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**Abstract**  
 Atmospheric deposition of nitrogen (N) leads to enhanced leaching of N species to surface waters in natural and semi-natural ecosystems. The reigning paradigm of freshwater primary productivity is limitation by phosphorus (P), which suggests that additional N does not affect growth of algae and higher aquatic plants. This literature review shows that increased availability of N, related to atmospheric N deposition, in nutrient-poor temperate, boreal and arctic lakes affect freshwater biology. Lake sediment studies show shifts in algal communities and increases in algal growth related to higher N concentrations. Regional surveys in boreal lakes show higher chlorophyll concentrations per unit P in areas with higher N deposition, indicative of higher primary production. Experimental nutrient additions in lakes (mesocosm studies and bioassays) support the finding of the regional surveys by showing that N limitation of algal growth is common, especially under conditions of low N availability. Increased N availability can stimulate productivity of sediment- and rock-dwelling algae (benthic algae) but here data are scarce. Water plant dynamics from oligotrophic lakes relate significant plant community shifts (loss of key species, dominance of new species) to increased ammonium deposition and increased availability of N species, but other factors (CO<sub>2</sub>, liming) may also explain some of the observed changes. The findings in this report are used to evaluate critical loads for atmospheric N in freshwater habitats. New empirical loads for nutrient-N are proposed, while an approach for modelling of critical loads of nutrient N with a mass balance model is suggested.

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CONVENTION ON LONG-RANGE  
TRANSBOUNDARY AIR POLLUTION

INTERNATIONAL COOPERATIVE PROGRAMME ON  
ASSESSMENT AND MONITORING EFFECTS OF AIR  
POLLUTION ON RIVERS AND LAKES

**Nutrient enrichment effects of atmospheric N deposition  
on biology in oligotrophic surface waters – a review**

Prepared at the ICP Waters Programme Centre  
Norwegian Institute for Water Research  
Oslo, August 2010

## Preface

The international cooperative programme on assessment and monitoring of 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 (LRTAP) in July 1985. Since then ICP Waters has been an important contributor to document the effects of implementing the Protocols under the Convention. Numerous assessments, workshops, reports and publications covering the effects of long-range transported air pollution has been published over the years.

The ICP Waters Programme Centre is hosted by the Norwegian Institute for Water Research (NIVA), while the Norwegian Climate and Pollution Agency (Klif) leads the programme. The Programme Centre's work is supported financially by Klif.

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 in the programme on a regular basis.

ICP Waters is based on existing surface water monitoring programmes in the participating countries, implemented by voluntary contributions. The ICP site network is geographically extensive and includes long-term data series (more than 20 years) for many sites. The programme conducts yearly chemical and biological intercalibrations.

This report focuses on the possible effects of N-deposition on freshwater biology and nutrient status of oligotrophic rivers and lakes. The report is a literature review.

We would like to thank all those who have contributed to this report with comments, especially Chris Curtis, Aldo Marchetto, Tom Clair and Anders Wilander who volunteered at the 2009 Task Force meeting to comment the draft report.



*Brit Lisa Skjelkvåle*  
ICP Waters Programme Centre

Oslo, September 2010

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## Abstract

Atmospheric deposition of nitrogen (N) leads to enhanced leaching of N species to surface waters in natural and semi-natural ecosystems. The reigning paradigm of freshwater primary productivity is limitation by phosphorus (P), which suggests that additional N does not affect growth of algae and higher aquatic plants.

This literature review shows that increased availability of N, related to atmospheric N deposition, in nutrient-poor temperate, boreal and arctic lakes has effects on freshwater biology. Lake sediment studies (paleolimnology) show shifts in algal communities and increases in algal growth related to higher N concentrations. Regional surveys in boreal lakes show higher chlorophyll concentrations per unit P in areas with higher N deposition, indicative of higher primary production. Experimental nutrient additions in lakes (mesocosm studies and bioassays) support the finding of the regional surveys by showing that N limitation of algal growth is common, especially under conditions of low N availability. Increased N availability can stimulate productivity of sediment- and rock-dwelling algae (benthic algae) but here data are scarce. Water plant dynamics from oligotrophic lakes relate significant plant community shifts (loss of key species, dominance of new species) to increased ammonium deposition and increased availability of N species, but other factors (CO<sub>2</sub>, liming) may also explain some of the observed changes .

To what extent N enrichment in oligotrophic waters eventually may cascade up the food chain and affect biodiversity is hard to predict. This will depend on possible changing dominance of pelagic versus littoral productivity and whether habitats will be affected by changing growth of macrophytes and benthic algae. Downstream nutrient enrichment due to N leaching from upland catchments is not evaluated here, but is known to lead to eutrophication in coastal zones and may also influence eutrophication in agriculturally affect freshwater systems.

The findings in this report are used to evaluate critical loads for atmospheric N in freshwater habitats. Existing empirical loads for nutrient-N are updated, while an approach for modelling of critical loads of nutrient N with a mass balance model is suggested.

## Extended summary

### Background and aim

Atmospheric deposition of nitrogen (N) leads to enhanced leaching of N species to surface waters in natural and semi-natural ecosystems. The reigning paradigm of freshwater primary productivity is limitation by phosphorus (P), which suggests that additional N does not affect growth of algae and higher aquatic plants. This paradigm is presently strongly challenged. The aim of this report is to give an overview of literature concerning effects of N enrichment on biology in nutrient-poor aquatic ecosystems in arctic, boreal and temperate regions. The choice for nutrient-poor ecosystems is guided by the primary focus of ICP Waters on natural ecosystems where local pollution sources (e.g. domestic sewage, industrial waste water, agriculture, etc.) are absent. Thus, any ecosystem responses to N will be most likely related to atmospheric long-range transported N. Downstream nutrient enrichment due to N leaching from upland catchments is not evaluated here, but is known to occur in coastal zones and may also influence eutrophication in agriculturally affected freshwater systems.

The review was predominantly focused on peer-reviewed literature. Historical records of lake sediments (paleolimnology), lake surveys and experimental studies were included. Effects on algae (free-floating phytoplankton and sediment-dwelling benthic algae), water plants (macrophytes) and invertebrates (insects, snails) were evaluated. The findings in this report are used to evaluate critical loads for atmospheric N in freshwater habitats. Existing empirical loads for nutrient-N are updated, while an approach for modelling of critical loads of nutrient N with a mass balance model is suggested.

### Paleolimnological evidence for N-limitation

Lake sediment studies (paleolimnology) show shifts in algal communities and increases in algal growth related to higher N concentrations. Most paleolimnological papers on possible effects of N enrichment on phytoplankton communities in oligotrophic lakes are studies published from 2000 and onwards. The studies have a wide geographical and climatic range and include temperate, alpine and arctic lakes from Europe and North-America, with N deposition levels between  $<1$  and  $>10$  kg N ha<sup>-1</sup> yr<sup>-1</sup>. The changes in community structures are in varying degrees attributed to N enrichment, climate and/or to interactions of these. The evidence presented support the hypothesis that N deposition increases algal productivity and can change algal community structure in oligotrophic lakes arctic, alpine and temperate lakes, at levels of N deposition as low as  $<1$  to 3 kg N ha<sup>-1</sup> yr<sup>-1</sup>, but also in lakes receiving higher loads of atmospheric N ( $>10$  kg N ha<sup>-1</sup> yr<sup>-1</sup>). However, not all studies show evidence of higher productivity related to increased N, and some changes in community structure are interpreted as climate effects rather than N enrichment effects.

### Phytoplankton and benthic algae

Whole-lake experiments, mesocosms and bioassays are all accepted methods used to experimentally investigate nutrient controls on freshwater primary productivity, in declining order of ecological realism and costs. Lake surveys along various gradients (deposition, climate, vegetation cover) are also used to statistically related water chemistry and indicators of algal growth. In many studies, a combination of all, or some, of these methods are used to gain insights in nutrient controls on productivity.

Whole-lake manipulations have often been done by manipulating both N and P supply, and conflicting interpretations exist with respect to whether N-limitation has been detected. However, where only N was added increases in productivity of both phytoplankton and sediment-dwelling algae were observed, and several authors interpret results from lakes receiving both N and P as showing evidence of N-limitation in addition to P-limitation and co-limitation.

Regional surveys in boreal lakes in Europe and North America with N deposition between  $<1$  and  $>14$  kg N ha<sup>-1</sup> yr<sup>-1</sup> show higher chlorophyll concentrations per unit P in areas with higher N deposition, indicative of higher primary production. Experimental nutrient additions in lakes (mesocosm studies

and bioassays) support the finding of the regional surveys by showing that N limitation of algal growth is common, especially under conditions of low N availability. Increased N availability can increase productivity of sediment- and rock dwelling algae (benthic algae) but here the number of studies is limited. It is suggested that N deposition may have shifted primary productivity in oligotrophic lakes from N limitation to P limitation. Also, some studies suggest that totN to totP ratios to evaluate nutrient deficiencies may underestimate N-deficiency as total N in oligotrophic lakes is often partly unavailable. Often, inorganic N to totP ratios are better predictors of responses to nutrient additions.

### **Macrophytes**

Effects of nitrogen on macrophytes in oligotrophic waters have not been investigated over a wide range of species, regions or atmospheric loads of N. The typical water plant communities of oligotrophic acid lakes (isoetids) – acid-tolerant and able to live in conditions of low N availability - have declined strongly in Western Europe in the 20<sup>th</sup> century. In some regions (Atlantic lowlands), the isoetid vegetation was replaced by other species like bulbous rush (*Juncus bulbosus*) and *Sphagnum* while in boreal lakes, coexistence of isoetids and bulbous rush is common. Lake surveys from Western Europe and some targeted experimental studies indicate that the shift in vegetation community is not related to acidification but rather to increases in nutrient availability, especially NH<sub>4</sub>. The links between community shifts, increased N availability and deposition are not clear-cut because other factors like liming, sediment processes, climate and interspecific competition also play a role. Species-specific characteristics like ability to utilise NO<sub>3</sub> or NH<sub>4</sub> will also co-determine dominance or loss of species. In Norway, liming has led to loss of isoetid vegetation, probably due to increased CO<sub>2</sub> concentrations and changing N availability in sediments. Most data on lake vegetation stem from the 1980s and 1990s and more recent data are generally lacking.

### **Invertebrates**

To what extent N enrichment in oligotrophic waters eventually may cascade up the food chain and affect organisms that depend on algae and water plants as a food source or as habitat is hard to predict. This will depend on possible changing dominance of pelagic versus littoral productivity and whether habitats will be affected by changing growth of macrophytes and benthic algae. Interactions between N availability and invertebrates in oligotrophic waters have hardly been studied.

### **Critical loads for nutrient N**

The findings in this report are used to evaluate critical loads for atmospheric N in freshwater habitats. Existing empirical loads for nutrient-N are updated and new critical loads are proposed, while an approach for modelling of critical loads of nutrient N with a mass balance model is suggested.

# 1. Introduction

After the industrial revolution, drastic changes have occurred in the global nitrogen (N) cycle. Availability and mobility of reactive N species have increased due to use of fertilizers and fossil fuel combustion, leading to long-range transport of N compounds from industrial and agricultural hotspots to background areas of the earth (Vitousek et al., 1997). The enhanced supply of N to natural ecosystems has led to changes and reductions in biodiversity (Krupa, 2003), increased plant growth (Vitousek and Howarth, 1991), N-enrichment of soils (Moldan et al., 2006; Nadelhoffer et al., 1999), higher emissions of N<sub>2</sub>O, increased binding of CO<sub>2</sub> (De Vries et al., 2006), long-term (Skjelkvale et al., 2005; Stoddard et al., 2001) and episodic (Murdoch and Stoddard, 1992; Schaefer et al., 1990; Stoddard, 1994) acidification of soils, groundwater and surface waters and larger riverine transfer of N to estuaries and coastal areas leading to eutrophication of coastal waters (Smith, 2003; Vitousek and Howarth, 1991).

