of variation in  $\text{chl}a$  on a broad geographic scale. The issue of autocorrelation between algal biomass and TP, however, may still be the explanation for the higher fraction of variation explained by TP.

**Table 2.** Summary of results of linear regressions from 1000 iterations of a stratified subsampling selecting  $n=60$  lakes of 4 combinations of high/low TP and high/low TN (see Methods). Mean and SD are the average and standard deviation of the regression  $R^2$  and parameters ( $a$ =intercept;  $b$  = slope) from the 1000 iterations.

	$\log(\text{chl}a) = a + b \log(\text{TP})$			$\log(\text{chl}a) = a + b \log(\text{TN})$			$R^2$ for $\log(\text{TP})$ vs $\log(\text{TN})$
$n=1000$	$R^2$	$a$	$b$	$R^2$	$a$	$b$	
Mean	0.58	-0.98	1.0	0.41	-7.46	1.52	0.36
SD	0.075	0.19	0.09	0.071	0.92	0.16	0.075

Even though TP explained most of the total variation in  $\text{chl}a$ , stoichiometric theory predicts that N should be limiting when the ratio of bioavailable N:P is below the ratio of N:P that the algae need for growth. This could still be true for lakes in the dataset, especially in areas of low N deposition. We tested this hypothesis by running linear regressions of  $\log(\text{chl}a)$  vs.  $\log(\text{TP})$  and  $\log(\text{TN})$  on subsets with low, medium and high N:P ratios (see methods, chapter 2.2.1.1).

The results supported the hypothesis that N is more limiting at low N:P ratios. In both the subsets of lakes with low TN:TP and low DIN:TP,  $\log(\text{TN})$  explained slightly more variation in  $\log(\text{chl}a)$  than  $\log(\text{TP})$  (Table 3). For lakes with medium and high TN:TP and DIN:TP ratios,  $\log(\text{TP})$  was clearly a better predictor for  $\log(\text{chl}a)$  than  $\log(\text{TN})$  (Table 3). The interaction term in a linear regression of  $\log(\text{chl}a)$  vs.  $\log(\text{TN})$  and  $\log(\text{TP})$  was significant ( $p=0.0005$ ), indicating that the effect of  $\log(\text{TN})$  depends on  $\log(\text{TP})$  or vice versa. Collectively, these analyses indicate that N progressively becomes a limiting nutrient as the ambient N:P ratio decreases, as one would expect from stoichiometric theory.

**Table 3.** Results ( $R^2$  and  $p$ -values) of linear regressions of  $\log(\text{chl}a)$  vs.  $\log(\text{TP})$  and  $\log(\text{TN})$  in lakes of low ( $< 20$ ;  $n=44$ ), medium (20-50;  $n=198$ ) and high ( $> 50$ ;  $n = 54$ ) TN:TP ratio, and low ( $< 1$ ;  $n=141$ ), medium (1-2;  $n=51$ ) and high ( $> 2$ ;  $n=101$ ) DIN:TP ratio. Ratios are by weight. All relationships are highly significant ( $p < 0.0001$ ).

	$\log(\text{chl}a)$ vs. $\log(\text{TP})$	$\log(\text{chl}a)$ vs. $\log(\text{TN})$
Low TN:TP	$R^2=0.52$	$R^2=0.59$
Medium TN:TP	$R^2=0.67$	$R^2=0.63$
High TN:TP	$R^2=0.60$	$R^2=0.45$
Low DIN:TP	$R^2=0.48$	$R^2 = 0.54$
Medium DIN:TP	$R^2=0.77$	$R^2=0.65$
High DIN:TP	$R^2=0.64$	$R^2=0.54$

### 2.3.2 N deposition, lake DIN concentrations, and responses of $\text{chl}a$ along the N deposition gradient.

Regional variation in N:P ratios in natural Nordic lakes, and hence the likelihood of phytoplankton N limitation, is mainly driven by regional variation in N deposition and less by regional variation in TP

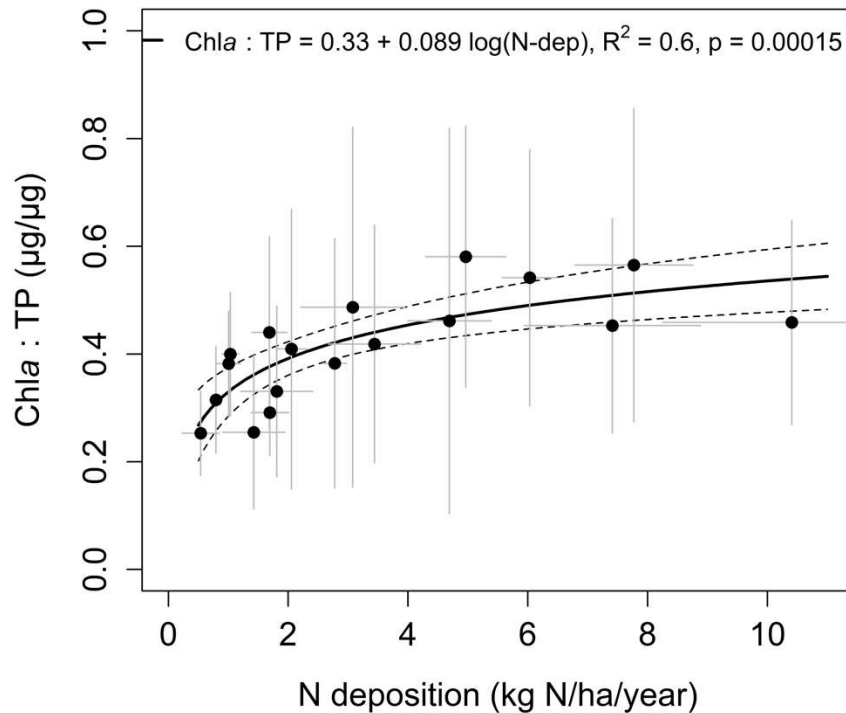
(Bergström et al., 2005; Elser et al., 2009; Isles et al., 2018). This was apparent also in our dataset, where N deposition was strongly related to latitude (and to some degree to longitude). Latitude and longitude explained 95% of the variation in N deposition in a generalized additive model (GAM; Figure 2). TP was less related to latitude and longitude (the fraction of deviance explained in a GAM was 33 %) and more to local conditions, especially TOC ( $R^2$  for the relationship between  $\log(\text{TP})$  and  $\log(\text{TOC})$  was 0.52,  $p < <0001$ ,  $n = 115$ ). TOC is highly correlated to the relative abundance of forests and bogs in the catchment (Larsen et al., 2011).

Notably, however, only 10 % of the variation in epilimnetic summer concentrations of DIN could be explained by N deposition ( $R^2$  for  $\log(\text{DIN})$  vs.  $\log(\text{N deposition})$  was 0.10,  $p < 0.0001$ ). Poor relationships between N deposition and aquatic DIN are often found because of N retention in catchments, which is controlled by climate and land cover (Austnes et al., in preparation). DIN is also taken up by actively growing phytoplankton, although the effect of algal uptake in natural lakes is small compared to the uptake by catchment vegetation. Adding the fraction of catchment forest cover and chl $a$  concentration as predictors of DIN revealed negative effects of both variables, and that the effect of forest cover was most significant (Table 4). Bergström et al. (2005) found a strong relationship ( $R^2 = 0.85$ ) between median N deposition and median DIN in Swedish lakes grouped by region. They used winter values for DIN, when catchment retention and phytoplankton uptake is minimal.

**Table 4.** Results from a linear regression of  $\log(\text{DIN})$  (epilimnetic summer averages) as a function of N deposition ( $\log(\text{N-dep})$ ;  $\text{kg N ha}^{-1} \text{ yr}^{-1}$ ), fraction of catchment forest cover (% forest cover) and lake chl $a$  concentration ( $\log(\text{chl}a)$ ;  $\mu\text{g/L}$ ).  $R^2$  for the full model was 0.29,  $n = 296$ .

	Estimate	SE	p
Intercept	3.12	0.145	<0.0001
$\log(\text{N-dep})$	0.752	0.077	<0.0001
$\log(\text{chl}a)$	-0.139	0.050	0.006
% forest cover	-0.015	0.002	<0.0001

The regional median concentrations of chl $a$  per unit P (chl $a$ :TP) increased significantly with median regional N deposition ( $p < 0.0001$ ,  $R^2 = 0.6$ ; Figure 4). This indicates a positive effect of long range transported N on the phytoplankton biomass of these lakes, similar to findings by Bergström et al. (2005) and Bergström and Jansson (2006) for Swedish and North American lakes. We found that the increase in chl $a$ :TP was most pronounced in the low range of N deposition and levelled off around 4  $\text{kg N ha}^{-1} \text{ yr}^{-1}$ . A curved function (chl $a$ :TP as a function of  $\log(\text{N-dep})$ ) gave a better fit than a linear, and a relationship that levels off is also expected based on stoichiometric theory. As N deposition increases, the levels of DIN rise, leading to higher DIN:TP ratios, and eventually a state of P limitation. When P is the limiting element, chl $a$  increases in proportion to P, leaving the chl $a$ :TP ratio relatively constant.



**Figure 4.** The relationship between median values of chlorophyll *a* per unit total phosphorus (*chl a* : TP) and median values of total N deposition for lakes in 18 regions in Norway, Sweden and Finland. Grey lines show median  $\pm$  one standard deviation for *chl a* : TP (vertical lines) and N deposition (horizontal lines) within each region.

