*Limnol. Oceanogr.*, 50(3), 2005, 987–994


# Effects of atmospheric nitrogen deposition on nutrient limitation and phytoplankton biomass in unproductive Swedish lakes

*Ann-Kristin Bergström*[1]
Department of Ecology and Environmental Science, Umeå University, SE-901 87 Umeå, Sweden

*Peter Blomqvist*[2]
Department of Limnology, Evolutionary Biology Centre, Uppsala University, Norbyvägen 20, SE-752 36 Uppsala, Sweden

*Mats Jansson*
Department of Ecology and Environmental Science, Umeå University, SE-901 87 Umeå, Sweden

*Abstract*

We used chemical data (3,907 lakes) and phytoplankton biomass (chlorophyll *a*) data (225 lakes) from Swedish lake monitoring programs to assess the effects of atmospheric nitrogen (N) deposition on nutrient limitation and phytoplankton biomass in unproductive Swedish lakes. There was a clear north–south gradient of increasing lake concentrations of dissolved inorganic nitrogen, which was related to the pattern of atmospheric N input. On the basis of positive relationships between total phosphorus (P) concentrations and phytoplankton biomass we conclude that lakes in areas of enhanced N deposition are mainly P limited during summer. This relationship was not detected in lakes in pristine areas with low N deposition, which, together with experimental evidence from the literature, suggest possible N limitation. During summer, lakes in high N-deposition areas had clearly higher phytoplankton biomass relative to the total phosphorus concentrations compared to lakes in low N-deposition areas. Thus, in Swedish unproductive lakes, high atmospheric N input is reflected by increased lake concentrations of dissolved inorganic nitrogen and, possibly, by a shift from natural N limitation of phytoplankton to P limitation. Our results also reveal that increased N input has caused a eutrophication with higher phytoplankton biomass as the result.

Increased atmospheric deposition of nitrogen (N), as a result of anthropogenic activities such as fossil fuel combustion and agricultural fertilizer application, is a common phenomenon in large areas of the world. The global rate at which reactive nitrogen is produced has doubled during the last century, which has led to an increased amount of excess nitrogen in nature (Galloway and Cowling 2002). Most aquatic research related to N deposition has concerned acidification of freshwater ecosystems. However, N is not only an acidifying component in freshwater ecosystems; it is also a nutrient, which together with phosphorus (P) regulates the growth of phytoplankton in lakes (Wetzel 2001). N limitation can be expected when the ratio of bioavailable N to bioavailable P on a mass basis is lower than 7:1, which is the mean ratio of N and P in phytoplankton nutrient demand (Redfield 1958). Thus, increased atmospheric sources of N, and especially the inorganic forms, can affect the N : P ratio of lakes and the balance between limiting nutrients. If lakes were N-limited in their natural state, excess input of N may cause a eutrophication.

Increased N loading due to atmospheric deposition has not been regarded as a eutrophication process since P is generally thought to be the most limiting factor to phytoplankton growth in northern temperate lakes. The concept of lakes being almost exclusively P-limited is to a large extent based on the results from whole-lake nutrient enrichment experiments conducted at the Experimental Lakes Area in Ontario, Canada (Schindler and Fee 1974; Schindler 1977), and on the positive relationships found between P loading and biomass of phytoplankton in mainly eutrophic lakes (Vollenweider 1968; Schindler 1978). However, there are several reasons to question the generality of the P-limitation concept when discussing unproductive northern temperate lakes. There is increasing evidence that phytoplankton in natural unproductive lakes, situated in more pristine areas with low N deposition, can be N-limited (Jansson et al. 1996; Levine and Whalen 2001; Fenn et al. 2003 and references therein), and enrichment with N or N + P often causes larger responses in phytoplankton growth than P (cf., Elser et al. 1990). Moreover, P limitation in unproductive lakes may be a derived character, evolved from increased N loading from the atmosphere during the last decades, as has been illustrated for Lake Tahoe (California–Nevada) (Goldman 1988; Jassby et al. 1995).

Against this background we used monitoring data to assess the effects of N deposition on nutrient limitation and phytoplankton biomass in Swedish unproductive lakes. In Sweden, there is a clear gradient, with the highest deposition of N in the southwestern part of the country (>1,500 kg km$^{-2}$ yr$^{-1}$), and the lowest in the north (<100 kg km$^{-2}$ yr$^{-1}$),

[1] Corresponding author (ann-kristin.bergstrom@emg.umu.se).
[2] Passed away during the course of the project.