Nitrogen deposition in natural ecosystems can lead to enhanced leaching of inorganic N from soils to surface waters, in cases where the supply of N is greater than plant and microbial demand. This is also referred to as 'nitrogen saturation' (Aber et al., 1989; Stoddard, 1994). Old forest, prolonged high N deposition and large soil N pools can in concert lead to significant N losses from terrestrial ecosystems. Regional datasets of surface water nitrate show that watersheds receiving over 10 kg N ha<sup>-1</sup> yr<sup>-1</sup> in atmospheric deposition leach considerably more nitrate than other watersheds, and that significant leaching can occur at deposition levels as low as 5 kg N ha<sup>-1</sup> yr<sup>-1</sup> (Stoddard et al., 2001). In temperate forests, N leaching was controlled by N deposition levels in combination with forest floor C to N ratio - a measure for soil N enrichment status (MacDonald et al., 2002).

In natural ecosystems, inorganic N species that are leached from soils to surface waters are predominantly in the form of nitrate (NO<sub>3</sub>). The relative contribution of NO<sub>3</sub> compared to sulphate (SO<sub>4</sub>) to acidification of surface waters can be expressed using the NAR ratio (Nitrogen Acidification Ratio – (NO<sub>3</sub>/(NO<sub>3</sub>+SO<sub>4</sub>) in µeq L<sup>-1</sup>). Sulphate is usually the dominating ion in anthropogenically acidified surface waters, but its importance has decreased with the ongoing surface water recovery. A survey in North America, referenced in Stoddard (1994), showed that the NAR in 1985 was most often below 0.1 but in regions with low SO<sub>4</sub> concentrations NAR was often between 0.2 and 0.4. Other NAR ratios reported for the early 1980s are usually below 0.2 (USA, Scotland, Sweden, Norway) but in Germany and Czecho-Slovakia (presently the Czech Republic and Slovakia) NAR values between 0.3 and 0.4 were common (Henriksen and Brakke, 1988). A more recent analysis of NAR values in ICP Water sites for the period 1990-2005 (Kaste in Skjelkvåle and De Wit (2007)) indicates that the vast majority of sites in North America have a NAR below 0.25 while for approximately 30% of ICP Waters sites in Europe NAR is between 0.25 and 0.5.

The hypothesis of N saturation suggests that the unavoidable effect of prolonged N-enrichment of ecosystems is increased N leaching. However, there is so far little evidence of widespread increasing trends in surface water nitrate concentrations in Europe and North America (Skjelkvale and de Wit, 2007). In fact, reports of nitrate decline appear to be more common, both in Europe and North America (Burns et al., 2006; de Wit et al., 2008; Goodale et al., 2005; Ulrich et al., 2006; Vesely et al., 2002). However, upward nitrate trends are reported too, for example in rivers draining forested catchments in the Lake Maggiore watershed in northern Italy (Rogora, 2007) and in the Rocky Mountains in North America (Baron et al., 2009). The observed trends do not appear to have one common control and are discussed in terms of changes in N deposition, temperature, forest regrowth, changes in winter climate and N saturation. Comparison of regional patterns in nitrate in remote lakes, however, indicates that higher nitrate concentrations are often found in lakes in areas with higher N deposition (Bergstrom and Jansson, 2006; Rogora et al., 2008).

So far, the main reason for studying controls of N leaching to surface waters was to predict effects of N deposition on future acidification of surface waters (Cosby et al., 2001; Curtis et al., 1998; Curtis et al., 2005; Dise and Wright, 1995; Stoddard, 1994; Wright et al., 2001). Effects of N leaching on

surface water eutrophication was explicitly stated as unlikely because of phosphorus (P) limitation of primary productivity (Stoddard, 1994). Any additional N in a P-limited system could thus not be expected to stimulate algal growth. Phosphorus limitation of primary productivity surface waters has been the dominating view since the 1970s (Hecky and Kilham, 1988; Schindler, 1971; Schindler, 1977). However, this view has been challenged recently in reviews (Elser et al., 2007; Lewis and Wurtsbaugh, 2008; Sterner, 2008) although earlier papers also stressed that N availability was important for algal production (Elser et al., 1990; Kratzer and Brezonik, 1981). New studies and reinterpretation of old results suggest that co-limitation of primary productivity by N and P is common, and that in some cases autotrophs respond similarly to single nutrient supply of N and P. A recent study challenges whether nutrient availability is the limiting factor for productivity in nutrient-poor, highly coloured lakes and suggests that light-limitation may be more important (Karlsson et al., 2009).

Policy and financial implications of nutrient productivity controls are potentially large as measures to reduce N loading are much more costly than those required to reduce P loading. This has been extensively debated in the journal *Science* (Bryhn and Hakanson, 2009; Conley et al., 2009a; Conley et al., 2009b; Jacoby and Frazer, 2009; Schelske, 2009; Schindler and Hecky, 2009). In their first paper, Conley and co-workers argue for reduced input of both N and P to combat coastal eutrophication, because N inputs can stimulate phytoplankton growth under certain conditions, especially where sediments are rich in P and P is rapidly recycled. They used in part model predictions to argue their case. In following letters to the editor, this view is criticized based on insights gained from whole-lake manipulations (Schindler and Hecky, 2009), examples of successful P-control to combat eutrophication, and discussion of the role of P in sediments (Schelske, 2009), the lack of models that properly describe and predict P and N dynamics in coastal systems (Bryhn and Hakanson, 2009) and the call for a wider view of recovery from eutrophication including 'legacy loads' and additional restoration measures beyond reducing future nutrient loadings (Jacoby and Frazer, 2009). Conley and co-workers address these issues in their reply (Conley et al., 2009a). This debate is important for environments that receive loadings of both N and P. For lakes and rivers that are primarily affected by increased N loadings – in background areas – no direct conflict exists between policy measures controlling N or P emissions.

The aim of this report is to give an overview of literature concerning effects of N enrichment on biology in nutrient-poor aquatic ecosystems in arctic, boreal and temperate regions. The choice for nutrient-poor ecosystems is guided by the primary focus of ICP Waters on natural ecosystems where local pollution sources (e.g. domestic sewage, industrial waste water, agriculture, etc.) are absent. Thus, any ecosystem responses to N will be most likely related to atmospheric long-range transported N.

The report provides background to the 'P limitation paradigm' and recent challenges to this paradigm, and then supplies literature reviews of N effects from paleolimnological studies, on phytoplankton, benthic algae, water plants and invertebrates. The report concludes by putting the results from the literature review in a policy-relevant context by evaluating present empirical critical loads for nutrient N in freshwaters and makes some suggestions for an approach for modelling of critical loads for nutrient N in freshwaters.

## 2. Nutrient limitation of primary productivity in freshwaters

### 2.1. The P-limitation paradigm

The ‘P-limiting paradigm’ of primary productivity in freshwaters is widely accepted in limnological textbooks and reviews as summarized by Sterner, 2008 (Table 1 in op.cit.). This ‘paradigm’ has been critically reviewed in three recent papers (Elser et al., 2007; Lewis and Wurtsbaugh, 2008; Sterner, 2008). Sterner (2008) provides an interesting overview on its historical development. The following is directly drawn from his paper.

*“Prior to mid-1970s, there was no consensus on controls of algal production in lakes. Eutrophication was a hot issue partly due to large public concern about eutrophied lakes and rivers. Commercial actors promoted the view of CO<sub>2</sub>-limitation rather than anthropogenically affected nutrients like N and P. The CO<sub>2</sub>-limitation hypothesis was rejected based on insights drawn from three types of empirical evidence: stoichiometrical observations from regional lake surveys, whole-lake fertilizations and trends within individual lakes. Schindler argued in his hallmark paper on the evolution of P-limitation in lakes (1977) that as C and N were available in large atmospheric reservoirs (CO<sub>2</sub> and N<sub>2</sub>), deficiencies of these elements would be overcome given sufficient time – a viewpoint considering an evolutionary scale. As there is no such reservoir for P, no similar mechanisms can compensate for P-deficiency. So, transient deficiencies of C and N may exist, but ultimately P is the controlling element. This view has dominated in literature and in textbooks although evidence of multi-resource limitation has accumulated. The obvious question is whether this new evidence can be reconciled with the P-limitation paradigm or whether significant adaptation is warranted.”* Adapted from Sterner, 2008, p 434-436.

### 2.2. Evidence for P-limitation from whole-lake manipulations

One of the most influential papers in modern limnology is written by David W. Schindler (1977) on P limitation of primary productivity in freshwaters. He argued on the basis of whole-lake nutrient enrichment experiments that P supply is the limiting nutrient for algal growth in most eutrophied lakes.

In the whole-lake experiments, P and N were added with N in adequate or excess amounts. Reducing the N input relative to the P input lead to changes in algal species composition with an increase in blue-green algae – capable of fixing atmospheric N – at the cost of algae that do not possess that ability. Fixation of atmospheric N led to stabilization of the N to P ratio, despite the reduced input of mineral N during the experimental manipulation. In another lake manipulation where only P fertilizer was added, no N fixers appeared, but still phytoplankton chlorophyll increased somewhat. The author concluded that on the long-term, N deficiency created by sudden increase of P, will be balanced by changes in phytoplankton species composition towards more blue-green algae (N-fixers) which are often toxic and objectionable from a water quality standpoint. Reducing N inputs to combat eutrophication may even have adverse effects because it may stimulate blue-green algae. Continuation of the whole-lake manipulations resulted in a 37-year time series of phytoplankton responses to nutrient supply and the conclusion of P-limitation was further solidified (Schindler et al., 2008).

Early results from Schindler’s whole lake experiments in the Experimental Lakes Area (ELA - [www.experimentallakesarea.ca](http://www.experimentallakesarea.ca)) were interpreted in apparently conflicting ways, as pointed out by Lewis and Wurtsbaugh (2008). Data from unfertilized lakes in the ELA area were interpreted as dual limitation (Fee, 1979) whereas Schindler (1980) argued for P limitation. Fee (1979) presented results of N, P and N+P additions in the ELA for the mid-1970s (Fee, 1979, Table 1; visually presented in Lewis and Wurtsbaugh (2008), Figure 4). Phytoplankton biomass showed similar responses to N and P additions and much stronger responses to dual additions of N and P. Additionally, mesocosm studies

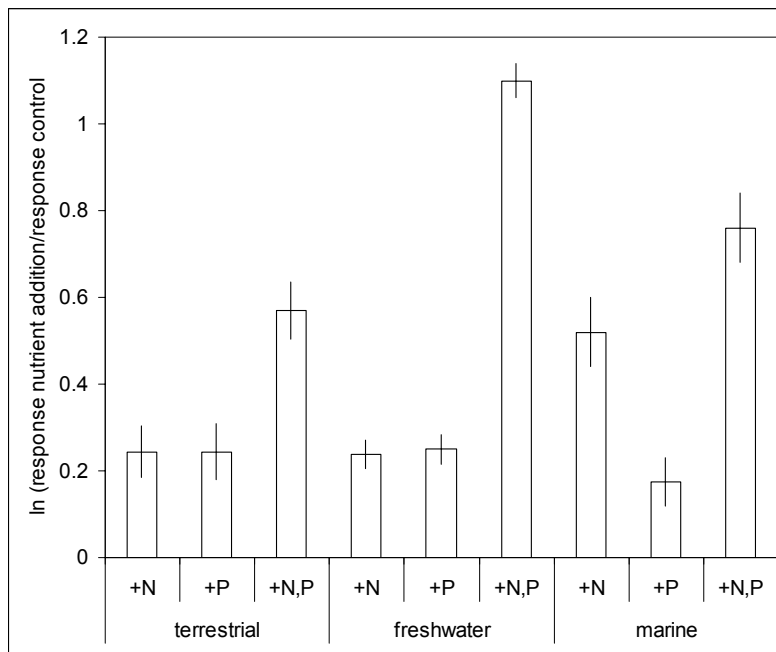
from ELA (to study compensatory nutrient response) were conducted in lakes that had previously received large inputs of N (Levine and Schindler, 1992). This may have influenced the results but was not accounted for in their interpretation.

Sterner (2008) points out that whole-lake manipulations were usually done in oligotrophic lakes, while single nutrient addition experiments in eutrophic lakes are generally lacking. This means that a relatively small dataset (nutrient enrichment experiments in oligotrophic lakes) is used to infer to mechanisms of phytoplankton growth by P-limitation on a much larger spatial and temporal scale. He also refers to Bergstrom and Jansson (2006) who suggest that historical anthropogenic loadings of N may have brought oligotrophic lakes from N to P limitation.