Our analysis suggests that N limitation, and hence a potentially stimulating effect of N deposition on *chl a*, is most likely in regions with N deposition between below 0.5 and ca. 4 kg N ha<sup>-1</sup> yr<sup>-1</sup>, after which the effect levels off (Figure 4). Above 4 kg N/ha/yr, algal productivity is only controlled by TP and is not affected by further increases in N deposition. This is slightly lower than the results of Bergström et al. (2005) from Swedish lakes, where increases in *chl a*:TP levelled off at around 5 kg/ha/yr (based on visual examination of fig. 2 in her paper). In Swedish and North American lakes, increases in *chl a*:TP levelled off at around 6 kg N ha<sup>-1</sup> yr<sup>-1</sup> (Bergström and Jansson, 2006). N deposition has declined since the late 1990s, where most data in Bergström et al. (2005) and Bergström and Jansson (2006) are from, but the ecological effect of N deposition could be related to leaching of N associated with long-term accumulation of N in the catchment, and direct atmospheric inputs. Thus, it is unclear whether the threshold values that we find here (i.e., a levelling off at ca. 4, rather than 5 to 6 kg N ha<sup>-1</sup> yr<sup>-1</sup>) indicate a higher sensitivity to N deposition or a delayed ecological response to reduced N inputs. Note that the exact point where the curve levels off in these studies is based on visual examination of the plots of *chl a*:TP vs. N deposition. The findings in the current study are supported by Elser et al. (2009), who, based on bio-assays and large scale data on N deposition and water chemistry, found that phytoplankton in Norwegian lakes shifted from generally being N-limited at N deposition below 4.5 kg ha<sup>-1</sup> yr<sup>-1</sup>, to being primarily P-limited at N deposition above 8.5 kg ha<sup>-1</sup> yr<sup>-1</sup>.

### 2.3.3 Seasonal pattern in N:P in natural rivers in Norway

The rivers covered wide geographical and climatic gradients, with catchment properties spanning from forested lowlands to high mountain-dominated (Table 5). Due to the low anthropogenic impact, the rivers were generally nutrient poor, with median TP and TN concentrations for the five ecoregions spanning from 2-3 µg P/L and 73-194 µg N/L (Table 5). Most of the rivers were relatively clear, and

median values of TOC for the five ecoregions ranged from 0.9-4.5 mg TOC/L. There were, however, several humic streams (TOC > 5 mg/L) in the southern, eastern and central ecoregions. For more details on water chemical parameters and ecological status of the rivers, see Sandin et al. (2021) and Thrane et al. (2020).

**Table 5.** Median values of stream and catchment characteristics from the six ecoregions in the study. For water chemistry, the numbers represent median values of monthly measurements from January to December. Standard deviation of yearly medians within each ecoregion is shown in brackets. Deposition in kg N ha<sup>-1</sup>yr<sup>-1</sup>, catchment elevation in m.a.s.l., July temperature in °C.

	South	West	East	Central	North, outer	North, inner
Number of rivers	14	10	18	7	14	11
Nitrogen deposition	6.9 [3.2]	6.1 [4.0]	2.9 [2.4]	3.2 [0.9]	1.5 [0.6]	1.3 [0.2]
Catchment elevation	680 [363]	1159 [385]	1039 [395]	512 [304]	425 [211]	285 [156]
% Forest	63 [31]	11 [20]	37 [37]	35 [26]	22 [13]	9 [20]
% Mountain	14 [30]	79 [24]	52 [36]	39 [30]	68 [14]	80 [32]
Mean July temperature	2.8 [2.5]	0.1 [3.0]	-1.5 [2.5]	1.4 [2.4]	1.3 [2.3]	-0.9 [1.4]
TN (µg/L)	194 [100]	103 [57]	135 [131]	109 [53]	73 [20]	73 [46]
DIN (µg/L)	54 [48]	37 [36]	49 [62]	27 [16]	16 [9]	13 [17]
TP (µg/L)	3 [1.4]	2 [1]	3 [4]	3 [1]	2 [0.5]	3 [1.5]
DIN:TP (w/w)	19.3 [11.3]	14.6 [16.8]	12.7 [17.4]	10.5 [5.5]	10 [8.7]	4.5 [5.3]
TN:TP (w/w)	60 [14]	34 [16]	38 [24]	40 [8]	35 [4]	22 [9]
TOC (mg/L)	4.5 [2.0]	0.9 [1.0]	1.5 [4.5]	2.9 [2.2]	1 [0.4]	0.9 [1.4]
Ca (mg/L)	1.2 [0.7]	1.2 [1.2]	2.4 [5.0]	3 [2.9]	3.4 [2.8]	4 [1.9]

There was a clear seasonal pattern in riverine DIN:TP, with lowest DIN:TP during summers in all ecoregions (Figure 5). The low summer DIN:TP ratios were most pronounced in the two northernmost ecoregions (North, outer; Figure 5E and North, inner; Figure 5F), where atmospheric N deposition is low (Table 1). Here, the DIN:TP on average dropped below the critical DIN:TP of ratio 2.2, which is indicative of N limitation (Bergström, 2010). We found the lowest DIN:TP values in ecoregion North, inner (Figure 5F).

In addition to low DIN:TP ratios, most of the rivers in the two northernmost ecoregions had DIN concentration < 10 µg/L during the growing season (Figure 6 and black dots in Figure 5). Myrstener et al. (2018) found consistent N limitation in Northern Swedish rivers, which in summer had a median DIN concentration of 12 µg/L. In another study of Swedish streams and lakes, results from 99 bioassays revealed that N limitation is common when the ambient DIN concentration is < 30-40 µg/L (Myrstener, 2020, chapter 4). Based on these findings, we conclude that N limitation is highly likely if both conditions apply: DIN:TP is < 2.2 (Bergström, 2010) and DIN is ≤ 10 µg/L. Applying these criteria, 17 of 18 rivers in the two northernmost ecoregions were likely N-limited for one or more months during the growing season (Figure 7). According to the same criteria, one third of the same rivers were likely N-limited for four or five of the five growing season months, with most rivers being in ecoregion Northern, inner.

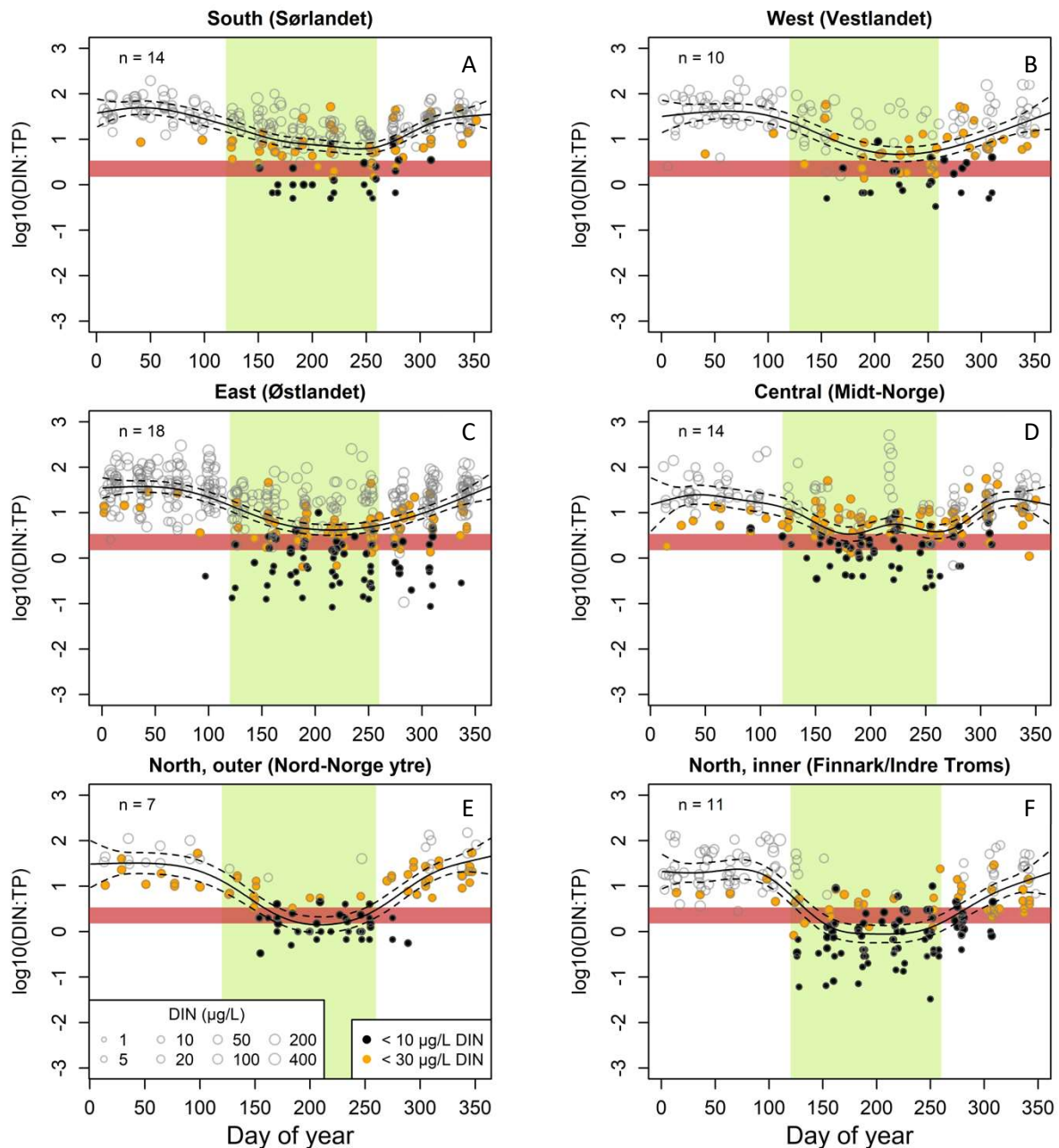
In the Central and East ecoregions, where N deposition is at a medium level (Table 1), about one third of the rivers were likely N limited for at least one month during the growing season, judging from the criteria above (Figure 7). The fraction was slightly higher in Central compared to East Norway. The rivers in the East ecoregion are in a mix of high-mountain catchments with low N deposition, and

lowland catchments with relatively high N deposition. Consequently, drivers of DIN and TP vary considerably between the rivers, resulting in a relatively large variation in DIN:TP in this region (Figure 5C). In Central Norway, most rivers had DIN concentrations below 30 µg/L during the growing season, and concentrations ≤ 10 µg/L was seen in 37-45% of the water samples during late summer (July-September; Figure 6G). Concentrations of DIN were higher in East Norway, and growing season DIN concentrations were rarely ≤ 10 µg/L (Figure 6E). DIN was, however, < 30 µg/L in more than half of the samples from each growing season month.