*Acknowledgments*

We thank Gunnar Persson, at the Department of Environmental Assessment Analyses, Swedish University of Agricultural Sciences, Uppsala, Sweden, for help with data from the Swedish Lake Inventory Programs.

This study was conducted with funds from the Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning.

a pattern related to precipitation and distance to continental and local sources of emissions. Atmospheric deposition increased rapidly between the 1950s and the 1980s, and has since then remained at a fairly constant level. A large part of the total N deposition is in inorganic form (www.internat.environ.se). We compiled data from 3,907 Swedish unproductive lakes and compared the nutrient composition in the lakes with the atmospheric N deposition. We also evaluated biological data from 225 unproductive lakes and compared the phytoplankton biomass (chlorophyll *a* [Chl *a*]) with N deposition. The following hypotheses were tested: (1) Atmospheric N deposition has affected the N : P ratios in Swedish unproductive lakes; (2) the phytoplankton production in natural unproductive Swedish lakes, outside areas with enhanced atmospheric N deposition, is N-limited; (3) increased atmospheric N deposition has changed the growth conditions for phytoplankton so that phytoplankton production has become P-limited; (4) enhanced atmospheric N deposition has caused eutrophication of originally N-limited lakes.

## Materials and methods

*Lake databases*—Two lake databases were used, with kind permission by the Department of Environmental Assessment Analyses, Swedish University of Agriculture Sciences, Uppsala, Sweden. The first database, the Swedish National Lake Survey (SNLS), is a national inventory of the chemical and physical conditions in Swedish lakes (Bernes 1991). The Swedish Environmental Protection Agency (SEPA) has conducted the SNLS every fifth year since 1975. We used data from the two latest inventories (1995 and 2000), each comprising approximately 4,000 lakes. Lakes included in the SNLS were randomly selected from four different size classes. The lakes in the largest size class ($>$10 km$^2$; 380 lakes) were all included in the SNLS. One thousand lakes in each of the three smaller-size classes were randomly selected and evenly distributed throughout the country. In the SNLS lakes, surface samples were taken (depths between 1 and 2 m) on one occasion during late autumn/winter (October to February). The second database is the Swedish Reference Lake Monitoring Program (SRLMP), also run by SEPA. Approximately 350 lakes are included in SRLMP. They are evenly distributed throughout the country, with somewhat higher numbers in southern Sweden. The SRLMP lakes vary in sizes between 0.03 and 30 km$^2$, and the mean lake surface area per catchment area for the SRLMP lakes is 9% (Wilander 1997). The SRLMP lakes were sampled at higher intensity, 4–8 times per year, and samples were taken at different water depths. In addition to chemical and physical data, the SRLMP also includes analyses of biological variables, such as Chl *a*. In this study we used data from SRLMP from the time period 1996–2001. To compensate for the somewhat higher distribution of SRLMP lakes in southern Sweden, we complemented our data set with published results from different research projects (Jansson et al. 1996 and 2000; Blomqvist et al. 2001; Karlsson 2001; Sobek et al. 2003) where sampling occasions and procedures, as well as analytical procedures, are comparable to the procedures in the SRLMP lakes. The contribution from different research projects was 37 lakes distributed as: 6 lakes in region 3, 5 lakes in region 9, 10 lakes in region 12, and 16 lakes in region 13.