### 2.3. Experimental evidence for co-limitation of primary production

Evidence for co-limitation of phytoplankton growth was gathered in an early paper (Elser et al., 1990). It covered a large range of nutrient enrichment bioassays (including whole-lake experiments) where algal growth responded most strongly to dual addition of N and P. Single additions of either N or P gave similar responses, while dual additions gave a ca 2.5 times stronger response than for single nutrients. The authors concluded that N as a limiting nutrient deserved more attention.

A more comprehensive review on nutrient controls of primary production in terrestrial, freshwater and marine ecosystems concluded similarly (Elser et al., 2007). This meta-analysis across ecosystems included data from 653 freshwater, 243 marine and 173 terrestrial experiments. Results (biomass and/or production or equivalent terms) were expressed in the logarithmic ratio of response to the nutrient addition divided by response in the control treatment (**Figure 1**). The authors concluded that all systems were surprisingly similar with respect to nutrient controls of photosynthesis and that co-limitation of productivity was most common, also in freshwaters. In freshwaters, the response to dual addition of nutrients was far stronger than to single nutrient additions.



**Figure 1** Responses of autotrophs to single enrichment of N or P or to combined N + P enrichment in terrestrial, freshwater and marine ecosystems. Data are given as natural-log transformed response ratios (RR<sub>x</sub>) in which autotroph biomass or production in the enriched treatment is divided by its value in the control treatment and then ln-transformed. Error bars indicate + or – one standard error. Redrawn from Elser et al. (2007) (Figure 1)

Almost simultaneously with the review by Elser and co-authors, a review appeared with P limitation in freshwaters as its main focus (Lewis and Wurtsbaugh, 2008). The authors argue that the paradigm of P as the main limiting element for algal growth has led to ignoring flaws in the evidence which support this paradigm and overlooking of contrasting results which suggest co-limitation or N limitation of photosynthesis in freshwaters.

Schindler (1977) uses the strong relation between tot P and Chl-a as a key argument for P-limitation. Relations between totN and Chl-a are usually much poorer. Lewis and Wurtsbaugh (2008) argue that phytoplankton biomass needs P as a constituent and therefore, it is logical that the variables are related. They further point out that the relationships – as documented for instance by Dillon and Rigler (1974) - have large uncertainty intervals and therefore low predictive value. Additionally, it is not always clear how much of total N and total P is bioavailable. A larger part of tot-N may be refractory than is common for P, which might explain the relatively poor relation between Chl-a and N compared with Chl-a and P.

Sterner (2008) maintained that totN:totP ratios are valuable proxies of relative availability of these elements despite the various forms with varying bioavailability. Referring to Guildford and Hecky (2000) he showed that oligotrophic lakes have larger totN:totP ratios than eutrophic lakes. Guildford and Hecky (2000) concluded that at a mass ratio of totN:totP below 9, P deficiency occurred. At a totN:totP over 22, N deficiency occurred and in between co-limitation was the case. Sterner interpreted the same data differently. According to Stern (2008), the data show that at totP below  $10\mu\text{g L}^{-1}$ , lakes are P limited, whereas at totP over  $1\text{ mg L}^{-1}$ , lakes are N limited. Stern suggests that oligotrophic lakes are P-limited, while eutrophic lakes are N-limited. Lewis and Wurtsbaugh (2008) in their review conclude, in contrast to Stern (2008), that N and P limitation in unpolluted lakes is equally likely. An analysis of enrichment studies suggested more frequent N-limitation at low lake totN:totP ratios ( $\leq 14$ ) and high totP concentrations ( $>30\mu\text{g L}^{-1}$ ) (Downing and McCauley, 1992).

## 2.4. Concluding

All three papers that critically review the P limitation paradigm (Elser et al., 2007; Lewis and Wurtsbaugh, 2008; Sterner, 2008) agree that there is compelling evidence that phytoplankton productivity can be limited both by N and P. They also agree that the mechanism proposed by Schindler of full compensatory N-fixation in P-limited freshwater ecosystems does not seem to be generally valid. Especially in oligotrophic lakes, the community shift of algal species to N-fixers under nutrient enrichment is not very well documented.

Stern (2008) concludes by reconciling the evidence for dual nutrient (or multi-resource, when also including micronutrients) limitation with the ‘P-limitation paradigm’ by pointing at the different time scales at which limitations operate.

*“I conclude that at time and space scales relevant to population growth, multi-resource control is the rule, not the exception and this view should at least be part of the paradigm of nutrient limitation of lakes. Primary producers in lakes are not regulated on day-to-day time scales by solely or even mainly by P. I further believe that though there is logical reasoning behind it, the paradigm for phosphorus limitation of whole lakes at multi-annual scales needs further examination, particularly in reference to eutrophic and other habitats. Many years after the carbon vs. phosphorus controversy was successfully resolved there still are relevant and interesting unanswered questions about limiting factors at different scales in freshwaters.”*  
(Sterner, 2008, p 442)

### 3. Nitrogen enrichment effects on lakes: paleolimnological evidence

Lake sediment records are useful tools to study temporal developments in algal communities in relation to potential drivers of change such as climate, deposition and nutrient availability. Paleolimnological studies aim to reconstruct former environments of inland waters by investigating sediment cores, including biological records in addition to chemical, physical and mineralogical data. Either by correlations or based on an understanding of environmental sensitivities of biological communities, historical changes in chemical and/or physical conditions in lakes can be inferred. A classical example is the reconstruction of lake acidification in northern regions in relation to atmospheric long-range transported acidifying deposition (Battarbee, 1984).

Most paleolimnological papers on possible effects of N enrichment on phytoplankton communities in oligotrophic lakes are rather recent. The overview in Table 1 shows only studies published from 2000 and onwards. The studies include mainly oligotrophic temperate, alpine or arctic lakes from Europe and North-America with changes in community structures that are in varying degrees attributed to N enrichment, climate or to interactions of these. In addition, there is one meta-analysis focusing specifically on abundance of certain diatom taxa in arctic, alpine and temperate lakes. Guiding hypotheses in some papers are related to climate, in others related to N deposition. Only a few papers address both hypotheses quantitatively, (i.e. Hobbs et al., 2010; Pla et al., 2009; Ruhland et al., 2008).

Sediments in alpine and arctic lakes receiving low ( $<5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ; mostly alpine lakes) to very low ( $<1 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ; arctic lakes) levels of N deposition show shifts in diatom and/or chrysophyte communities after the onset of the 20<sup>th</sup> century (most references in Table 1). The most conspicuous changes happened usually post-1950 or even more recently, during the 1990s (i.e. Holmgren et al., 2009). Where data on  $^{15}\text{N}$  isotopes were collected additionally (Baron et al., 2000; Holmgren et al., 2009; Pla et al., 2009; Wolfe et al., 2001; Wolfe et al., 2006), depletion of  $^{15}\text{N}$  was a consistent feature which was interpreted as enrichment of the sediments by reactive N from atmospheric sources. Depletion of  $^{15}\text{N}$  in organic matter in lake sediments with increasing N deposition was also found in 12 lakes in the UK (Curtis and Simpson, 2007).

Higher productivity was observed both in arctic and in alpine lakes either by measuring higher concentrations of taxa in later periods or by measuring higher organic matter accumulation. Changes in planktonic size were noted – smaller chrysophyte cysts in a Scottish lake (Pla et al., 2009) and larger diatoms in the Beartooth Mountains (Saros et al., 2003). In the Scottish lake, the reduced planktonic cell size was suggested to be related to enhanced P-limitation due to N enrichment from atmospheric deposition: smaller cells with a larger area to volume ratio can compete more effectively for P. Saros et al. (2003) explained the larger cell size as a sign of less fierce competition for nutrients in an N-enriched lake.

For lakes in Svalbard, observed community changes were attributed to climate warming while arguing against N enrichment effects on the grounds of lacking evidence of any responses or impacts of atmospheric N (Birks et al., 2004). For mountain lakes in Europe, community shifts and higher productivity were also attributed to climate warming although nutrient enrichment was mentioned as a confounding factor (Battarbee et al., 2002). The data were not analysed systematically to test the effect of N enrichment, however. A follow-up study of the lake study of Svalbard showed signs of  $^{15}\text{N}$  depletion in addition to drastic changes in diatoms and chrysophyte taxa (Holmgren et al., 2009). The authors raised the question if current climate change is large enough to explain the recent changes. They showed that earlier climate changes in the Holocene were associated with community shifts, but that current climate change appeared to be too little to explain the recently observed drastic shifts. They concluded that the community shifts and increased production were probably related to a combined effect of climate warming and nutrient enrichment from atmospheric N pollution. The conclusions are similar to those reached for a Scottish lake in mountain conditions (Pla et al., 2009),

where both climate and atmospheric pollutants are considered as explaining variables for diatoms and chrysophyte community changes. The shifts in the diatom community were better correlated with atmospheric pollutants (observed in the form of spherical carbonaceous particles in the sediment record) while the chrysophyte community shifts were explained by a combination of climate and pollutants. Alpine lake sediments in the Colorado Front Range showed shifts from oligotrophic to more mesotrophic diatom taxa, which were interpreted as primarily related to N deposition (Baron et al., 2000; Wolfe et al., 2001). Arctic lakes in Eastern Canada showed similar changes as observed in the Colorado Front Range but here they were interpreted as a combined effect of increased N availability and climate change (Wolfe et al., 2006). A meta-analysis of diatom records in 200 lakes in Europe and North America showed community shifts in arctic lakes around 1850 and in temperate lakes in 1970. This was attributed to climate warming. For one lake, diatom community shifts were quantitatively correlated to N deposition, but no significant relations were found (Ruhland et al., 2008).

In a synthesis of dated sediment diatom records from 52 lakes in arctic, alpine and forested temperate regions, relations between diatom species turnover and climate variables, lake characteristics and deposition was tested statistically. The ecological relations found were complex but for arctic lakes, rapid temperature changes seemed to control diatom species turnover. For temperate lakes, N deposition appeared to be the controlling factors (Hobbs et al., 2010).

Concluding, there is a fair amount of qualitative and statistical evidence from paleolimnological studies that indicate effects of N enrichment on algal productivity and communities in oligotrophic lakes, at levels of N deposition as low as  $<1$  to  $3 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , but also in lakes receiving  $>10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . The relations between N enrichments and community changes are not present in all investigated lakes, and some changes in diatom records are interpreted as climate effects rather than N enrichment effects. However, the evidence from paleolimnological studies is sufficient to conclude that atmospheric N deposition can affect phytoplankton communities in arctic, alpine and temperate lakes.

Table 1 Responses to increased N supply from paleolimnological studies in remote nutrient-poor lakes. N deposition in kg N ha<sup>-1</sup> yr<sup>-1</sup>