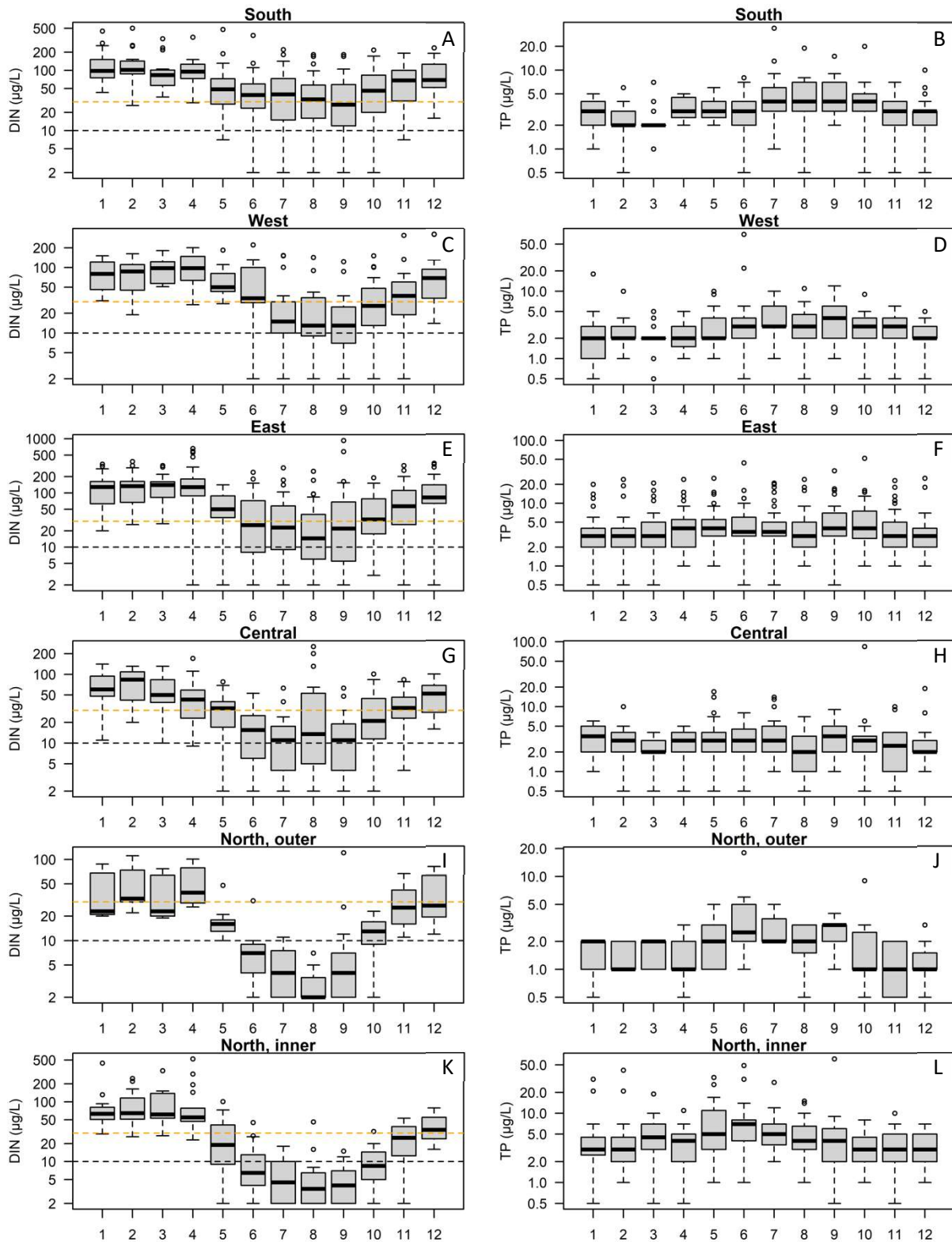
The South and West ecoregions of Norway receive relatively high N deposition (Table 1). Most rivers in these regions had relatively high DIN:TP and high DIN concentrations also during the summer (Figure 5A and B, and Figure 6A and C). Hence, only a few rivers were apparently N limited, and P limitation is likely most common in these regions. In total, only 5 of 24 rivers were likely N limited for one or more months during the growing season in these regions (Figure 7). Highest DIN:TP and highest concentrations of DIN were found in ecoregion South. Here, median DIN was > 30 µg/L in all growing season months, except for September, where DIN was close to 30 µg/L. In ecoregion West, most DIN concentrations were below 30 µg/L but rarely below 10 µg/L (Figure 6).

The temporal and regional patterns in N:P stoichiometry and DIN concentrations (Figure 5 and Figure 6) indicate a high likelihood and prevalence of N limitation in northern Norway, both N and P limitation in central- and eastern Norway, and mostly P limitation in southern and western Norway. Accurate prediction of nutrient limitation solely based on water chemistry, however, is difficult (Keck and Lepori, 2012). Our analysis can therefore only give an indication of the likelihood of N limitation. A true test of nutrient limitation would need *in situ* investigations, for instance through bioassays experimentally adding N and P in a factorial design (see e.g., Myrstener et al. 2018). Still, the threshold DIN:TP from Bergström (2010) is based on bioassays and water chemistry from catchments in broadly the same geographical region, suggesting it is a good indicator of N vs. P limitation in these rivers. Moreover, DIN:TP is a better indicator than TN:TP for assessing N vs. P limitation in northern freshwaters, as a large fraction of the TN in natural catchments often is organically bound and not easily available for uptake by autotrophs, especially in humic surface waters. Even though the critical DIN:TP of Bergström (2010) is based on phytoplankton responses, it should apply also to streams, because the N:P demand of benthic algae or periphyton is not inherently different from that of freshwater phytoplankton (Hillebrand and Sommer, 1999).

Note that the concentrations of phosphate were low during the growing season, with median dissolved phosphate concentration in May-September being below the detection limit of 1 µg P/L in all ecoregions (Table 6). With such low levels of phosphate, we cannot exclude P limitation or co-limitation by N and P even in the rivers with DIN:TP below the critical ratio of Bergström (2010) and DIN concentrations below 10 µg/L. In such ultraoligotrophic rivers with low values of both N and P, co-limitation is likely common, at least during certain periods. Myrstener et al. (2018), however, found consistent N limitation in bioassays from rivers in Northern Swedish streams even though PO<sub>4</sub> concentrations were often low (below 1 µg/L) and theoretically in deficit relative to N. Hence, although dissolved phosphate is low, DIN can be supplied in even lower quantities relative to biological demand. N limitation at PO<sub>4</sub> concentrations below 1 µg/L in Myrstener et al. (2018) could indicate that fractions of the total P pool likely also were bioavailable, so that P limitation was avoided even though the concentrations of dissolved inorganic P was low.



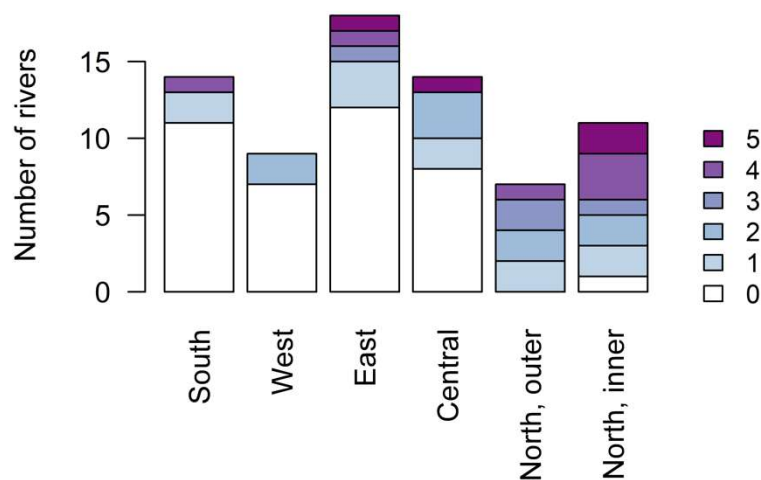
**Figure 5.**  $\log_{10}(\text{DIN:TP})$  plotted as a function of day of year for 74 rivers in six different eco-regions in Norway. The green band show the “growing season”, here defined as May to September. The red band shows the range ( $\text{DIN:TP} = 1.5\text{-}3.4$ ; at  $\text{DIN:TP} = 2.2$ , there was a 50 % chance of N limitation) where phytoplankton shifted from P to N limitation according to Bergström (2010). The dots are scaled to DIN concentration. Samples with  $\text{DIN} < 30 \mu\text{g/L}$  are shown as filled orange circles and  $\text{DIN} < 10 \mu\text{g/L}$  as filled black circles. The trend lines (GAMs) show the average  $\text{DIN:TP}$  over the year for each eco-region. Note: data from both sampling years (2017/2019 or 2018/2020) are shown in the figure.