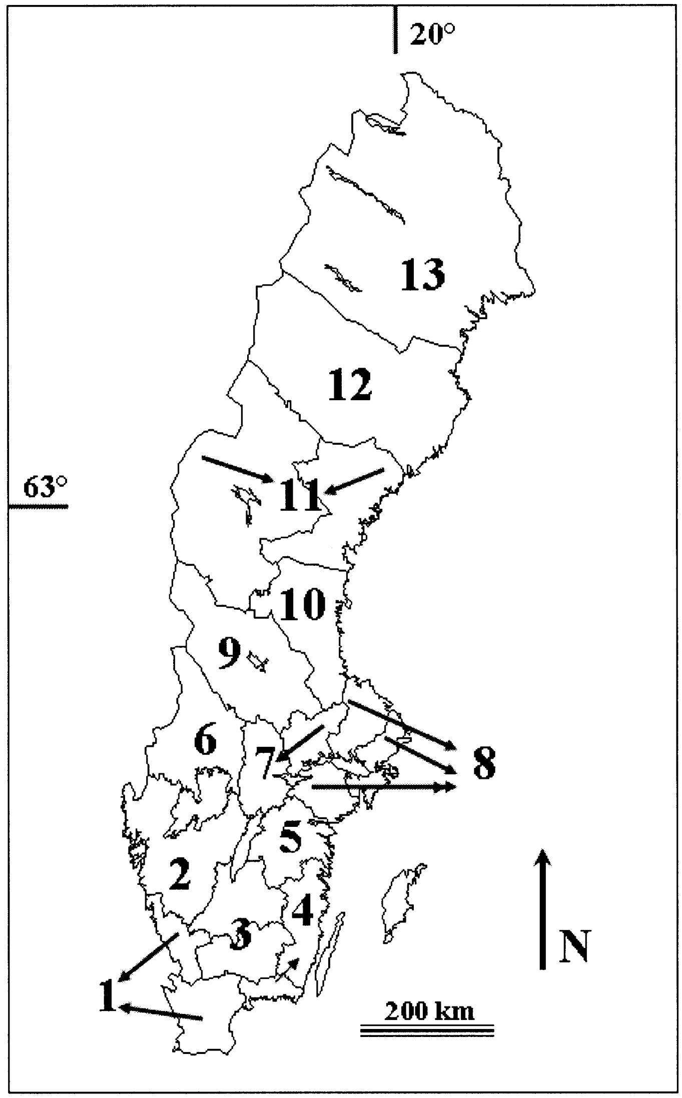


Fig. 1. The different Swedish regions used in this study.

Lake monitoring was administered by Swedish county administration, organizing lake databases following county borders (Fig. 1). As the size of the Swedish counties varies, small counties in southern Sweden situated close to each other were pooled into larger regions, to avoid large differences between total numbers of lakes within regions used in this study. In addition, two counties in northern Sweden were pooled into one region, region 11, to make this region comparable with regions 12 and 13, which both are geographically similar to region 11.

We checked for possible influences of differences in lake morphometry and lake water renewal times on water chemical characteristics between different regions. Data on lake area and catchment area were available for 10–30% of the lakes in each region in the Swedish Lake Register of the Swedish Meteorological and Hydrological Institute. We used these data and calculated the mean drainage ratio (DR = catchment area : lake area) in each region. We found no sta-

Table 1. Drainage ratio (DR), specific runoff (SR), and proxy of water renewal time (SR × log DR) for lakes in each Swedish region (mean values with standard deviations for DR presented within parentheses).

| Region | $n$ | log DR | SR (mm yr$^{-1}$) | Water renewal time proxy (SR × log DR) |
|---|---|---|---|---|
| 1 | 32 | 1.4(0.7) | 400 | 560 |
| 2 | 39 | 1.5(0.6) | 400 | 600 |
| 3 | 67 | 1.5(0.6) | 300 | 450 |
| 4 | 32 | 2 (0.6) | 150 | 300 |
| 5 | 26 | 1.4(0.6) | 100 | 140 |
| 6 | 53 | 1.5(0.6) | 100 | 150 |
| 7 | 47 | 1.7(0.7) | 250 | 425 |
| 8 | 52 | 1.5(0.5) | 300 | 450 |
| 9 | 103 | 1.5(0.6) | 400 | 600 |
| 10 | 64 | 1.9(0.8) | 300 | 570 |
| 11 | 80 | 1.8(0.7) | 500 | 900 |
| 12 | 43 | 1.7(0.5) | 500 | 850 |
| 13 | 52 | 1.6(0.6) | 500 | 800 |

tistically significant differences in DR between regions (Table 1). To assess possible impacts of water renewal times on lake water chemistry we calculated the product between mean specific runoff (mm yr$^{-1}$) (www.sna.se) and mean DR for each studied region as a proxy for lake water renewal time (Table 1). We found no correlation between the proxy of lake water renewal time and mean values of Chl *a*, total nitrogen (Tot-N), dissolved inorganic nitrogen (DIN), and total phosphorus (Tot-P) estimated for each region (see below: Chemical status in unproductive Swedish lakes); i.e., the parameters that are used in our analyses of the effects of N deposition on nutrient limitation and phytoplankton biomass in Swedish unproductive lakes (cf., below). The correlation coefficients ($r^2$) were 0.13, 0.20, 0.10, and 0.30, respectively, and $p > 0.05$. The proxy of lake water renewal time was not correlated to mean conductivity ($r^2 = 0.14$; $p > 0.05$), which shows that differences in lake water renewal times between regions do not to any large extent influence lake water chemistry.