Reference	Sites	N deposition ecosystem type	Data/method.	Response	Interpretation
Baron ea 2000; Wolfe ea 2001	USA, Colorado. Rocky Mountain National Park. Sky Pond, Lake Louise	3-5; Oligotrophic alpine lakes 3000 m a.s.l.	Ca 200 yr records. Diatoms. N deposition. Stable N isotopes. No climate data. Qualitative	Community shift after 1900 from oligotrophic towards more mesotrophic species. Higher algal production; Depletion of <sup>15</sup> N	Response to N deposition; Enrichment with anthropogenic N from atmosphere
Battarbee ea 2002	7 lakes in Europe (Norway, Austria, Slovakia, Spanish Pyrenees, Finland, Switzerland, Slovenia)	Varying, low (<2) to medium (>10)*; remote alpine lakes; oligotrophic to mesotrophic	200 yr records. Diatoms, chrysophytes, Cladocera and chironomids; C and N. Climate, no deposition. Correlations.	Various lake-dependent responses. Planktonic diatom assemblages correlated with temperature at most sites. Shifts in community structure not studied explicitly.	Climate change; nutrient enrichment and acidification mentioned as confounding factor but not analysed separately
Saros ea 2003	USA, Wyoming/Montana, Rocky Mountains, Beartooth Mountain Range, 4 lakes	1-1.5; Oligotrophic alpine lakes	400 yr record. Diatoms. No climate or deposition. Qualitative.	Shift in community from alpine diatom flora pre-1990 to dominance other species post-1995. Larger taxa, more typical for mesotrophic conditions. Increased in-lake production.	Shifts related to increased N loadings, longer ice-free periods and changed thermal stratification
Birks ea 2004	Arctic Europe, Svalbard, 23 lakes	<1 ; Arctic, mostly oligotrophic lakes	500-1400 yr records. Diatoms; cysts; chironomids; organic matter accumulation. Statistics.	Large community shifts in the last 50-100 yrs; increased OM accumulation	Climate change, no indication of response to N enrichment.
Wolfe ea 2006	Eastern Canadian Arctic, Baffin Island, 2 lakes	<1; Arctic, oligotrophic lakes	160 yr records. diatoms, stable N isotopes. No climate. PCA statistics.	Pronounced community shifts after 1950; depletion of <sup>15</sup> N, increases in organic matter	OM accumulation related to N enrichment, superimposed on community shifts that relate to climate
Ruhland ea 2008	200 lakes in North-America and Europe	varying, low (<5) to very low (<1)*; nonenriched; nonacidified; arctic, alpine, temperate lakes	Ca 200 yr records. Planktonic and benthic diatoms. Climate, no deposition. Correlational statistics.	Meta-analysis of taxon-specific shifts: Cyclotella_Aulacoseira-Fragilaria species. Timing and abundance. Changes: arctic - 1850; temperate: 1970	Shifts are response to climate change (lake-ice cover). Possible effect of N deposition investigated for 1 lake only (see below)
Ruhland ea 2008	Canada, Ontario, Whitefish Bay	4*; temperate lake. Forested catchment	Ca 200 yr records. Planktonic and benthic diatoms. Climate, deposition.	No significant correlations between N deposition trends and taxon-specific trends in abundance	No effect of N deposition on diatoms
Holmgren ea 2009	Arctic Europe, Western Svalbard, 4 lakes	<1; Arctic, oligotrophic lakes	160 yr records. Diatoms, chrysophytes, stable N isotopes. Species turnover (beta diversity). No climate. Qualitative.	Community shifts after 1920; more drastic in last 2 decades. Depletion of <sup>15</sup> N. Higher concentrations of diatoms and chrysophytes	Climate change and N enrichment drive to new ecological state
Pla ea 2009	UK, North-West Scotland. Loch Coire Fionnaraich	9*; Oligotrophic lake, 236 m a.s.l.; blanket peat, heather. Not acidified.	350 yr record. Benthic diatoms; Planktonic chrysophyte cysts. Climate and deposition. Correlational statistics.	Diatoms: Shift in 19 <sup>th</sup> century from deep water productivity to shallower rock habitats. Chrysophytes: Increased dominance of smaller cysts. Depletion of <sup>15</sup> N	Diatoms (benthic) shift related to N enrichment; Chrysophytes (planktonic) shift related to interaction with warming and N enrichment. Switch from N- to P- or co-limitation?
Hobbs ea 2010	Western North America and Western Greenland, 52 lakes	0.8-2 (<50° N) to <0.5 (>50° N); Arctic (15), alpine (17) and forested (17)	Ca 400 yr records. Diatoms, diatom compositional turnover (β-diversity). Climate, deposition. PCA analysis.	β-diversity increased in arctic and mid-latitude alpine lakes after onset 20 <sup>th</sup> century	New ecological state in arctic relates to temperature changes; in mid-latitude alpine areas to N deposition. Mixed responses in-between

\*data derived from other published data, no data in paper

## 4. Nitrogen enrichment in lakes: phytoplankton and benthic algae

Different types of evidence have been produced to test nutrient controls on lake primary productivity. The whole-lake experiments by Schindler and co-workers were initiated because bio-assays had artefacts and limitations which made hypothesis testing in ecologically relevant conditions difficult. Bio-assays are usually operated on short time scales in order to avoid artificial limitation of CO<sub>2</sub> and nutrients caused by lacking interaction with the lake ecosystem. An intermediate type of experiment in terms of ecological realism and costs are in-situ mesocosm studies, which allow exchange with the atmosphere and sometimes, in shallow lakes, exchange with the sediments. A whole different type of data is produced by lake surveys along various gradients (deposition, climate, vegetation cover) where insights in nutrients are produced from correlations between water chemistry and indicators of algal growth. Also, element ratios in organisms and in water (stoichiometry) are used as indicators of nutrient limitation. Studies on effects of N-enrichment on phytoplankton and benthic algae in oligotrophic lakes are presented in Table 2 to Table 5.

### Whole-lake experiments (Table 2)

Few whole-lake experiments specifically designed to test hypotheses on nutrient controls on phytoplankton productivity have been conducted. The whole-lake experiments in the experimental lakes area (ELA) in Canada involve combined N and P additions and single nutrient additions to oligotrophic lakes (Fee, 1979; Schindler, 1977; Schindler et al., 2008). Schindler's papers only reported results from the N+P and single P additions, while Fee also reported from single N additions. The question of single nutrient versus dual nutrient controls on primary productivity was debated in the beginning of the 1980s (Fee, 1980; Schindler, 1980). In this debate, the P-limitation hypothesis emerged as the dominating view.

The longest ongoing whole-lake eutrophication experiment is in lake 227 in ELA, where dual NP and single P additions were done (Schindler et al., 2008). In lake 227, total P and total N concentrations were raised from 6-10 to on average 42 µg P/L, respectively 0.3-0.5 to 0.8-1.2 mg N/L. Mass ratios of totN to totP in the lake varied around 22. Phytoplankton biomass increased after increasing the nutrient loads, but N-fixers became abundant only after the N:P ratio in the nutrient addition was reduced from 14 to 5. The totN:totP ratio in the lake was relatively constant even after N additions were stopped because blue-green algae fixed significant amounts of N from the atmosphere. It was concluded that reduced mineral inputs of N were compensated for by algal activity. In other words, there are compensatory mechanisms that avoid C and N limitation on freshwater productivity in the long run so that P is the element that ultimately controls plant growth in freshwaters. The data presented in Schindler et al. (2008) were interpreted differently by two independent authors (Scott and McCarthy, 2010). Instead of a constant totN:totP ratio as indicated by Schindler et al. (2008), they found a decline in totN:totP caused by declining totN concentrations after the N addition to the lake had ceased. They also found a decline in phytoplankton biomass and concluded, contrary to Schindler, that N-fixation was not enough to compensate for declining external N inputs, and that primary production was progressively limited by N in lake 227.

A whole-lake nutrient addition experiment in humic lakes in Northern Sweden (Jansson et al., 2001) showed that P addition had no effect on phytoplankton while N addition increased productivity. The authors concluded that there was a large pool of bioavailable P in humus-rich lakes, and that growth of bacterio- and phytoplankton was limited by access to inorganic N. Another whole-lake nutrient addition experiment in Northern California was not designed to test nutrient controls on primary productivity and lacked a reference treatment (Axler and Reuter, 1996). However, bioassays indicated some N-limitation of phytoplankton. The experiment demonstrated that benthic periphyton can compete with phytoplankton for available nutrients in the water column, and that N uptake in plankton and periphyton is rapid.

Summarizing, evidence from whole-lake nutrient addition experiments cannot be used to falsify the hypothesis of N-limitation of primary productivity, and is in some cases supportive of N-limitation or co-limitation of primary productivity.

Table 2 Whole-lake nutrient addition experiments

Reference	Site description	Manipulation	Lake chemistry	Response	Interpretation
Schindler, 1977	Canada, Ontario, Experimental Lake Area. Shallow, oligotrophic lakes N deposition ca 4 kg N ha <sup>-1</sup> yr <sup>-1</sup>	N:P mass ratio in fertilizer: Lake 227 N:P 14:1 – N in excess ('69-'74). N:P 5:1 – N deficient ('75 start) Lake 226 N:P 5:1. ('73-'75)	Fertilized and unfertilized lakes: Range totP: 6-40 µg/L, range totN: 0.3-1.5 mg /L.	Lake 227: '69-'74: green algae dominate, no N-fixers. '75: N-fixing algae dominate Lake 226: blue-green N-fixing algae dominate.	Short-term limitation by N is possible, but long-term (evolutionary) mechanisms work towards correction of N-deficiency. Controls to limit N might favour N-fixing algae, leading to toxic blooms. Eutrophication is controlled by P-supply
Fee, 1979	as above 20 experimental lakes. 1974-1976.	Various fertilizations: +NP, +P, +N. Also unfertilized lakes.	Annual means of nutrient loading and Chl-a.	Higher Chl-a with N+P additions than with N or P addition alone	Both N and P in short supply for phytoplankton production.
Schindler ea, 2008	as above Lake 227.	N:P mass ratio in fertilizer: Lake 227: '69-'74, N:P 12:1 – N in excess. '75-'89, N:P 4:1. N deficient. '90 onwards, only P addition.	tot-P: 20-80 µg/L, mean 42 µg/L. totN - before '79 <1 mg/L. '80-95 1.2 mg/L. 96-decline towards .8 mg/L. mean tot N:P mass ratios >20 (no N limitation) TotN and totN:totP ratio decreased after 1990 when only P was added to lake	'69-'74, see above. '75-'89: N-fixing algae dominate. '90+: N-fixers dominated phytoplankton biomass. Chl-a to biomass ratio lower when N was deficient. Chl-a decreased. Phytoplankton biomass decreased from '97-2005.	Significant N-fixation by cyanobacteria when N-additions decreased, to keep stable totP tot totN ratios.  Eutrophication is controlled by P-supply
Scott and McCarthy, 2010	As above. Lake 227.	Reinterpretation of data presented in Schindler ea 2008			Progressive N-limitation in lake 227. N-fixation not sufficient to compensate for external N inputs. Eutrophication can be controlled by managing N and P inputs.
Schindler 1977	as above Lake 261	Only P fertilizer, 3 yrs.	TotP 15-20 µg/L. totN 0.5 mg/L	No N-fixers in phytoplankton. Luxuriant growth of periphyton	Low N:P ratios stimulate N-fixing cyanobacteria.
Jansson ea 2001	Northern Sweden, humic, oligotrophic, shallow lakes. Low (<2?*) N deposition	Reference lake. P addition: + 10 µg/L P N addition: +100 µg/L N	Pretreatment: totN:totP mass ratio 19-21 TotN 0.44-0.53 mg/L TotP: 21-30µg/L DIN ≤ 10 µg/L.	P addition had no effect on bacterio- or phytoplankton. Phytoplankton stimulated by N addition.	Pool of bioavailable P in DOC-rich lakes. Growth of phyto- and bacterioplankton limited by inorganic N.
Axl and Reuter 1996	Northern California, Castle Lake. Meso-oligotrophic alpine lake. Large littoral zone.	'Before-after' experiment. Fertilizer: NH <sub>4</sub> NO <sub>3</sub> addition to epilimnion. No reference lake. Parallel mesocosm experiments. Bioassays.	TotP ~ 5 µg/L. After fertilizer addition: DIN to ca 80 µg/L, decline to <5 µg/L	Most N uptake by periphyton, 10-30% by phytoplankton. Bioassays: no consistent sign of P-limitation phytoplankton. Additions of Mo increased productivity	No particular focus on nutrient deficiency. Phytoplankton N-deficient. Periphyton might outcompete phytoplankton for nutrients in water column. Effects on Chl-a or phytoplankton biomass not reported.

### Regional surveys Table 3

A survey of European lakes including dystrophic to eutrophic water bodies indicated that Chl-a was positively related to both totN and totP, where totP was the best predictor in most lakes except humic lakes. Here, Chl-a was related best to totN (Phillips et al., 2008). They conclude that these humic lakes are more likely to be N-limited than P-limited.

Surveys covering oligotrophic lakes in Europe and North America (Bergstrom and Jansson, 2006), Sweden (Bergstrom et al., 2005) were used to test whether higher N supply lead to higher Chl-a to tot P ratios. They hypothesized that in N-limited lakes, a higher supply of N would lead to a higher ratio

of Chl-a to totP. Both studies found higher dissolved inorganic N concentrations where N deposition was higher, and higher Chl-a to tot P ratios in regions with higher N deposition. The Chl-a to tot P ratio almost doubled from along the gradient of 2 to 5 kg N ha<sup>-1</sup> yr<sup>-1</sup>. The authors concluded that N deposition had contributed to higher phytoplankton biomass in oligotrophic lakes, i.e. eutrophication. Also, they argued that it was likely that N deposition in oligotrophic lakes has caused a shift from naturally N-limited lakes to P-limited lakes.