**Figure 6.** Boxplots of DIN ( $\mu\text{g/L}$ ) and TP ( $\mu\text{g/L}$ ) (log-scale on y-axis) per month (1-12, x-axis) and ecoregion. Data from two years (2017/2019 and 2018/2020) are included. In the plots of DIN, the orange dotted line marks a concentration of  $30 \mu\text{g/L}$  while the black line marks  $10 \mu\text{g/L}$ . Note: data from both sampling years (2017/2019 or 2018/2020) are shown in the figure.

**Table 6.** First quantile (Q1), median, and third quantile (Q3) concentrations of dissolved phosphate ( $\mu\text{g P/L}$ ) in the growing season (May-September) per ecoregion. Values for total phosphate ( $\mu\text{g P/L}$ ; dissolved + particle bound) are shown in brackets.

Ecoregion	Q1	Median	Q3
South	< 1 (< 1)	< 1 (< 1)	1 (1.25)
West	< 1 (< 1)	< 1 (1)	1 (2)
East	< 1 (< 1)	< 1 (1)	1 (2)
Central	< 1 (< 1)	< 1 (< 1)	1 (1)
North, outer	< 1 (< 1)	< 1 (< 1)	1 (1)
North, inner	< 1 (< 1)	< 1 (2)	2 (3)



**Figure 7.** Prevalence of N limitation or co-limitation by N and P in the growing season (May-September) based on DIN:TP and DIN concentration. The y-axis shows the number of rivers per ecoregion, and the color scale indicates the number of months with DIN:TP < 2.2 and DIN < 10  $\mu\text{g/L}$ .

### 2.3.3.1 Drivers of seasonal variation in DIN:TP

The seasonal pattern in DIN:TP in the rivers was mainly driven by variation in DIN (Figure 6, left panels), which showed a similar seasonal pattern as DIN:TP (Figure 5). Low DIN and DIN:TP during summer was likely caused by retention of DIN by vegetation and soils in the catchment during the growing season (Kaste et al., 2020). Seasonal variation of DIN is often strongly correlated with air temperature (Vuorenmaa et al., 2018). Due to the relatively warm climate and longer growing seasons, the DIN:TP ratio started its seasonal decline already in April in the rivers located in the ecoregions South and West (Figure 5). In Central Norway, which has several lowland coastal rivers, DIN also started to decline in April. In the northernmost ecoregions (especially in North, inner), DIN:TP did not start to decline before May as a consequence of the colder climate and later onset of vegetation growth. In the East ecoregion, > 50% of the rivers were draining high mountain catchments (> 1000 masl) and areas with relatively cold climate, similar to the North regions. Hence, the seasonal decline in DIN:TP also started in May.

TP was relatively stable throughout the year (Figure 6, right panels) and therefore contributed less to the seasonal variation in DIN:TP. Slightly higher values of TP were seen during early summer (May and June) in the two northernmost ecoregions, probably related to the spring snow melt (Figure 6J and L). In South and West, TP tended to be slightly higher during late summer/fall (Figure 6B and D), perhaps related to more precipitation and increased runoff of DOM and particle-bound P during the fall. In the



East and Central ecoregions, TP was more stable, but with slightly higher concentrations in spring (April/May) and fall (October/November) in the east. Overall, the concentration of TP was linked to the high-flow events during spring and/or autumn. We found a significant ( $p < 0.01$ ) correlation (Pearson's  $r$ ) between  $\log(\text{TP})$  and  $\log(\text{TOC})$  in all ecoregions (North, outer:  $r = 0.38$ ; North, inner:  $r = 0.61$ ; Central:  $r = 0.59$ ; East:  $r = 0.50$ ; West:  $r = 0.36$ ; South:  $r = 0.60$ ). Moreover,  $\log(\text{TP})$  was significantly ( $p < 0.01$ ) correlated with  $\log(\text{turbidity})$  (North, outer:  $r = 0.24$ ,  $p = 0.01$ ; North, inner:  $r = 0.59$ ; Central:  $r = 0.45$ ; East:  $r = 0.50$ ; West:  $r = 0.61$ ; South:  $r = 0.54$ ), showing that TP is linked to DOM and particles that are released from the catchments during snowmelt and/or precipitation events. Hydrology is an important control of DOM (Boyer et al., 1996; de Wit et al., 2007).

DIN concentration varied with a factor of 13.5-18 within a year in most rivers in the three northernmost ecoregions (North outer, North inner, and Central Norway; Table 7). I.e., DIN concentration was 13.5-18 times lower during the summer than during the winter in these regions. In the ecoregions West, South and East, DIN in most rivers varied by a factor of 7-9 (Table 7). In comparison, TP varied only by a factor of 5-7 over the season in North inner and outer, and the Central ecoregions, and 3-4 in ecoregions South, West and East (Table 7). Hence, the relative seasonal variation in DIN in each river was two to three times higher than the variation in TP for all ecoregions. This indicates that DIN drove the variation in DIN:TP.

**Table 7.** Median relative variation in DIN and TP over the year in the rivers in the different ecoregions. The numbers were calculated by dividing the highest monthly DIN concentration by the lowest monthly DIN concentration for each river and taking the median per ecoregion. Standard deviation for the relative change is given in brackets.  $n$  = the number of rivers per ecoregion

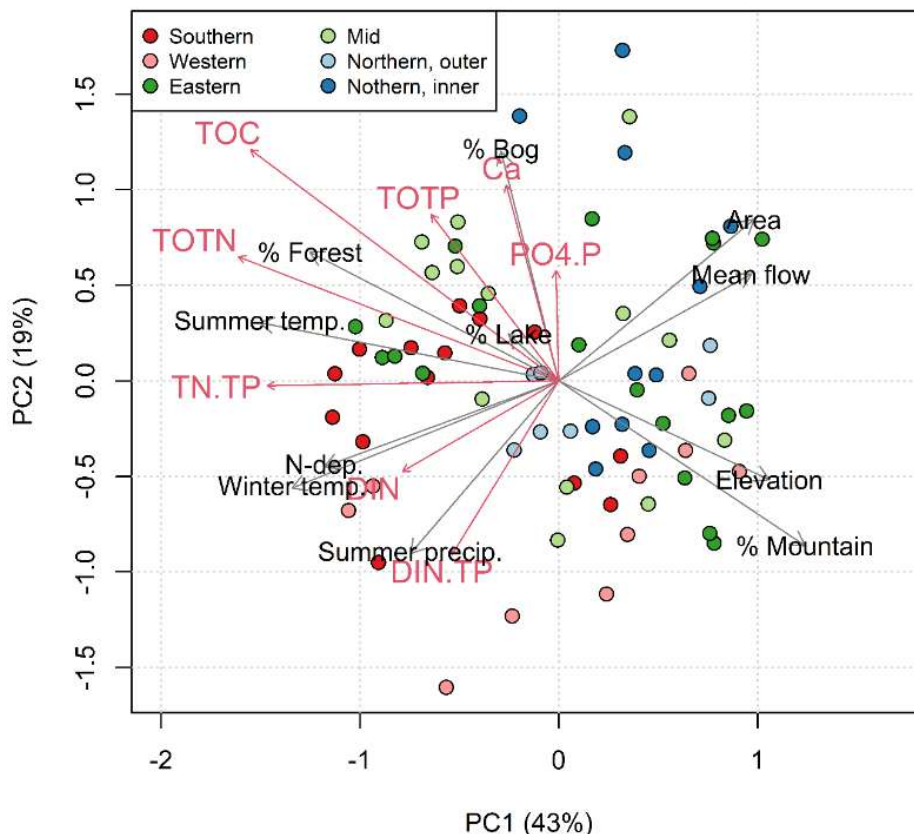
Ecoregion	Relative yearly variation in DIN (median [sd])	Relative yearly variation in TP (median [sd])	n
South	6.5 [2.5]	2.9 [1.5]	14
West	8.9 [2.8]	3.8 [2.3]	10
East	9.0 [3.2]	4.2 [1.9]	18
Central	13.5 [2.6]	5.0 [2.1]	14
Northern, outer	15.8 [1.2]	4.9 [1.6]	7
Northern, inner	18.3 [2.4]	6.8 [1.8]	11

### 2.3.4 Factors affecting the likelihood of N limitation

Because of the limited local anthropogenic pressures in the catchments, the DIN:TP ratio (and thus the likelihood of N limitation) in the rivers is likely to be primarily influenced by atmospheric nitrogen deposition and factors affecting retention of N in the catchments. Additionally, the DIN:TP is influenced by TP from the catchment. However, TP varies much less than DIN (see section 2.3.3.1). A PCA on catchment and climate variables revealed that the main underlying gradient (PC1; explaining 43% of the variation in the data) spanned from relatively warm, forested catchments with high N deposition (to the left in Figure 8) to relatively cold catchments with low forest cover and N deposition (to the right in Figure 8), which are both high-latitude and high-elevation catchments. PC axis 2 only explained 19 % of the variation in the data and was most strongly related to the fraction of bogs (wetlands) in the catchment.