*Chemical status in unproductive Swedish lakes*—Chemical and physical data were compiled from both lake databases. Data were gathered from late autumn and winter sampling from water depths of 1–2 m. Thus, the data represent the chemical status when phytoplankton production is low, and can be regarded to represent the potentially available pools of nutrients for phytoplankton. If a lake was included in both inventory programs, data from the latest sampling occasion was chosen; i.e., all compiled lake data represent one sampling occasion in each lake in late autumn/early winter during the time period 1995–2001. Only unproductive lakes, with Tot-P and total organic carbon (TOC) concentrations of $\leq 25$ $\mu$g L$^{-1}$ and $\leq 25$ mg L$^{-1}$, respectively, were selected from the two databases. Tot-P in such lakes generally covaries with TOC (humic content) (Meili 1992). For each region we therefore plotted the Tot-P concentrations against the TOC concentrations to assure that increased P concentrations were related to an increasing lake humic content (cf., Meili 1992; Nürnberg and Shaw 1998). We excluded data from lakes that did not follow the Tot-P–TOC relationship to avoid lakes with unnaturally high P concentrations or TOC concentrations possibly affected by agricultural activities or sewage water. On the basis of the mentioned criteria, 200 lakes were excluded from the original database of approximately 4,100 lakes. Our data set then comprised 3,907 lakes, i.e., 4% of the total number of lakes in Sweden. We did not exclude limed lakes from our selection of lakes, as the variations in nutrient concentrations between limed and unlimed lakes were insignificant (data not shown). Thus, except for liming, the only anthropogenic influence on the selected lakes was from forestry (which is performed at similar intensity throughout the country) and from atmospheric deposition. Mean values with standard deviation of the chemical and physical data were then calculated for lakes in each of the different Swedish regions (Fig. 1). The mean chemical and physical status of lakes in each region was then compared with the atmospheric N deposition in Sweden.

*Atmospheric deposition*—Wet deposition of DIN has been monitored monthly since 1983 at 25 stations distributed over all of Sweden within the national environmental surveillance program administered by SEPA (www.internat.environ.se). We used data (www.ivl.se) for the time period 1995–2001 and calculated the mean annual wet deposition of DIN for the different regions (Fig. 1) used in this study. Tot-N deposition is not included in regular monitoring of atmospheric deposition in Sweden. However, we estimated Tot-N deposition from the different regions by using data on Tot-N deposition in Sweden presented by the SEPA (which is performed using the so-called MATCH model) (www.internat.environ.se). Comparison of Tot-N and DIN deposition (Table 2) showed that regional variation of organic N (Tot-N minus DIN) was very small compared to DIN variation. Similar results have been reported in a large national survey in Finland where N deposition has the same sources and similar north–south gradients as in Sweden (Anttila et al. 1995). Therefore, we consider that DIN represents the major anthropogenic impact on atmospheric N deposition. P deposition is not included in Swedish monitoring programs. However, P deposition was reported to be low in Sweden (range between 10 and 100 kg km$^{-2}$ yr$^{-1}$) (Knulst 2001), and the N : P ratios (weight) of the deposition is considerably higher (range between 20 and 80) than, e.g., the Redfield ratio of phytoplankton. Similarly the DIN : Tot-P ratio in bulk wet deposition in Finland varied between 20 and 100 (weight) (Anttila et al. 1995) and regional variation in P deposition was small. Therefore, the regional N : P balance should have been changed mainly by increasing DIN deposition and not by deposition of organic N and P.