Lakes in Sweden and Norway had higher totN to totP ratios in areas with higher N deposition (Elser et al., 2009). Nutrient enrichment bioassays in 6 lakes in Sweden, Norway and Colorado (USA) showed that phytoplankton was consistently N-limited in low N deposition lakes, while high N-deposition lakes showed P-limitation (Elser et al., 2009). At totN:totP < 20 (mass ratio), lakes were generally N limited. At totN:totP > 50, lakes were generally P-limited. Deposition categories 'high' and 'low' were relative and different for lakes in different countries.

In four acid-sensitive lakes in Minnesota, nutrient limitation of phytoplankton was studied by nutrient bioassays, physiological assays and nutrient deficiency indices (Axler and Reuter, 1996). All evidence indicated a growth response to N although N+P additions yielded the largest effect. Deposition of N at the lakes was sufficient to meet daily algal N demand and it was concluded that N supply through N deposition had increased the fertility of the lakes.

Other surveys (i.e. lakes in the UK (Curtis and Simpson, 2007; Maberly et al., 2002); arctic lakes in Canada (Ogbebo et al., 2009)) analysed element ratios in water and related these to ratios developed to evaluate nutrient deficiencies in algal growth (Smith, 1982). All of the studied arctic lakes in Canada were severely P-deficient (i.e. above the NP mass ratio of 17) according to these criteria. In the UK, co-limitation was most common. Nutrient assays for the arctic lakes indicated co-limitation rather than P-limitation (Ogbebo et al., 2009). The use of previously established fixed criteria to assess nutrient deficiencies is criticized by Maberly and co-workers, as total element concentrations are not the same as bioavailable concentrations. In four acid-sensitive lakes in Minnesota (Axler et al., 1994), totN to totP ratios were not consistent with growth responses from bioassays, in contrast to DIN to totP ratios. Similarly, lake TN:TP ratios and physiological bioassays assessing nutrient deficiency in temperate lakes in British Columbia (Davies et al., 2004) indicated P limitation and co-limitation, respectively. Using totN to totP ratios for assessing nutrient deficiencies may underestimate the extent of N limitation, alternatively overestimate P limitation, when most nitrogen is not bioavailable.

Thus, surveys, in combination with nutrient bioassays, indicate that N-limitation and/or co-limitation of algal growth is common, especially in oligotrophic lakes. Several studies conclude that N deposition has increased lake primary productivity.

Table 3 Phytoplankton responses increased N supply in regional surveys. N deposition in kg N ha<sup>-1</sup> yr<sup>-1</sup>

Reference	Site	Data	Response	Interpretation
Axler ea 1994	USA, Northern Minnesota. 4 acid-sensitive lakes. Forest. Oligotrophic to mesotrophic N deposition 5-9*	Phytoplankton, chl-a; water chemistry. Bioassays (+N, +P, +NP)	No N-fixing algae. Growth responses to all enrichments. DIN:totP agreed with growth responses. N deposition high enough to meet daily algal N demand	N deposition has increased primary productivity in lakes.
Maberly ea 2002; Curtis and Simpson, 2007	UK, 43 upland lakes from 6 regions along an N- and P-limitation gradient	Phytoplankton, chl-a; water chemistry. Bioassays (+N, +P, +NP)	N, P and co-limitation found in bioassays. N limitation at DIN <90µg/L and DIN:TDP <24 (mass ratio). P limitation at DIN>0.180µg/L and DIN/TDP>113	Seasonal changes in nutrient limitation. Colimitation more common as summer progresses. Effect of N deposition dependent on N retention of catchment.
Davies ea 2004	Canada, British Columbia. 10 temperate lakes and reservoirs.	Element ratios in lake water and seston. Bioassays.	TN:TP ratios indicate P limitation. Physiological bioassays indicate co-limitation.	Co-deficiency is common.
Bergström ea 2005	Sweden, unproductive lakes from 10 regions. N deposition .1-15	225 lakes with Chl-a data; 3907 lakes with water chemistry.	DIN, resp DIN:totP, resp Chl-a: tot P positively correlated with N deposition.	N deposition increases productivity of oligotrophic lakes. Clearest effect at N deposition < 5
Bergström and Jansson 2006	Europe and North America. 4295 unproductive lakes in 42 regions N deposition .1-14	Chl-a; water chemistry	DIN, resp DIN:totP, resp Chl-a: tot P positively correlated with N deposition.	N deposition has contributed to higher phytoplankton biomass in unproductive lakes, and has caused shift from natural N limitation to P limitation. Clearest effect at N deposition < 5
Bergström ea 2008	Sweden, 12 unproductive lakes in 4 regions along north-south gradient. N deposition <1-10	Chl-a; water chemistry. Bioassays (+N, +P, + NP)	DIN, resp DIN:totP, resp Chl-a: tot P positively correlated with N deposition. N limitation common during summer	P-limitation is induced by N deposition.
Phillips ea 2008	Europe, >1000 lakes. Large variation in climate, alkalinity, humic content, depth. All trophic states.	Chl-a, totP, totN.		Chl-a related to totN and totP. totP best predictor, except for humic lakes where totN is best predictor.
Elser ea 2009	Sweden, 1668 lakes. Norway, 385 lakes. In high and low N deposition areas (high: 6-8; low: 2-5)	Water chemistry in >2000 lakes in Sweden and Norway. Bioassays (+N, +P, + NP) in lakes in Colorado, Sweden, Norway (90 experiments)	TotN:totP positively related to N deposition, due to higher TN. Below TN:TP 20 (mass ratio), N limitation.	At low N deposition, phytoplankton N limited. High N deposition, P limitation.
Ogbebo ea 2009	Canada, North West territories. 9 oligotrophic, arctic lakes.	Chl-a; water chemistry; element ratios phytoplankton. Bioassays (+N, +P, +NP)	NP additions give highest response, small effects of single additions. Lake water and seston TN:TP ratios indicate P limitation. Nitrate below detection limit.	Bioassays indicate co-limitation. TN:TP ratios of lake water indicate P limitation.

### Benthic algae Table 4

Pelagic phytoplankton is not the sole contributor to lake productivity. Especially in shallow lakes, productivity of benthic algae in the littoral zone can be significant and could be affected by increased N supply from N deposition. Increased nutrient loads have repeatedly been associated with shifts in the benthic algae communities from a dominance of periphyton to filamentous green algae. Such shifts could affect secondary littoral productivity negatively, as many filamentous green algae are assumed to be inedible for macroinvertebrates (Sandjensen et al., 1989; Thiebaut and Muller, 1999).

A similar survey as presented by Bergstrom and co-workers for phytoplankton (2005) was done for epilithic periphyton in shallow lakes in Sweden along a N deposition gradient (Liess et al., 2009). Periphyton is a complex mixture of algae, cyanobacteria, heterotrophic microbes, and detritus that is attached to submerged surfaces in most aquatic ecosystems. Periphyton mats on stones and rocks are

called epilithon and are formed by benthic algae together with detritus, bacteria and eukaryotic microorganisms. The productivity of periphyton mats is limited by light, the supply of nutrients and by grazing. Element ratios and abundance of cyanobacteria in periphyton mats were investigated in unproductive lakes in north and south Sweden. Lake total N concentrations and tot N: P ratios were highest where N deposition was highest, in south Sweden. N deposition was weakly positively related to epilithic N:P ratios. Epilithic C:P and N:P ratios were higher in the southern than in the northern lakes, which was interpreted as P being more limiting in the south than in the north. Epilithic N:P ratios in the south indicated P-limitation, contrary to the northern lakes where N-limitation was more common. Benthic algal composition in the north had a higher contribution from N-fixing cyanobacteria than the south. The ratio between Chl-a and totN was positive and significant in the north and the south (A. Liess, pers. comm.). The results were interpreted as in rough agreement with the Swedish study on phytoplankton (Bergstrom et al., 2005). The higher proportion of N-fixing cyanobacteria (although only consisting of 5% of the algal biovolume) found where N-deposition was lowest – thus to some extent compensating for N-limitation – appears to support Schindler's suggested mechanism of compensatory N-fixation where N supply is deficient (Schindler, 1977). However, the low absolute volume of these N-fixing micro-organisms ruled out a significant contribution to N availability.

Nutrient limitation in periphyton was also studied in the UK, using bioassays and evaluation of element ratios (Maberly et al., 2002). The *in-situ* bioassays for periphyton gave fewer responses than bioassays for phytoplankton, possibly due to interaction with sediments or to intense grazing. However, co-limitation was the most common response. In Australian streams from catchments with various land use, N limitation of periphytic algae was frequently found (Chessman et al., 1992).

Table 4 Responses of benthic algae to increased N supply. N deposition in kg N ha<sup>-1</sup> yr<sup>-1</sup>

Reference	Site	Data	Response	Interpretation
Chessman et al 1992	Australia, Victoria, streams draining subalpine, forested, agricultural, urban catchments	Nutrient diffusion strata for assessing limiting nutrients periphyton; Chl-a	Chl-a density related to N access	Periphytic algae in streams limited by N
Maberly et al 2002	UK, 30 upland lakes from 6 regions	Periphyton; water chemistry. Yield limitation of periphyton growth (+N, +P, +NP)	No response to nutrient additions in 40% of tests. Otherwise co-limitation most common. Lack of response due to light limitation or grazing pressure?	Periphyton less responsive to nutrient addition than phytoplankton. Co-limitation common.
Liess et al 2009	6 unproductive lakes in middle, and 6 in south Sweden N deposition 2-12	Epilithon, water chemistry	Lake water totN and totN:P positively related to N deposition. Epilithic N:P lower in north; proportion of cyanobacteria higher in north	More N limited in north, more P limited in south. N deposition might intensify P-limitation of epilithic algae.

### Mesocosm experiments Table 5

Nutrient enrichment enclosure experiments in oligotrophic shallow alpine lakes in the Rocky Mountains (USA) were done with a focus on phytoplankton and zooplankton (Lafrancois et al., 2004) and phytoplankton and epilithon (Nydick et al., 2004). In two lakes in Wyoming, phytoplankton responded strongly to N and NP additions but not to P additions. Biomass (Chl-a and cell density) and photosynthesis increased, while the community structure shifted from chrysophytes dominance towards more cyanophytes, chlorophytes and diatoms. The latter taxa are known to have higher N optima than the original chrysophyte-dominated community. Benthic algae on tiles responded also to N and NP additions, while benthic algae on sediments did not respond. Biomass of benthic algae dominated biomass of phytoplankton, which is the reason why total lake productivity did not increase significantly as a response to nutrient additions. A second nutrient enrichment enclosure experiment involved one of the lakes used in Nydeck et al (2004), in addition to a shallow alpine lake in Colorado (Lafrancois et al., 2004). An additional factor of acid addition was included in the experimental design. The lake in Colorado had considerably higher nitrate concentrations than the lake in Wyoming. Phytoplankton biomass in the low-nitrate lake responded strongly to N and N+P additions while biomass in the high nitrate lake only responded to combined N and P additions. No responses to P

alone were observed. Shifts in phytoplankton taxa were both related to increased nutrient availability and lower pH.

To test the effect of N:P ratios – with P constant, and N varying - on algal production and composition, mesocosms were placed in pelagic and littoral at ELA (Levine and Schindler, 1999). N-deficiency was detected in all treatments at N:P ratios below 17. Phytoplankton biomass was highest at high N:P ratios while periphyton biomass was stimulated by increased inorganic N availability at low N:P ratios. It was concluded that low N:P ratios, or N-limitation of phytoplankton, stimulated cyanobacterial growth and that increased N-availability could increase productivity of benthic algae at the sediment surface.