High concentrations of DIN and high DIN:TP were associated with high N deposition and high temperature catchments with relatively high precipitation. These conditions are typical for the southern and western ecoregions in Norway. Using a simple linear regression model with  $\log(\text{median}$

summer DIN:TP) as response, we found a highly significant positive effect of N deposition ( $p < 10^{-5}$ ;  $R^2 = 0.29$ ,  $n = 74$ ) and a slight negative effect of summer temperature ( $p = 0.03$ ). Summer temperature was highly correlated with fraction of forest in the catchment, and including fraction of forest instead of temperature resulted in a negative, though non-significant ( $p = 0.07$ ) effect. The negative “effect” of temperature or forest cover could be related to increased retention of DIN by forest. However, it could also be related to increased TP (and with this lower DIN:TP) associated with DOM from forest in the catchment. Overall, however, N deposition was clearly the best predictor for both DIN and DIN:TP. Rivers with low DIN:TP and low DIN were typically found in relatively cold areas with low N deposition – typical for the northern parts of Norway.



**Figure 8.** Ordination diagram (PCA) showing the main gradients in catchment characteristics (% mountain, forest, lake and bog in the catchment; catchment area (km<sup>2</sup>); median catchment altitude (elevation, masl); mean water flow (m<sup>3</sup>/s)), climate (average summer and winter temperature (°C)), and N deposition (kg/ha/year) for the 74 rivers. The water chemical variables (plotted in red) were fitted as passive variables (see methods). The rivers are colored according to ecoregion.

## 2.4 Summary and conclusions

We addressed five main questions relating to the importance of N limitation for phytoplankton in lakes and the potential for seasonal N limitation in rivers. The results of analysis of the natural lake dataset from Norway, Sweden and Finland indicated that the amount of chl<sub>a</sub> correlated better with TP than with TN, but that TN explained more of the variation in chl<sub>a</sub> than TP at low TN:TP and DIN:TP ratios. This implies that N limitation becomes progressively more important as the availability of N relative to

P decreases, as one would expect from stoichiometric theory. Using median values from 18 geographical regions, we found that the amount of chl $a$  per unit of TP increased with N deposition up to deposition levels of circa 4 kg N ha $^{-1}$  yr $^{-1}$  and then levelled off. Even though the chl $a$ :TP ratio was highly variable within regions, the pattern of increasing chl $a$ :TP with N deposition illustrates that N limitation is common in lakes in areas with deposition levels below 2-4 kg N ha $^{-1}$  yr $^{-1}$ . Moreover, it shows that N deposition has caused an increase in the amount of algal biomass in N-limited, low N deposition areas. The increase of chl $a$ :TP with N deposition is consistent with other studies from oligotrophic lakes in low deposition areas (Bergström et al., 2005; Bergström and Jansson, 2006; Elser et al., 2009), although these studies found a levelling off at slightly higher N deposition rates (5-6 kg N ha $^{-1}$  yr $^{-1}$ ).

When it comes to seasonal and regional patterns in N:P stoichiometry in rivers, DIN:TP ratios and DIN concentrations indicated that N limitation is common in northern Norway. Most of the rivers in the two northern ecoregions (North, outer and North, inner; N deposition 1.3 to 1.5 kg N ha $^{-1}$  yr $^{-1}$ ) were N-limited for at least one month during the growing season from May to September. About one third of the rivers in the two northern ecoregions were N-limited in  $\geq$  four of the growing season months. The rivers in the ecoregions Central and East Norway (N deposition 2.9 to 3.2 kg N ha $^{-1}$  yr $^{-1}$ ) were a mix of both N- and P-limited, while the rivers in ecoregion South and West (N deposition > 6.0 kg N ha $^{-1}$  yr $^{-1}$ ) were generally P-limited. Low values of DIN:TP were primarily driven by retention of DIN by vegetation and soils in catchments, which resulted in DIN concentrations well below 10  $\mu$ g/L from June to September in most rivers furthest north and DIN concentrations above/around 30  $\mu$ g/L further south, where N deposition was higher. However, phosphate levels were generally also low and < 1  $\mu$ g/L on average in all ecoregions, highlighting the possibility of co-limitation by N and P.

The findings in this chapter support that natural surface waters can be N limited and that N deposition can stimulate phytoplankton growth. This information along with other recent findings in the literature serve as basis for the proposed empirical critical loads in chapter 3.

## 3 Literature review on nutrient limitation in freshwaters

### 3.1 Introduction

de Wit and Lindholm (2010) reviewed literature on eutrophying effects of N availability and N deposition on oligotrophic freshwater ecosystems. The current chapter is an update of the 2010-review with a similar structure, reviewing literature that was published from 2010 and onwards and serves as background for the contribution to the ongoing revision of empirical critical loads for nitrogen (Bobbink et al., in preparation). de Wit and Lindholm (2010) discussed the widely supported hypothesis of P limitation in freshwaters which was challenged especially in the 2000s. The literature review summarized peer-reviewed studies that demonstrated that increased availability of N, related to atmospheric N deposition in nutrient-poor temperate, boreal and arctic lakes affect freshwater biology. The literature was grouped after type of evidence, i.e. on algal productivity from paleolimnological studies, whole-lake experiments, regional lake surveys and experimental nutrient additions (mesocosm studies and bioassays).

Since 2010, evidence for N limitation in oligotrophic freshwaters has become substantiated. Howarth et al. (2021) present a review of evidence for N and P limitation in aquatic ecosystems. For lakes in the US, they conclude that the N:P ratio on average is above the limit needed by phytoplankton, suggesting that P limitation is most common. However, it was underlined that this especially concerns lakes where the nutrient sources are in the catchment rather than from deposition. In lakes in low N deposition areas without local catchment N and P sources, they concluded that N limitation or co-limitation is common and can be driven towards P limitation when N deposition is high and/or catchment N retention is low. Additionally, evidence of cyanobacterial blooms in oligotrophic lakes is accumulating and the paradigm of the necessity of high-nutrient conditions for cyanobacterial blooms is challenged (Reinl et al., 2021). However, factors that trigger such blooms in oligotrophic lakes remain poorly understood and the role of nitrogen availability is unclear.

In recent years, focus has been drawn to the role of dissolved organic matter (DOM) as a mediating factor for freshwater productivity, i.e. by limiting light availability and by transporting organic forms of N and P (Karlsson et al., 2009; Seekell et al., 2015). Northern lakes have become more DOM-rich during the past decades, related to reduced acid deposition (Monteith et al., 2007) and to increased precipitation (de Wit et al., 2016). DOM in boreal and alpine lakes varies with land cover (forests and peatlands) and climate (Larsen et al., 2011). Results from a whole lake experiment (Deininger et al., 2017a) suggested that effects of enhanced inorganic N are weakened along a positive DOM gradient, which could imply that algal productivity in clear-water lakes is more sensitive to increased N availability than darker lakes.

Establishing links between changes in atmospheric inputs and changes in N species in surface waters remains challenging (Amos et al., 2018), in part because of confounding effects of climate and climate change impacting aquatic processing of reactive N (Baron et al., 2013). Integrated monitoring has helped to determine sources and has shown that reduction of N in inland waters, related to reduced N deposition, has also helped to reduce eutrophication in coastal waters (Amos et al., 2018). Surface waters in catchments that receive high N deposition have a higher risk for elevated inorganic nitrogen concentrations (Wright et al., 2001). The risk for higher nitrate concentrations is mediated by catchment properties such as vegetation cover or lack thereof, as demonstrated in a dataset of water chemistry, catchment characteristics, climate and deposition from North America and Europe (Austnes

et al., in preparation). The possibly greater sensitivity of productivity to increased N availability in low-DOM lakes in combination with a relatively low N-retention capacity of their catchments implies that clear-water lakes are especially sensitive to N deposition. Low DOM and low nitrogen retention capacity are causally linked through low pools of soil organic matter in their catchments, which is a consequence of sparsely vegetated areas and shallow soils (Evans et al., 2006).

### 3.2 Paleolimnological evidence

Recent studies are referenced in a review of paleolimnological studies for assessing eutrophication and its impact on lakes (Davidson and Jeppesen, 2013). These authors conclude that N in addition to P impacts eutrophication, especially in remote lakes with no local catchment sources of N and P. However, it is difficult to derive levels of N deposition from paleolimnological studies from remote lakes (Catalan et al., 2013) because N is not conserved in lake sediments. Catalan et al. (2013) report that in lakes receiving elevated N deposition that did not warm or acidify, evidence for changes in diatom assemblages is limited. Laird et al. (2017) tested if N deposition from oil sand exploitation in the Athabasca Oil Sands Region (AOSR) in Canada had an impact on diatom assemblages in lake sediments. Cores were collected from N- and P-limited lakes downwind of the AOSR emissions, and no effect was found. Thus, recent paleolimnological studies continue to support that N is a limiting nutrient in remote lakes in agreement with previously reviewed studies de Wit and Lindholm (2010), but do not offer significant new contributions suitable for determining threshold values for empirical critical loads of N deposition.

### 3.3 Whole-lake experiments

de Wit and Lindholm (2010) summarized evidence from whole-lake experiments that test nutrient controls on algal productivity. Some of these experiments were interpreted as being supportive of nitrogen- or co-limitation. One recent whole-lake experiment, designed with one pre- and two post-manipulation years, was reported by Deiningner et al. (2017b). Here, nitrogen was added to three oligotrophic lakes in Sweden in a paired, whole-lake design with three control lakes along a dissolved organic carbon (DOC) gradient in a low N deposition area ( $< 2 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ) Deiningner et al. (2017b). The observed responses included increases in phytoplankton production, biomass (supportive of N as a limiting nutrient) and the ratio of primary versus secondary productivity. Zooplankton biomass (the next trophic level) did not change, which suggests that the increase in primary productivity did not cascade to the next trophic level. Whole-lake experiments are powerful for testing hypotheses on nutrient limitations in lakes, and the new study demonstrates N limitation in nutrient-poor lakes at N-deposition levels below  $2 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , in agreement with existing empirical critical loads.