Table 5 Phytoplankton and periphyton responses to increased N supply from mesocosm-studies. N deposition in kg N ha<sup>-1</sup> yr<sup>-1</sup>

Reference	Site	Manipulation	Data	Response	Interpretation
Levine and Schindler 1999	Canada, Experimental lake area. Oligotrophic lakes.	Mesocosm in-situ; pelagic and littoral; +NP (NP 8; NP 50)	Phytoplankton, periphyton. Water chemistry; Chl-a; C-fixation. species.	At low N:P ratio N limitation; N <sub>2</sub> -fixing bacteria dominate only at low N:P in pelagic; Periphyton biomass increased with higher DIN at low N:P. Phytoplankton biomass larger at high N:P	Low N:P may encourage N <sub>2</sub> fixers or increase productivity at sediment surface where more N is available.
Nydick et al 2004	USA, Wyoming, 2 shallow oligotrophic alpine lakes. N deposition 3	Enclosures, in situ, to lake bottom. +P, +N, +NP (10:1)	Phytoplankton, epilithon, epipelon. Chl-a; water chemistry; Species	Phytoplankton growth response to N and NP additions. Shift from chrysophytes towards cyanophytes, chlorophytes, diatoms. Epilithon: Late summer, algae on tiles responded to more N and N+P. Epipelon: Algae on sediments responded hardly	N controlled productivity and community structure of phytoplankton and algae on hard substrates, but not benthic community on sediments.
Lafrancois et al 2004	USA, Wyoming, 2 shallow oligotrophic alpine lakes. N deposition 3	Enclosure experiments, to lake bottom. +P, +N, +NA (pH 4.5-5) (A=acid), +NAP (NP as 33:1)	Water chemistry, phytoplankton, zooplankton. Chl-a, species composition	In low-N lake: Phytoplankton response with N and NP, NAP additions: chl-a. Changes in community (more large chlorophytes). In high N lake, few responses to N, but responses to P and NAP	Under high nitrate conditions, P-limitation and co-limitation; under low nitrate N limitation and co-limitation.
Andersson and Brunberg, 2006	Sweden, shallow oligotrophic clearwater lake.	Enclosure experiments, to lake bottom. +N, +P, +NP	Water chemistry, Chl-a, phytoplankton, microphytobenthos. Biomass, species composition. Primary production.	Biomass microphytobenthos stimulated. Phytoplankton biomass not affected. Pelagic P limited, benthic habitat N limited. Bacteria negatively affected by N addition.	Increased nutrient supply increases primary production, but not necessarily pelagic production. Benthic habitat N-limited

The results from these American nutrient enclosure experiments are in agreement with the conclusions from the surveys, that phytoplankton productivity in nutrient-poor lakes is N-limited. An exception was the nitrate-rich lake in Colorado, which only responded to dual N and P additions, but here it was argued that N deposition had driven the lake from N limitation to co-limitation by P and N.

A mesocosm study in an oligotrophic, clearwater lake in Sweden showed that the benthic habitat was N-limited, as sediments supplied enough P (Andersson and Brunberg, 2006). Also, they showed that increased nutrient supply does not necessarily increase production in the pelagic habitat but instead leads only to increased production in the benthos. The mesocosm experiments from the Experimental Lake Area also indicate that benthic algae were sensitive to N-availability (Levine and Schindler, 1999).

**Bio-assays Table 6**

A simpler methodology to study nutrient enrichment responses than mesocosms is by adding nutrients to bottles of lake water, either under controlled laboratory conditions or *in-situ*. The methodology is criticized because of reduced ecological realism and missing interactions with the environment surrounding it, especially the atmosphere and sediments. Some of the surveys reported in Table 3 included bioassays.

Table 6 Phytoplankton responses to increased N supply from bio-assays. N deposition in kg N ha<sup>-1</sup> yr<sup>-1</sup>

Reference	Sites	Manipulation	Data	response	Interpretation
Morris and Lewis, 1988	USA, Colorado. 8 alpine nutrient-poor lakes N deposition 3	In-situ; 4-7 days incubation; 5-8 times during growing season. Treatments +P, +N, +NP, ratio: 2:1	Water chemistry, phytoplankton. Chl-a, cell counts, POC. Species composition	3 lakes N limited, 1 lake P-limited, 4 lakes co-limited. Seasonal switches also observed. Best indicator of phytoplankton limitation was DIN to totP, and DIN to Tdissolved P, not totN to totP ratio	Nutrient-poor alpine lakes can be both N and P limited, and co-limitation and switches are common
Nydick et al. 2003	USA, Colorado. 3 alpine nutrient-poor lakes N deposition 3-5	In-situ, 2-L bottles, 1 week incubation. Seasonal. +P, +N, +NP (5.4:1)	Chl-a; water chemistry; Phytoplankton species; zooplankton species	Less P deficiency with higher P concentrations. Chl-a very similar despite different lake chemistry	Mostly P limiting for phytoplankton growth but not early in the season. Also co-limitation
Saros et al. 2005	USA, Rocky Mountains. 2 alpine nutrient-poor lakes N deposition 3-5	In-situ, 5 days incubation. +N, +P, +NP(1.6:1); ; also NP 18:1 enrichment	Two diatom taxa, Water chemistry, cell density, chl-a	Response to +N, and NP; dependent on taxa. Higher NP ratio gives higher response than low NP ratio	The two taxa thrive at low totP and additional N will make them more abundant.
Thompson et al. 2008	Canada, Rocky Mountains. 1 Alpine nutrient-poor lake	Laboratory. Treatment +N +temp, +(N, temp)	Plankton (phyto and zoo), water, sediment	Positive effect on phytoplankton abundance dependent on warming	Interaction between nutrient enrichment and warming at several trophic levels

An early *in-situ* bioassay tested responses to single nutrient and dual NP additions in 8 oligotrophic lakes in Colorado, and concluded that N was limiting in most lakes, alone or in combination with P (Morris and Lewis, 1988). Canadian arctic lakes were all co-limited by N and P, with little responses to single nutrient additions (Ogbebo et al., 2009). Arctic lakes in the Rocky Mountains were found to be limited by P except early in the growing season (Nydick et al., 2003). Nitrogen limitation in a Canadian arctic lake was found to be dependent on warming, in a growth chamber experiment where effects of P were not investigated (Thompson et al., 2008). Two diatom taxa in two lakes in the Rocky Mountains in Colorado responded positively to N and NP additions, where higher NP ratios resulted in the highest response. It was concluded that these two taxa thrived at low P concentrations and would become more abundant with higher N availability (Saros et al., 2005).

In Sweden, bioassays were done in oligotrophic lakes along a N deposition gradient (Bergstrom et al., 2008). More frequent responses in Chl-a to N additions than to P additions were found, and N-limitation was strongest at mass ratios of DIN to totP < 1.5. A compilation of data from oligotrophic lakes in Europe and USA shows that the DIN to totP ratio is a better indicator of N-limitation of phytoplankton than the totN to totP ratio (Bergstrom, 2010). Responses to N and P additions were always higher than to single-nutrient additions – in agreement with the review by Elser and co-workers (2007). Only in high N deposition areas, P-limitation was found in early summer when DIN concentrations were highest. As a result of the declining DIN pool over the summer, lakes switched to dual limitation by N and P, followed by N limitation. The authors argued against compensatory N-fixation, as cyanobacteria are not abundant in nutrient-poor lakes in Sweden, and even extreme N-limitation induced by P-addition in such lakes did not lead to dense populations of cyanobacteria (Jansson et al., 2001).

**Concluding**

Whole-lake experiments, mesocosms and bioassays are all accepted methods used to experimentally investigate nutrient controls on freshwater primary productivity, in declining order of ecological realism and costs. Lake surveys along various gradients (deposition, climate, vegetation cover) are also used to statistically related water chemistry and indicators of algal growth. In many studies, a combination of all, or some, of these methods are used to gain insights in nutrient controls on productivity.

Whole-lake manipulations have often been done by manipulating both N and P supply, and conflicting interpretations exist with respect to whether N-limitation has been detected. However, where only N was added increases in productivity of both phytoplankton and sediment-dwelling algae were observed, and several authors interpret results from lakes receiving both N and P as showing evidence of N-limitation in addition to P-limitation and co-limitation.

Regional surveys in boreal lakes in Europe and North America with N deposition between  $<1$  and  $>14$   $\text{kg N ha}^{-1} \text{ yr}^{-1}$  show higher chlorophyll concentrations per unit P in areas with higher N deposition, indicative of higher primary production. Experimental nutrient additions in lakes (mesocosm studies and bioassays) support the finding of the regional surveys by showing that N limitation of algal growth is common, especially under conditions of low N availability. Increased N availability can increase productivity of sediment- and rock dwelling algae (benthic algae) but here the number of studies is limited. It is suggested that N deposition may have shifted primary productivity in oligotrophic lakes from N limitation to P limitation. Also, some studies suggest that totN to totP ratios to evaluate nutrient deficiencies may underestimate N-deficiency as total N in oligotrophic lakes is often partly unavailable. Often, inorganic N to totP ratios are better predictors of responses to nutrient additions.

## 5. Nitrogen enrichment effects on macrophytes

Effects of nitrogen on macrophytes in oligotrophic waters have not been investigated over a wide range of species, ecosystem types or atmospheric loads of N. However, targeted studies were done to investigate water plants mostly in Atlantic soft clearwater lakes in Western Europe during the 1980s and 1990s, while a large body of evidence exists from eutrophic waters. More recent studies on macrophytes in oligotrophic lakes in relation to nitrogen deposition are not available. In general, N supply is assumed to affect macrophyte communities in mesotrophic and eutrophic aquatic ecosystems, calling for a dual control of P and N for restoration of water plant communities in eutrophic lakes (Gonzalez Sagrario et al., 2005; Jeppesen et al., 2007; Li et al., 2008). However, the primary aim of this report is to document effects of atmospheric N deposition on aquatic ecosystems. Therefore, this chapter focuses mostly on higher plants in oligotrophic, acid-sensitive lakes where direct waterborne loadings of agricultural N and P are notably absent.

### Macrophyte communities in oligotrophic lakes

Higher plants in oligotrophic clearwater lakes are dominated by submerged and rooted growth forms, while emergent and floating-leaved plants are far less common. The dominating plant communities, commonly denoted as isoetids (phytosociological termed *class* Littorelletea), are typically small (5-20 cm high), slow-growing water plants, with large belowground root systems. The entire plant is submerged, and mats of isoetids may occasionally cover the entire bottom of large areas down to 3-4 m depth, with the worldwide occurring fern-related pteridophyte *Isoetes* as name-giving genus. Typical widespread genera belonging to the group are *Isoetes*, *Lobelia* and *Littorella*. Light availability is critical for these bottom-dwelling plants, and they are easily outcompeted if turbidity increases. While most aquatic species take up CO<sub>2</sub> from stems and leaves, many isoetids assimilate CO<sub>2</sub> from the roots (Rorslett and Brettum, 1989), an assumed adaptation to oligotrophic acidic conditions, where water CO<sub>2</sub>-concentration is normally low. Their elaborate root systems in sandy-rocky substrate make phosphorus limitation less probable (Barko et al., 1991; Vandonk et al., 1993). Moreover, isoetids are pronounced acid-tolerant (Bobbink and Roelofs, 1995), and survive on lower nitrogen concentrations than any other macrophyte (Arts, 2002).

The occurrence of isoetid vegetation depends on the presence of oligotrophic lakes. Isoetids are common in mountainous and alpine areas, but also under other climatic conditions where the substrate is sufficiently poor in nutrients - in Europe in Scandinavian highlands, in north-west Britain's deep lakes, and in alpine lakes in Switzerland and Austria. Best investigated are probably the Atlantic soft clear-water lakes of Western Europe found in the Netherlands, Western France and Belgium. These are predominantly rainwater-fed and to some extent vulnerable to desiccation during dry summers. Located on sandy soil, they are poor in calcium carbonate and correspondingly poorly buffered (alkalinity 50-500 µeq l<sup>-1</sup>). Plant nutrients (N and P) are low under pristine conditions, and the water is slightly acidic even under natural conditions (pH 5-6).

During the 20<sup>th</sup> century, soft-water macrophytes have declined dramatically in the West European lowlands (Denmark and the Netherlands) but also in Scotland, Norway and Sweden (Arts, 2002; Brandrud, 2002; Brandrud and Roelofs, 1995; Farmer and Spence, 1986; Grahn, 1977; Roelofs, 1983; Roelofs, 2002; Vandam, 1988). Causes for this decline – acidification, alkalisation, eutrophication – are discussed in a review by Arts (2002).

### Shifts in oligotrophic macrophyte communities (Table 7)

Macrophytes in soft-water lakes have been extensively studied in the Netherlands. A survey of more than 70 lakes that still comprised a largely pristine isoetid vegetation in 1950, revealed that this vegetation type was strongly reduced or had disappeared thirty years later in 53 of the investigated lakes (Roelofs, 1983). Only twelve lakes were eutrofied by local pollution sources. The original isoetid plant community had typically changed by losing key species (*Isoetes lacustris*, *Littorella uniflora*) to biased dominance of one single species, *Juncus bulbosus* (bulbous rush) and submerged *Sphagnum* mosses. *J. bulbosus* is widely distributed and a common species in nutrient poor lakes, and

possesses several of the isoetid characteristics. It is, however, less dependent on rooting for sufficient nutrient uptake and can alter growth form and thus effectively outcompete other plants under conditions of increasing nutrient availability. Increased N availability due to atmospheric deposition of nitrogen and changes in alkalinity were assumed to have contributed to the vegetation shifts (Roelofs, 1983; Roelofs et al., 1984).

Table 7 Macrophyte responses to increased N supply. N deposition in kg N ha<sup>-1</sup> yr<sup>-1</sup>

Reference	Sites	Type of data	Observation / Response	Interpretation
Roelofs, 1983	Netherlands	Survey, time series. 70 lakes. 1950s and 1980s.	1950s: all lakes had well-developed isoetid vegetation. 1980s: 17 lakes unchanged and still low in [NO <sub>3</sub> ]. 12 lakes were eutrophied from agriculture. 41 lakes: isoetids replaced by <i>Juncus</i> and high in [NO <sub>3</sub> ]	Detrimental effects on isoetid vegetation from enrichment in N by atmospheric N deposition.
Roelofs et al 1984	Culture experiments and field data	Factorial design: pH/alkalinity; sediments; NH <sub>4</sub> . <i>Littorella uniflora</i> , <i>Juncus</i>	Experiments: shifts in CO <sub>2</sub> can cause shifts in the macrophyte community. Field data: soetids replaced by (mainly) <i>Juncus</i> and <i>Sphagnum</i> , due to acidification, or (more rare), due to phosphate sediment enrichment.	Nutrient enrichment favours <i>Juncus</i> only when CO <sub>2</sub> enrichment (due to acidification) occurs
Schuurkes et al 1986	Mesocosms in greenhouse, simulating softwater ponds.	7 treatments. Control, various combinations and levels of NH <sub>4</sub> , SO <sub>4</sub> , NO <sub>3</sub>	+NH <sub>4</sub> SO <sub>4</sub> : acidified (through nitrification) and eutrophied. +NH <sub>4</sub> gave highest biomass of <i>Littorella u.</i> +NO <sub>3</sub> favoured <i>Juncus</i> only. +SO <sub>4</sub> : no effect on vegetation	Plants are sensitive to NH <sub>4</sub> , not to acidification. <i>Juncus</i> sensitive to NO <sub>3</sub> .
Schuurkes et al 1987	Mesocosms simulating lentic oligotrophic softwater lakes in the Netherlands	Manipulation: 1. sulphuric rain with NO <sub>3</sub> (pH 5.6–3.5) addition 2. (NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> (pH 5.6) addition	Treatment 2: <i>Littorella uniflora</i> disappeared. Luxuriant growth of <i>Sphagnum cuspidatum</i> , <i>Juncus bulbosus</i> Increase of filamentous green algae. No response to treatment 1	Plants are sensitive to NH <sub>4</sub> , not to acidification. For survival of ecological community oligotrophic waters, NH <sub>4</sub> deposition no higher than 19 kg N/ha/yr
Robe and Griffiths, 1994	Culture experiments with <i>L. uniflora</i>	Treatment: 3 NO <sub>3</sub> levels (low, near-natural, high); 2 light intensities. <i>Littorella uniflora</i>	Supply of NO <sub>3</sub> failed to cause enhanced growth or photosynthesis, and indicates that <i>L. uniflora</i> is not NO <sub>3</sub> -limited in natural oligotrophic lakes.	<i>L. uniflora</i> not limited by NO <sub>3</sub> -supply in natural oligotrophic habitats due to low growth rate and strategy to avoid N-stress
Roelofs et al, 1994	Norway	Macrophyte responses to liming in 6 norwegian lakes, while using five unlimed lakes as reference.	Liming caused increased CO <sub>2</sub> -production, and mobilisation of N, P and Fe in sediments. This may explain the observed increase in <i>J. bulbosus</i> in limed lakes.	Increased CO <sub>2</sub> makes <i>J. bulbosus</i> competitive superior when oligotrophic lakes are exposed to liming.
Brandrud and Roelofs 1995	SW Norway.	Survey 1992-1994. 55 lakes SW Norway. Limed and unlimed acidic. <i>Juncus Bulbosus</i>	Limed lakes had high coverage of <i>Juncus</i> compared with reference. Water covered with surface mats. Most conspicuous in lakes furthest west.	Liming leads to high NH <sub>4</sub> and CO <sub>2</sub> in sediments. High availability of NH <sub>4</sub> favours <i>Juncus</i> over original isoetid vegetation. Mild winters may also favour <i>Juncus</i> .
Lucassen et al 1999	Mesocosms experiment in unlimed lake in Norway	Factorial design: limed/not limed sediment; acidified/reference water. <i>Juncus Bulbosus</i>	Combination of limed sediment (more NH <sub>4</sub> ) and acidified water (more CO <sub>2</sub> ) lead to large plant growth. Only more CO <sub>2</sub> did not lead to increased plant growth.	Combination of increased NH <sub>4</sub> in sediments (due to liming) and higher CO <sub>2</sub> promotes growth of <i>Juncus</i> .
Brandrud 2002	Norway and Sweden.	Review on effects of liming on broad range of limed lakes in Norway and Sweden.	Recolonisation of acid-sensitive macrophytes, but temporary massive expansion of <i>J. bulbosus</i> .	Liming leads to eutrophication of sediment and alkalisation of water, resulting in less favourable conditions for original isoetid vegetation, and increased growth of <i>J. bulbosus</i> .
Wen et al 2003	China	Field data from chinese lakes.	Comparison of oligotrophic lakes where decline or disappearance of <i>Isoetes</i> has been reported, with unaffected lakes.	Lakes where <i>Isoetes</i> has disappeared had increased concentrations of NO <sub>3</sub> , hardness, CO <sub>2</sub> and heavy metals.

Acidic deposition in the Netherlands during the 1980s was dominated by ammonium sulphate, leading to surface water acidification from leaching of  $\text{SO}_4$  and  $\text{NO}_3$  and to nutrient enrichment with  $\text{NH}_4$  (Vanbreemen and Vandijk, 1988). This made it difficult to differentiate between the possible causes of macrophyte deterioration (Roelofs, 1983). In a mesocosm study, macrophyte responses were studied in an acidification treatment and a  $(\text{NH}_4)_2\text{SO}_4$  addition treatment (Schuurkes et al., 1987). No changes in macrophyte vegetation were found under the acidification treatment while in the other treatment the sensitive isoetid species disappeared and luxuriant growth of *Spagnum* and *Juncus* were observed, in addition to an increase in filamentous algae. It was concluded that the observed macrophyte vegetation shifts in the Netherlands were primarily related to nutrient enrichment. The increasingly dominating *Juncus bulbosus* was found to be more nitrophilous than common isoetids, with an ammonium-dominated nitrogen-utilisation, with leaves as major uptake sites for both nutrients and  $\text{CO}_2$  (Reuss et al., 1987; Roelofs et al., 2002). Moreover, this species, which in many localities is reported to become dominant, is shown to have a low photosynthetic compensation point, allowing it to grow and thrive in rather dimmed light and at larger depth than its competitors (Svedang, 1990). Such ecological features may explain the extensive growth of *Juncus bulbosus* in many acidified soft clear-water lakes. Schuurkes et al. (1987) pointed out that extensive growth of filamentous, mucous-rich green algae, where detritus-particles accumulate, has negative implications for macrophyte photosynthesis and can lead to deterioration of the original vegetation. Based on field observations of macrophytes, atmospheric N loads and the experimental studies in the mesocosms, Bobbink and Roelofs (1995) estimated an atmospheric nitrogen load of 5-10 kg N ha<sup>-1</sup> yr<sup>-1</sup> as critical for the typical isoetid vegetation.

The results from the mesocosm studies from Schuurkes and coworkers are supported by other studies. Most isoetids, though generally acid tolerant, are apparently adapted to nitrogen poor conditions, and predominantly utilize nitrate, with roots as the major uptake site. Treatment of the isoetids *Luronium natans* and *Littorella uniflora* (shoreweed) with ammonium-enriched water led initially to improved growth, but secondary to reduced flowering, increased fungal infections, withering and declined plant density (Robe and Griffiths, 1994; Vanbreemen and Vandijk, 1988). Wen et al (2003) observed reduced occurrence of Chinese *Isoetes sinensis* associated with increased concentrations of  $\text{CO}_2$  and  $\text{NO}_3$ . Arts (2002) suggests that eutrophication in deep lakes lead to increased phytoplankton and chlorophyll concentrations, implying enhanced turbidity and upcoming leaf floating vegetation (nymphoids) in shallow areas, which may cause shading effects on the isoetids at the bottom sediments, or overgrowth by larger macrophytes or epiphytic algae. Such increased dominance of leaf-floating species also banishes the original isoetids from deeper to more shallow zones, which are prone to desiccation during the summer periods (ibid).

Reduced distribution of the isoetid *Littorella uniflora* in Denmark has also been associated with eutrophication, but without accounting for the possible contribution from atmospheric deposition. Historical and recent data on distribution of *Littorella uniflora* from 472 lakes were studied. In almost 50% of the lakes (218 lakes), the macrophyte had disappeared. Secondary effects of eutrophication, associated with reduced water transparency, increased phytoplankton dominance, overgrowth by epiphytes and increased competition for light by growth of elodeids were thought of as possible factors responsible for the reduction of the isoetid (Pedersen et al., 2006).

Roelofs et al. (1994) recorded massive invasions of *Juncus bulbosus* and disappearance of the original isoetid vegetation in alpine Norwegian lakes after liming. Liming of lakes is a common restoration tool to improve water chemistry to sustain fish populations. Higher N concentrations in the sediments were also observed after liming. They related the vegetation shift both to elevated  $\text{CO}_2$  – caused by liming - and increased N availability. Liming can lead to anoxic sediments, promoting ammonium production and increased  $\text{CO}_2$ , as well as additional release of phosphorus. Such conditions may be unfavourable for isoetids. Higher  $\text{CO}_2$  concentrations in the water can lead to shifts in the interspecific competitive regimes. It was found experimentally that the isoetid *Littorella uniflora* was replaced by the elodeid *Myriophyllum alterniflorum* after increased  $\text{CO}_2$ -concentrations in water (Spierenburg et al., 2009; Spierenburg et al., 2010).

To restore the original isoetid vegetation in soft-water Atlantic lakes, removal of N-enriched bottom sediments was attempted but not successful, as parts of the seed bank were removed along with the sediments leading to a delay in the establishment of the original vegetation (Brouwer et al 1996, referred in Roelofs (2002)).

The cause for the observed loss of many isoetids from Atlantic soft clear-water lakes and the increase in *Juncus bulbosus* is hence assumed to be an effect of interspecific competition, due to shifts in CO<sub>2</sub>, the nitrate/ammonium ratio and epiphytic growth as reviewed by Bobbink and coauthors (Bobbink et al., 2010), where species with nutrient uptake through leaves and adapted to ammonium utilisation will be competitively superior when NH<sub>4</sub> accumulates and CO<sub>2</sub>-concentration has increased. However, the coexistence of isoetids and *Juncus* in many oligotrophic lakes, and the observed increase in *Juncus* in southern Norway, is still poorly understood and may be controlled by other factors than liming and N-availability. Water regulation for hydropower and climate change have also been mentioned as important factors that determine the dynamics of macrophyte vegetation.

### **Conclusion**

Effects of nitrogen on macrophytes in oligotrophic waters have not been investigated over a wide range of species, regions or atmospheric loads of N. The typical water plant communities of oligotrophic acid lakes (isoetids) – acid-tolerant and able to live in conditions of low N availability - have declined strongly in Western Europe in the 20<sup>th</sup> century. In some regions (Atlantic lowlands), the isoetid vegetation was replaced by other species like bulbous rush (*Juncus bulbosus*) and *Sphagnum* while in boreal lakes, coexistence of isoetids and bulbous rush is common. Lake surveys from Western Europe and some targeted experimental studies indicate that the shift in vegetation community is not related to acidification but rather to increases in nutrient availability, especially NH<sub>4</sub>. The links between community shifts, increased N availability and deposition are not clear-cut because other factors like liming, sediment processes, climate and interspecific competition also play a role. Species-specific characteristics like ability to utilise NO<sub>3</sub> or NH<sub>4</sub> will also co-determine dominance or loss of species. In Norway, liming has led to loss of isoetid vegetation, probably due to increased CO<sub>2</sub> concentrations and changing N availability in sediments. Most data on lake vegetation stem from the 1980s and 1990s and more recent data are generally lacking.