### 3.4 Gradient studies

Gradient studies are potentially valuable for establishing critical thresholds because it is possible to relate ecological responses to measured levels of a particular stressor, such as nitrogen deposition. However, interpretation of gradient studies can be hampered by the presence of confounding factors, such as other environmental gradients. However, some gradient studies aimed to test nutrient limitation were supported with results from bioassays designed to test short-term responses to additions of N and/or P. Here, we present new studies focusing on algal productivity and on macrophytes.

With regard to phytoplankton, some new gradient studies and synoptic surveys from Europe and North America focusing on effects of N availability in remote lakes were found with varying support for N limitation. A regional survey of 29 remote alpine ponds and lakes in Canadian Rocky Mountains,

sampling water and doing bioassays of phytoplankton to evaluate the impact of N deposition, showed that shallow ponds were more likely to be N-limited than lakes (Murphy et al., 2010). However, only 14% of the investigated sites demonstrated N limitation. A lake in the Spanish Pyrenees showed signs of P limitation induced by long-term enrichment of reactive N from deposition. In the lake, a decline in lake DIN was found despite increase in inlet DIN and N deposition for the period 1998-2011 (Camarero, 2017). However, the lake also increased in TP concentrations related to inputs of atmospheric P from Saharan dust. Also, algal biomass increased, suggesting that the observed changes were related to changes in P, and thus indicated P limitation.

Atmospheric N deposition, lake water quality, and sediment diatoms were measured in ultra-oligotrophic lakes in national parks in Washington State (USA) (Sheibley et al., 2014). A shift in diatom community was found, indicative of N-enrichment and supporting a critical load of 1-1.2 kg N ha<sup>-1</sup> yr<sup>-1</sup> of wet deposition for these lakes. Nelson and Carlson (2011) presented in-situ short-term (<2 weeks) microcosm experiments in a high-elevation (alpine) lake (Emerald Lake, Sierra Nevada, California, USA) to evaluate responses in bacterioplankton growth, carbon utilization, and community structure to short-term enrichment by nitrate and phosphate. In both experiments, phosphate enrichment (but not nitrate) caused significant increases in bacterioplankton growth, changed particulate organic stoichiometry, and induced shifts in bacterial community composition. No support was found for N limitation. Long term average levels of N deposition were around 2 N ha<sup>-1</sup> yr<sup>-1</sup>.

A meta-analysis of field studies and experimental data from a global lake dataset focused on effects of nutrients and DOM on relative and absolute rates of primary production of phytoplankton and secondary bacterial production (Faithfull et al., 2011). Experiments and field surveys were not consistent in their responses. The study concluded that in field surveys, ratios of secondary to primary production were highest at low N availability and at high latitudes, as a consequence of lower primary productivity. Thus, the field surveys – including N-gradients - supported that N is a limiting nutrient in oligotrophic lakes while results from experimental studies could not be interpreted in the same way.

Some of the key studies used for establishing empirical critical loads for nitrogen in freshwaters in 2011 only used phytoplankton data (chlorophyll *a*) and water chemistry (Bergström et al., 2005; Bergström and Jansson, 2006). The studies were based on data collection from the 1990s and early 2000s. Deposition of N in Europe has declined since 1990 (Torseth et al., 2012; Engardt et al., 2017) while concentrations of DIN have either remained constant or have declined, probably driven by trends in deposition and climate (Rogora et al., 2012; Lucas et al., 2016; Kaste et al., 2020).

The current report (Chapter 1) presents an analysis of algal productivity in relation to water chemistry and N deposition in Nordic lakes from undisturbed catchments for the years 2006 to 2017. Concentrations of chlorophyll *a* (*chl<sub>a</sub>*) are used as a proxy for algal biomass. The analysis documents a positive relationship between the ratio of *chl<sub>a</sub>* to the concentration of total phosphorous, and N deposition, for lakes grouped into 18 regions in the Nordic countries. More algal biomass per unit P is found with increasing N deposition, at low levels of deposition (i.e., between 0.5 and 4 kg N ha<sup>-1</sup> yr<sup>-1</sup>), indicative of N limitation. At higher levels of N deposition (>4 kg N ha<sup>-1</sup> yr<sup>-1</sup>), the relationship between *chl<sub>a</sub>* and P remains constant. Thus, the threshold range where the lakes switch from N limitation to P or co-limitation is close to, or slightly lower, than the range found in the aforementioned studies (Bergström et al., 2005; Bergström and Jansson, 2006), i. e. 5 to 6 kg N ha<sup>-1</sup> yr<sup>-1</sup>. It should be noted that our report uses the sum of dry and wet deposition (total deposition), while many other studies, including Bergström and Jansson (2006), use wet N deposition only. In our study, we calculated that median wet N deposition in their dataset is 68% (25% and 75%percentiles: 63% and 73%, respectively) of total N deposition (EMEP data, for 2006-2018) in the Nordic countries.

de Wit and Lindholm (2010) summarized nutrient enrichment effects on macrophytes and concluded that the decline in water plant communities could be related to increases in nutrient availability, but that links between community shifts, increased N availability and deposition were

difficult to establish because nutrient responses were confounded by other factors like liming, sediment processes, climate and interspecific competition. This conclusion is supported by two post-2010 studies. Moe et al. (2013) present a field survey on the invasive species *Juncus bulbosus* in 139 lakes in southern Norway with the aim to explain distribution patterns. Deposition of N was suspected as a driving factor but it was concluded that N deposition was not important. Curtis et al. (2014) explored where water chemical limits could be used to protect sensitive macrophyte communities. However, identifying potential impacts of nutrient N in naturally nutrient-poor systems of conservation importance and links to biodiversity was severely inhibited by lack of reference data from unimpacted reference water bodies. The current empirical critical loads to protect macrophyte communities of shallow softwater lakes in Europe (5-10 kg ha<sup>-1</sup> N yr<sup>-1</sup>) are exceeded in the UK, but the empirical evidence for the threshold values is limited, which limits the application of empirical critical loads for nitrogen and thus also the protection of sensitive ecosystems. In summary, it remains difficult to develop critical threshold values for responses of macrophytes to nitrogen.

### 3.5 Mesocosm experiments and in-situ bio-assays

de Wit and Lindholm (2010) summarized experimental evidence on nutrient limitation from mesocosm experiments. Trommer et al. (2017) presented results on foodwebs from mesocosm experiments in three temperate lakes in nature reserves in Germany with high N:P ratios (DIN:P >50), to mimic effects of N deposition. However, background concentrations of NO<sub>3</sub> varied between 1.3 to 17 mg/L in these lakes which far exceeds concentrations in remote lakes in Europe and North America (Austnes et al., in preparation). The additions of N multiplied background N deposition (ca. 50 kg N ha<sup>-1</sup> yr<sup>-1</sup>; estimated from information in Trommer et al. (2017)) with a factor 1, 2, 8, 16, and 32). This background deposition level exceeds current empirical critical loads for nitrogen for any type of lakes (Bobbink and Hettelingh, 2011). Reported effects included removal of N by phytoplankton, despite the initially high N concentrations, and increases in chl<sub>a</sub> in two out of three lakes. Mesozooplankton abundance (the lower foodweb) decreased, which was interpreted as an N-enrichment effect. Given the large background N concentrations, it is noteworthy that additional N still can result in an increase in algal biomass, which suggests co-limitation even in relatively N-rich lakes.

Brittain and Strecker (2018) present a multi-stressor mesocosm test of zooplankton community responses to N addition in mesocosms in two lakes, with and without fish in Mount Rainer National Park (USA). The responses were surprising and counter-intuitive and demonstrate that such ecological interactions are difficult to predict. Thus, no new evidence relevant to empirical critical loads has emerged from mesocosm studies in the literature after 2010.

de Wit and Lindholm (2010) summarized responses of benthic algae to increases in N supply primarily based in-situ experiments manipulating nutrient supply. New studies demonstrate some support for N limitation. In-situ nutrient-diffusion strata placed in alpine lakes in the French Alps with and without access to grazing showed that phyto-benthic biomass was higher in N-enriched substrates, and that green filamentous algae were favoured over diatoms and cyanobacteria (Lepori and Robin, 2014). Myrstener et al. (2018) present nutrient-diffusion strata in arctic streams in northern Sweden to test for various limitations on microbial communities in biofilms. They found a strong N limitation where N was added. Temperature regulated productivity in tundra streams and light regulated productivity in birch and coniferous streams.

These new studies, combined with earlier evidence, suggest that benthic algal productivity is driven by many factors including N availability. However, threshold values for N deposition are difficult to derive for benthic algae because of the complexity of interactions between nutrient availability and other factors.

### 3.6 Empirical critical loads for surface waters

In the US, empirical critical loads for surface waters were developed (Baron et al., 2011) shortly after the European empirical critical loads were published (Bobbink et al. 2011). Baron et al. (2011) included the capacity of the catchment to retain atmospheric nitrogen as a factor for the critical loads. Here, the nutrient enrichment critical load for western lakes was given a range of 1.0 to 3.0 kg N ha<sup>-1</sup> yr<sup>-1</sup>, reflecting areas covered with little or no vegetation in complex, snowmelt-dominated catchments. This type of ecosystem would be similar to European alpine lakes. The nutrient enrichment critical load for northeastern lakes was given a range of 3.5 to 6.0 kg N ha<sup>-1</sup> yr<sup>-1</sup>. The Northeastern US is forested and seasonally snow-covered and could be compared with boreal and temperate Europe.

In the 2011 report (Bobbink et al. 2011), a single critical load range for EUNIS Class C1.1 (permanent oligotrophic water bodies) of 3 to 10 kg ha<sup>-1</sup> yr<sup>-1</sup> (reliable) was proposed. The range should only be applied to oligotrophic waters with low alkalinity with no significant agricultural or other direct human inputs. Furthermore, the lower end of the range would apply to boreal, sub-Arctic and alpine lakes, whereas the upper end of the range would apply to Atlantic soft-water lakes. It was also concluded in 2011 that dystrophic lakes (C1.4) may also be sensitive to N deposition. The latter is now further substantiated, and it can be stated with more confidence. However, results from a whole lake experiment (Deininger et al., 2017b) suggested that effects of enhanced N are weakened along a positive DOM gradient, indicating that algal productivity in clear-water lakes is more sensitive to increased N availability than darker lakes. This implies that clear-water lakes are especially sensitive to N deposition because low DOM and low nitrogen retention capacity are causally linked through low pools of soil organic matter in their catchments, a consequence of little vegetation and shallow soils (Evans et al., 2006). Thus, we find that dystrophic lakes are indeed sensitive to N deposition but less so than clear-water lakes, which is because of a) the effect of DOM on light availability, which limits the ecological response to increases in nitrogen, b) the catchment properties that lead to high DOM also lead to higher retention of atmospheric nitrogen deposition.

The evidence that has accumulated since 2010 allows for a better quantification of critical loads (CLs) for EUNIS habitat type C1.1 (permanent oligotrophic lakes, ponds and pools) and C1.4 (peaty, dystrophic waters). Clear-water sub-Arctic and alpine lakes (included in C1.1) in catchments with little vegetation and limited soil cover have a slightly lower critical threshold for responses to N deposition than boreal lakes (also in C1.1) from forested regions. Based on the evidence referred to above, i.e. a combination of experiments (whole-lake manipulations, mesocosm experiments) and gradient studies (alone, or in combination with bioassays) that document responses of algal productivity to N deposition, we propose a critical load range for nitrogen as a nutrient of 2-6 kg N ha<sup>-1</sup> yr<sup>-1</sup>, where clear-water sub-Arctic and alpine lakes have a CL of 2-4 kg N ha<sup>-1</sup> yr<sup>-1</sup> while boreal (non-dystrophic) lakes have a CL range of 3-6 kg N ha<sup>-1</sup> yr<sup>-1</sup>. Dystrophic, humic lakes (C1.4) in catchments with forests, wetlands and well-developed forest soils are less sensitive to N deposition because more N is retained in the catchment, while high DOM limits the eutrophying effects because of light limitation. Hence, their CL range is proposed to be 5-10 kg N ha<sup>-1</sup> yr<sup>-1</sup>. The critical load should only be applied to waters with low alkalinity and no significant agricultural or other direct human inputs.

It is possible to argue that the range for sub-Arctic and alpine lakes should start at a lower limit, given that N-limitation has been found at N-deposition levels between 0.5 and 2 kg N ha<sup>-1</sup> yr<sup>-1</sup> (Chapter 2 in this report; the critical load range of 1.0-3.0 kg N ha<sup>-1</sup> yr<sup>-1</sup> for western lakes in the USA presented by Baron et al. (2011)). Biological effects of enhanced N deposition also depend on DOC concentrations (controlling light limitation), which are associated with catchment carbon density and



climate in high-latitude and high elevation catchments. The data analyses we have used to justify ranges of empirical critical loads of N do not differentiate between biological responses in relation to DOC concentrations. Thus, we now conclude that further data analysis would be preferable in order to make robust recommendations for the lower end of the CL range.

The above literature review, as well as Bobbink and Hettelingh (2011) focused on critical loads for lake ecosystems. However, the analysis of river water elemental stoichiometry presented in Chapter 2 of this report suggests that also natural rivers can be driven from N limitation to P limitation as a consequence of N deposition, similar to lakes. We conclude that ecological responses in rivers related to N deposition is an area that deserves more attention.

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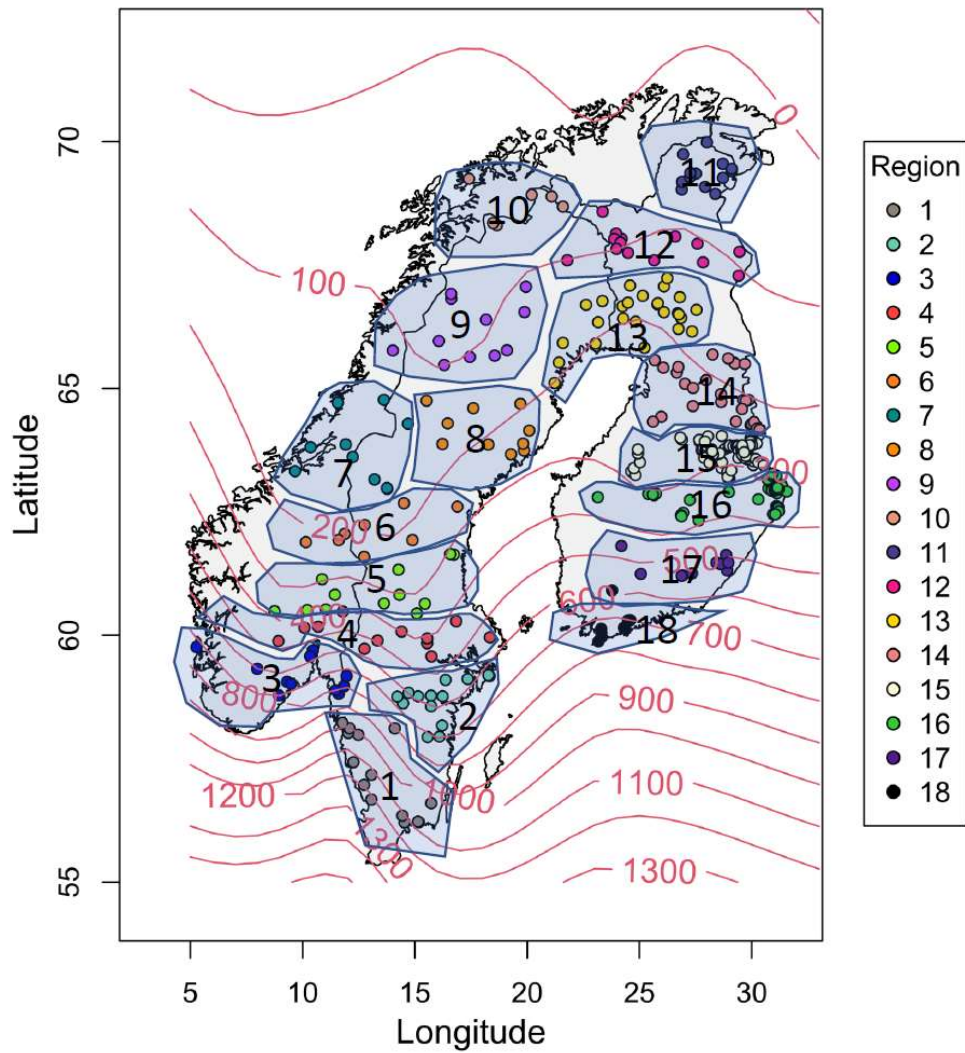
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## Appendix A: Supporting information

### Chapter 2



**Figure A. 1** Overview of the 296 natural lakes with predicted N deposition (kg N ha<sup>-1</sup> yr<sup>-1</sup>) from a GAM added as isolines. The lakes were distributed to 18 regions, which are marked with different colors and enclosed by blue polygons.

**Table A. 1** Overview of the regions, the number of lakes per region (# lakes) and median (standard deviation) values of N deposition ( $\text{kg ha}^{-1} \text{year}^{-1}$ ), chlorophyll *a* (chl*a*) and relevant water chemical parameters (TP = total phosphorus; TN = total nitrogen; DIN = dissolved inorganic nitrogen) per region.