## 6. Nitrogen enrichment effects on invertebrates

Invertebrates (such as crustaceans, insects or snails) are, especially in streams and rivers, used worldwide as biomonitoring agents to quantify the impact of organic pollution (defined as biological oxygen demand, BOD<sub>5</sub>: the amount of oxygen consumed by bacteria after five days). The concepts are founded on the general observation that specific invertebrate presence and absence vary in accordance with BOD, mainly caused by sewage and manure. This premise is also applied in the methodology of the EU Water Frame Directive (Buffagni et al., 2006), where the ASPT-index system of macroinvertebrates is widely used.

When focussing on specific elements, however, their reliability as biomonitoring agents is drastically reduced, and reports on responses of invertebrates to atmospheric nitrogen deposition are scarce. Friberg et al (2009) analysed data on macroinvertebrates from 594 danish streams in relation to different chemical variables, including total N and ammonium. More specifically, they investigated how the occurrence of taxa related to gradients in water chemistry, and whether some taxa exhibited distinct threshold or optimum values. Their results indicated the significance of organic pollution as an anthropogenic main driver for taxonomic composition: Various levels of BOD were significantly associated with presence or absence of defined taxa. It was, however, impossible to separate the effect of BOD from those of plant nutrients associated with eutrophication. In two cases, solely (the Trichopterid family Glossosomatidae and the Coleoptera genus *Limnius sp.*), shifts in occurrence were negatively correlated with tot N. In all other cases, no response was specifically related to tot N or to NH<sub>4</sub>. As BOD and NH<sub>4</sub> were strongly correlated, it is not possible to consider the effect of NH<sub>4</sub> separately.

Indirect effects of increased atmospheric N deposition on macroinvertebrates and zooplankton in oligotrophic lakes and streams due to a fertilization effect of N on phytoplankton and periphyton (see relevant chapters in this report) are conceivable. However, the responses may be complex and difficult to predict, and have so far not been quantified. Increased mats of filamentous algae related to increased N deposition in oligotrophic streams could possibly imply a more extensive habitat for specific taxa (as for Chironomid larvae), while at the same time reduce periphyton growth which serves as a diet for other taxa. A considerable body of research shows responses of pelagic zooplankton communities to various C:P and C:N-ratios in diet (Andersen and Hessen, 1991). Limitation of phosphorus or nitrogen will affect the phytoplankton species composition and nutrient quality of seston, which may then provoke shifts in the zooplankton grazer community (McCarthy et al., 2006). In general, Copepods have higher nitrogen needs, while Daphnids have higher phosphorus needs. Quite recently, McCarthy and Irvine (2010) compared soft and alkaline low nutrient lakes in relation to stoichiometric C:N:P-ratios in seston, and found these ratios reflected in differences in the zooplankton compositions between lakes. Zooplankton species composition was shown to have significant effects both on phytoplankton and fish recruitment and production. Enrichment of surface waters by atmospheric deposition of N has the potential to affect the entire food chain (Elser et al., 2009).

## 7. Policy implications – critical N loads for freshwaters

The report presents a review of nitrogen enrichment effects in oligotrophic waters, a process which also could be covered by the term ‘eutrophication’. Eutrophication occurs when nutrient fertilization of an ecosystem leads to increased primary productivity. It is a global problem in freshwater and coastal ecosystems with profound negative effects on water quality (Hutchins, 1973; Smith, 2003; Smith and Schindler, 2009; Smith et al., 1999). The most alarming effect of severe eutrophication is probably the increased presence of bloom-forming cyanobacteria (blue-green algae) which can impair water quality by forming scums, and excrete toxins with hazardous effects on humans and cattle. Cyanobacterial poisoning of humans, pets and livestock has been extensively documented (Falconer, 2008; Mez et al., 1997; Rao et al., 2002). The symptoms of N enrichment documented in this report are limited to oligotrophic waters where risks of cyanobacterial blooms are small (Downing et al., 2001). In aquatic ecosystems that receive nutrient inputs from sewage and agriculture, symptoms of severe eutrophication are more common. Additionally, atmospheric N leaking from natural vegetation and channelled by rivers can contribute to eutrophication in coastal and marine ecosystems (Howarth, 2008; Howarth et al., 1996). Policy aimed at reduction of atmospheric emissions of N should also take into account the fertilizing effect of leached N in downstream aquatic habitats – natural, agricultural, urban or coastal. However, critical loads of N deposition for freshwaters presented here are related to responses in the freshwaters themselves.

### Status for empirical critical loads of N

Critical loads nutrient N for freshwaters were considered in an Expert workshop on Critical loads for Nitrogen in Berne in 2002 (Anonymous, 2002). Empirical critical loads for ecosystems were tied to habitats in the EUNIS classification (Moss and Davies, 2002). The freshwater habitat for which an empirical critical load was defined was surface standing waters, further restricted to oligotrophic softwater lakes ( $5\text{-}10\text{ kg N ha}^{-1}\text{ yr}^{-1}$ ) (Bobbink and Roelofs, 1995) and dune slack pools ( $10\text{-}20\text{ kg N ha}^{-1}\text{ yr}^{-1}$ ). The critical loads were evaluated in a declining order of reliability from ‘reliable’ to ‘quite reliable’ to ‘expert judgement’. The highest degree of reliability was given when a number of published papers showed comparable results; the critical loads were deemed ‘quite reliable’ when results of some studies were comparable; ‘expert judgement’ was pronounced when no empirical data were available for the particular ecosystems but results from other studies were likely to be relevant to this ecosystem. The critical loads for softwater lakes were considered to be ‘reliable’. In oligotrophic lakes, negative effects for macrophytes (isoetid species) were the indication of exceedance of the critical loads. For the dune slack pools (“expert judgement”) the indication was increased biomass and rate of succession. The background data supporting the choice of these critical loads were mainly from the Netherlands with references dating from the 1980s and 1990s.

In 2010, a new workshop on the on empirical critical loads was held in Noordwijkerhout. Here, a draft report on updated critical loads was presented and discussed (Bobbink et al., 2010). Based on much of the same literature that is reviewed in this ICP waters report and on a discussion between experts, it was agreed to provide a single critical load range for permanent oligotrophic clearwater lakes with a range of  $3\text{-}10\text{ kg N ha}^{-1}\text{ yr}^{-1}$  which should be described as reliable. It is recommended that the lower end of the range is used for boreal, sub-arctic and alpine lakes, and the upper end of range is used for Atlantic softwaters.

Critical loads for N as an acidifier in acid-sensitive surface waters were also summarized in the proceedings from the workshop in Berne (Anonymous, 2002) and repeated in the new report presented in Noordwijkerhout (Bobbink et al., 2010). These critical loads were calculated using the steady state approach - based on the assumption that N is the only source of acidification and that all N deposition, except for a small fraction retained in the catchment, leaches and acidifies the lakes. Thus, they represent a ‘worst-case’ situation.

### Updated empirical loads for nutrient N for freshwaters Table 8

One of the aims of this report is to contribute to a further development of empirical critical loads for nutrient N, by updating the literature review, extending the present critical loads to more types of habitats, and document in a better way which type of data, and from what regions the studies come that support the critical loads. Only studies where N deposition is related to a certain response are used in Table 8 to quantify the critical load. Reference is made to Tables in this report with the relevant information. Also, previously established critical loads from the workshop in 2002 are referred to.

In arctic lakes, only paleolimnological studies related responses (shifts in phytoplankton community) to N deposition. The critical load here is very low ( $1 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ), which might be related to the low N retention capacity of arctic catchments that usually lack vegetation and soils. Paleolimnological studies and lake surveys indicate that phytoplankton communities in alpine have changed in relation to N deposition, and productivity has increased. Thus, this critical load is supported by several paleolimnological studies, but not by other types of studies using experimental approaches, and should receive the medium grade of reliability, “quite reliable”.

Most responses to N deposition were noted in alpine, boreal and temperate catchments – from lake sediment records to algal growth (benthic and pelagic) to water plants. The critical load for boreal and temperate catchment types was the highest of the three types of catchments. Again, this may be related to N retention capacity of catchments, which is known to be dependent on land cover (Baron et al., 1994; Dise and Wright, 1995; Hessen et al., 2009; Sjoeng et al., 2007). The critical loads for the alpine, boreal, temperate and atlantic catchments is supported by a number of published papers from very different regions, focused on different organisms and employing different methods and are evaluated as ‘reliable’. Regarding the dunes, we refer to the previous evaluation from expert workshops as ‘expert judgement’. Critical loads for dystrophic lakes are based primarily based on regional surveys and not by other types of studies and should receive the medium grade of reliability, “quite reliable”.

Table 8 Proposed new empirical critical loads for nutrient N in freshwaters based on this review. N deposition in  $\text{kg N ha}^{-1} \text{ yr}^{-1}$ . Table numbers refer to Tables in this report. In *italics*, critical loads that were suggested in other reviews.

EUNIS	description	catchment type	Regions	Response	Critical load
C1.1	Oligotrophic softwater lakes	arctic	Europe, Canada, Greenland	1. Phytoplankton community shift at N deposition $< 1-1.5$ (Table 1)	1
		Alpine, boreal	USA, Europe	1. Phytoplankton community shift at N deposition 3-5 (Table 1) 2. Higher phytoplankton productivity at N deposition $< 5$ (Table 3)	3-5
		Temperate, boreal	Canada, USA, UK, Scandinavia, Netherlands	1. Phytoplankton community shift at N deposition 2-9 (Table 1) 2. Higher phytoplankton productivity at N deposition $< 5$ (Table 2, 3) 3. Shift of N to P limitation of benthic algae at N deposition 2-12 (Table 2, 4) 4. Productivity of benthic algae increases at N deposition 2-12 (Table 4) 5. <i>Macrophytes: loss of key isoetid species, increase of species like juncus bulbosus and sphagnum (Bobbink and Roelofs, 1995)</i>	5-10
		dunes	Netherlands	1. <i>Increased biomass and rate of succession (Bobbink et al., 2003)</i>	10-20
C1.4	Dystrophic lakes	Temperate, boreal	Sweden, Canada	1. Higher phytoplankton productivity, especially at N deposition $< 5$ (Table 3)	3-5

### Recommendations for further improvement of critical loads of nutrient N for freshwaters

A compilation of data from oligotrophic lakes in Europe and USA shows that the DIN to totP ratio is a better indicator of N-limitation of phytoplankton than the totN to totP ratio (Bergstrom, 2010), suggesting that modelling  $\text{NO}_3$ -leaching is a key challenge for critical loads of nutrient N. The results in Table 8 suggest that including land cover may be included as a proxy for catchment N retention

capacity, which is in line with a wealth of literature that indicates relations between catchment N-leaching and catchment characteristics.

For future establishment of reliable critical loads for freshwater ecosystems, not only greater insights in responses of aquatic organisms to N enrichment are required, but also a better understanding of N leaching from catchments to surface waters is necessary. Presently, relations between N deposition and N leaching are not very well understood (Wright and Dise, 1995; Goodale et al., 2005; Wright et al., 2001; Aber et al., 1998 & 1989). Consequently, future N leaching, especially given prolonged N enrichment and climate change, is difficult to predict (Wright et al., 2006; Stoddard, 1994).

Development of critical loads for nutrient N for the UK was addressed in a report summarizing effects of N deposition and climate change on freshwaters in the UK (Curtis and Simpson, 2007). Here, it is suggested to use the FAB (First-order Acidity Balance) model (Henriksen and Posch, 2001), which is used to calculate acidity critical loads of N deposition by simulation of NO<sub>3</sub> leaching (reference to Manual M&M). If a criterion based on NO<sub>3</sub> concentrations could be established for harmful effects in terms of nutrient status in waters, then the FAB model could in theory be used to set a critical load for nutrient N.

Other factors except NO<sub>3</sub> leaching that a nutrient N critical load would have to take into account would be P-availability, presence of sensitive organisms and growth-limiting factors other than N and P. However, N:P ratios in streamwaters vary strongly with water discharge and its controls remain poorly understood (Green and Finlay, 2010).

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