region	# lakes	N deposition ( $\text{kg ha}^{-1} \text{year}^{-1}$ )	Chl <i>a</i> ( $\mu\text{g/L}$ )	TP ( $\mu\text{g/L}$ )	TN ( $\mu\text{g/L}$ )	DIN ( $\mu\text{g/L}$ )	TN:TP	DIN:TP
reg1	14	10.4 (2.2)	3.3 (3.9)	7.6 (6.2)	385 (135)	41.8 (73)	55 (68)	5.2 (49.2)
reg2	15	6.0 (0.5)	4.3 (8.7)	8.3 (7.9)	401 (158)	5.1 (11)	46 (12)	0.6 (1.3)
reg3	11	7.4 (1.5)	1.6 (3.4)	4.3 (3.5)	343 (53)	42.0 (47.2)	62 (38)	7.5 (18.9)
reg4	12	4.7 (0.7)	3.0 (15.1)	7.9 (13.5)	310 (225)	26.2 (24.5)	38 (14)	3.0 (4.0)
reg5	12	3.4 (0.8)	2.2 (5.7)	5.8 (9.7)	277 (78)	5.9 (19.3)	45 (17)	1.2 (3.9)
reg6	9	1.7 (0.3)	1.8 (0.9)	6.8 (2.1)	210 (79)	4.5 (23.6)	28 (7)	0.5 (3.6)
reg7	10	1.4 (0.5)	1.0 (0.4)	3.6 (6.5)	168 (79)	3.8 (14.8)	46 (14)	1.1 (6.5)
reg8	10	1.8 (0.6)	3.0 (4.5)	10.8 (8.9)	323 (122)	4.0 (8.4)	31 (12)	0.5 (0.7)
reg9	11	1.0 (0.2)	1.3 (0.7)	3.5 (1.7)	139 (39)	5.0 (10.5)	42 (42)	1.4 (5.1)
reg10	6	0.5 (0.3)	0.8 (0.8)	4.0 (2.2)	132 (40)	2.5 (2.4)	37 (27)	1.0 (1.9)
reg11	13	0.8 (0.1)	1.0 (0.8)	4.5 (1.8)	170 (69)	2.4 (6.3)	40 (11)	0.5 (1.4)
reg12	13	1.0 (0.2)	5.8 (4.1)	15.8 (7.1)	335 (109)	2.6 (14.7)	23 (7)	0.2 (0.7)
reg13	26	1.7 (0.3)	5.0 (4.8)	12.8 (7.5)	306 (145)	2.3 (3.3)	25 (11)	0.2 (0.5)
reg14	23	2.0 (0.3)	5.8 (9.2)	14.2 (11.3)	355 (164)	2.5 (1.7)	24 (12)	0.2 (0.2)
reg15	68	2.8 (0.2)	6.0 (8.2)	17.5 (8.7)	400 (103)	2.5 (9.3)	23 (9)	0.2 (0.9)
reg16	42	3.1 (0.9)	5.4 (9.2)	12.5 (8.2)	356 (127)	2.5 (7.9)	28 (13)	0.2 (0.7)
reg17	11	5.0 (0.7)	5.1 (5.5)	10.0 (4.6)	420 (111)	2.8 (6.9)	37 (14)	0.4 (1.2)
reg18	9	7.8 (1.0)	3.2 (4.5)	8.2 (7.0)	306 (126)	2.6 (1.3)	47 (18)	0.5 (0.2)



## Appendix B: Reports and publications from the ICP Waters programme

All reports from the ICP Waters programme from 2000 up to present are listed below. Reports before year 2000 can be listed on request. All reports are available from the Programme Centre. Reports and recent publications are also accessible through the ICP Waters website; <http://www.icp-waters.no/>

- Garmo, Ø., Furuseth, I.S., and Austnes, K. (editors) 2021. Proceedings of the 37<sup>th</sup> Task Force meeting of the ICP Waters Programme held on-line, April 28-29, 2021. NIVA report SNO 7657-2021. **ICP Waters report 145/2021**
- Velle, G., Birkeland, I.B., Johannessen, A. and Landås, T.S. 2020. Biological intercalibration: Invertebrates 2020. NIVA SNO 7556-2020. **ICP Waters report 144/2020**
- Gundersen, C.B. and Bryntesen, T. 2021. Intercomparison 2034: pH, Conductivity, Alkalinity, NO<sub>3</sub>-N, Cl, SO<sub>4</sub>, Ca, Mg, Na, K, TOC, Tot-P, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA SNO 7621-2021. **ICP Waters report 143/2021.**
- Gundersen, C.B. 2020. Intercomparison 2034: pH, Conductivity, Alkalinity, NO<sub>3</sub>-N, Cl, SO<sub>4</sub>, Ca, Mg, Na, K, TOC, Tot-P, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA SNO 7549-2020. **ICP Waters report 143/2020.** *Obs! This report has been revised (see ICP Waters 143/2021 above).*
- Garmo, Ø., Arle, J., Austnes, K. de Wit, H., Fölster, J., Houle, D., Hruška, J., Indrikson, I., Monteith, D., Rogora, M., Sample, J.E., Steingruber, S., Stoddard, J.L., Talkop, R., Trodd, W., Ułańczyk, R.P. and Vuorenmaa, J. 2020. Trends and patterns in surface water chemistry in Europe and North America between 1990 and 2016, with particular focus on changes in land use as a confounding factor for recovery. NIVA report SNO 7479-2020. **ICP Waters report 142/2020**
- Gundersen, C.B. 2019. Intercomparison 1933: pH, Conductivity, Alkalinity, NO<sub>3</sub>-N, Cl, SO<sub>4</sub>, Ca, Mg, Na, K, TOC, Tot-P, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA SNO 7445-2019. **ICP Waters report 141/2019.**
- Velle, G., Birkeland, I.B., Johannessen, A. and Landås, T.S. 2019. Biological intercalibration: Invertebrates 2019. NIVA SNO 7433-2019. **ICP Waters report 140/2019**
- Garmo, Ø., Austnes, K. and Vuorenmaa, J. (editors) 2019. Proceedings of the 35<sup>th</sup> Task Force meeting of the ICP Waters Programme in Helsinki, June 4-6, 2019. NIVA report SNO 7437-2019. **ICP Waters report 139/2019**
- Velle, G., Johannessen, A. and Landås, T.S. 2018. Biological intercalibration: Invertebrates 2018. NIVA report SNO 7314-2018. **ICP Waters report 138/2018**
- Escudero-Oñate, C. 2018. Intercomparison 1832: pH, Conductivity, Alkalinity, NO<sub>3</sub>-N, Cl, SO<sub>4</sub>, Ca, Mg, Na, K, TOC, Tot-P, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA report SNO 7316-2018. **ICP Waters report 137/2018.**
- Garmo, Ø., Ułańczyk, R. and de Wit, H. (eds.) 2018. Proceedings of the 34<sup>th</sup> Task Force meeting of the ICP Waters Programme in Warsaw, May 7-9, 2018. NIVA report SNO 7298-2018. **ICP Waters report 136/2018.**
- Austnes, K. Aherne, J., Arle, J., Čičendajeva, M., Couture, S., Fölster, J., Garmo, Ø., Hruška, J., Monteith, D., Posch, M., Rogora, M., Sample, J., Skjelkvåle, B.L., Steingruber, S., Stoddard, J.L., Ułańczyk, R., van Dam, H., Velasco, M.T., Vuorenmaa, J., Wright, R.F., de Wit, H. 2018. Regional assessment of the current extent of acidification of surface waters in Europe and North America. NIVA report SNO 7268-2018. **ICP Waters report 135/2018**

- Escudero-Oñate, C. 2017. Intercomparison 1731: pH, Conductivity, Alkalinity, NO<sub>3</sub>-N, Cl, SO<sub>4</sub>, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA report SNO7207-2017. **ICP Waters report 134/2017.**
- Halvorsen, G.A., Johannessen, A. and Landås, T.S. 2017. Biological intercalibration: Invertebrates 2017. NIVA report SNO 7198-2017. **ICP Waters report 133/2017.**
- Braaten, H.F.V., Åkerblom, S., de Wit, H.A., Skotte, G., Rask, M., Vuorenmaa, J., Kahilainen, K.K., Malinen, T., Rognerud, S., Lydersen, E., Amundsen, P.A., Kashulin, N., Kashulina, T., Terentyev, P., Christensen, G., Jackson-Blake, L., Lund, E. and Rosseland, B.O. 2017. Spatial and temporal trends of mercury in freshwater fish in Fennoscandia (1965-2015). NIVA report SNO 7179-2017. **ICP Waters report 132/2017.**
- Garmo, Ø., de Wit, H. and Fölster, J. (eds.) 2017. Proceedings of the 33rd Task Force meeting of the ICP Waters Programme in Uppsala, May 9-11, 2017. NIVA report SNO 7178-2017. **ICP Waters report 131/2017.**
- Anker Halvorsen, G., Johannessen, A. and Landås, T.S. 2016. Biological intercalibration: Invertebrates 2016. NIVA report SNO 7089-2016. **ICP Waters report 130/2016.**
- Escudero-Oñate, C. 2016. Intercomparison 1630: pH, Conductivity, Alkalinity, NO<sub>3</sub>-N, Cl, SO<sub>4</sub>, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA report SNO 7081-2016. **ICP Waters report 129/2016.**
- De Wit, H. and Valinia, S. (eds.) 2016. Proceedings of the 32st Task Force meeting of the ICP Waters Programme in Asker, Oslo, May 24-26, 2016. NIVA report SNO 7090-2016. **ICP Waters report 128/2016.**
- Velle, G., Mahlum, S., Monteith, D.T., de Wit, H., Arle, J., Eriksson, L., Fjellheim, A., Frolova, M., Fölster, J., Grudule, N., Halvorsen, G.A., Hildrew, A., Hruška, J., Indriksone, I., Kamasová, L., Kopáček, J., Krám, P., Orton, S., Senoo, T., Shilland, E.M., Stuchlík, E., Telford, R.J., Ungermanová, L., Wiklund, M.-L. and Wright, R.F. 2016. Biodiversity of macro-invertebrates in acid-sensitive waters: trends and relations to water chemistry and climate. NIVA report SNO 7077-2016. NIVA report SNO 7077-2016. **ICP Waters report 127/2016.**
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- De Wit, H., Hettelingh, J.P. and Harmens, H. 2015. Trends in ecosystem and health responses to long-range transported atmospheric pollutants. NIVA report SNO 6946-2015. **ICP Waters report 125/2015.**
- Fjellheim, A., Johannessen, A. and Landås, T.S. 2015. Biological intercalibration: Invertebrates 1915. NIVA report SNO 6940-2015. **ICP Waters report 124/2015.**
- Escudero-Oñate, C. 2015 Intercomparison 1529: pH, Conductivity, Alkalinity, NO<sub>3</sub>-N, Cl, SO<sub>4</sub>, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA report SNO 6910-2015. **ICP Waters report 123/2015.**
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