*Nutrient limitation*—To evaluate spatial differences in nutrient limitation, linear regression analyses were performed between Chl *a* and Tot-P concentrations with the data from lakes for each of the different Swedish regions. Thus, with P-limited lakes, the Chl *a* concentrations should be positively related to Tot-P concentrations (cf., Vollenweider 1968;

Table 2. Chemical characteristics (3,907 lakes) and Chl *a* data (225 lakes) of lakes and the atmospheric N deposition from different Swedish regions during the time period 1995–2001 (mean values with standard deviations within parentheses are given for all parameters except for total N deposition where the range is given).

| Region | pH | Cond. (mS m$^{-1}$) | TOC (mg L$^{-1}$) | Absorbance (420/5) | $NH_4$ ($\mu$g L$^{-1}$) | $NO_2$ + $NO_3$ ($\mu$g L$^{-1}$) | DIN ($\mu$g L$^{-1}$) | Tot-N ($\mu$g L$^{-1}$) | Tot-P ($\mu$g L$^{-1}$) | Tot-N : Tot-P | DIN : Tot-P | Wet DIN-Dep* (kg km$^{-2}$ yr$^{-1}$) | Tot-N-Dep† (kg km$^{-2}$ yr$^{-1}$) | Chl *a* ($\mu$g L$^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 (*n*=171) | 6.2(0.8) | 10.1(11.7) | 10.2(4.9) | 0.222(0.164) | 84(96) | 245(191) | 330(217) | 728(295) | 14(7) | 68(49) | 36(37) | 1100(320) | 900–1800 | 8.1(4.8) |
| 2 (*n*=312) | 6.2(0.8) | 6.8(5.3) | 10.9(4.6) | 0.21 (0.133) | 60(58) | 160(113) | 220(123) | 441(164) | 9(5) | 60(27) | 31(19) | 750(270) | 900–1500 | 4.9(4.3) |
| 3 (*n*=208) | 6.4(0.6) | 7.7(3.2) | 13.7(5.1) | 0.278(0.164) | 60(56) | 164(135) | 224(154) | 524(234) | 13(5) | 45(19) | 19(14) | 750(150) | 900–1500 | 6.2(3.5) |
| 4 (*n*=213) | 6.5(0.5) | 9.3(3) | 12.6(4.8) | 0.191(0.131) | 61(65) | 115(117) | 176(131) | 606(220) | 12(6) | 58(25) | 17(14) | 490(140) | 700–1200 | 4.6(2) |
| 5 (*n*=118) | 6.8(0.5) | 8.7(4.2) | 12.2(4.6) | 0.167(0.128) | 55(49) | 75(83) | 130(104) | 501(195) | 12(5) | 48(17) | 12(7) | 400(100) | 600–900 | 4.4(2) |
| 6 (*n*=170) | 6.4(0.5) | 3.6(1.5) | 11.2(4.5) | 0.2 (0.11) | 27(17) | 58(59) | 85(61) | 352(154) | 9(4) | 50(34) | 14(22) | 560(130) | 600–900 | 3.6(2.6) |
| 7 (*n*=249) | 6.5(0.6) | 5 (2.9) | 11.8(5) | 0.21 (0.139) | 34(39) | 58(55) | 93(69) | 504(240) | 12(5) | 48(24) | 10(9) | 490(80) | 600–900 | 3.2(2.3) |
| 8 (*n*=258) | 7.2(0.5) | 18.3(22.3) | 13.5(4.7) | 0.13 (0.1) | 47(53) | 33(36) | 80(66) | 722(344) | 15(6) | 56(33) | 6(5) | 400(100) | 500–900 | 7.2(3.6) |
| 9 (*n*=498) | 6.4(0.6) | 2.8(2.5) | 8.7(4.3) | 0.162(0.117) | 12(14) | 24(29) | 36(34) | 420(186) | 10(4) | 51(26) | 4(5) | 380(80) | 300–700 | 2.8(1.6) |
| 10 (*n*=259) | 6.6(0.4) | 3.5(2.7) | 10.2(4.4) | 0.195(0.114) | 22(28) | 24(24) | 45(41) | 433(92) | 10(5) | 49(30) | 5(4) | 470(100) | 300–600 | 3.5(2.1) |
| 11 (*n*=473) | 6.7(0.6) | 3.7(3.7) | 8(4.4) | 0.135(0.106) | 18(24) | 21(30) | 39(41) | 312(153) | 8(5) | 62(53) | 7(8) | 250(110) | 200–500 | 3 (1.2) |
| 12 (*n*=344) | 6.5(0.6) | 2.8(1.7) | 10 (6.3) | 0.157(0.137) | 15(15) | 18(18) | 33(28) | 290(131) | 7(5) | 64(57) | 6(5) | 220(100) | 150–400 | 2.3(1.5) |
| 13 (*n*=634) | 6.7(0.5) | 2.5(2.5) | 4.8(3.6) | 0.08 (0.088) | 10(11) | 7(8) | 17(15) | 286(116) | 7(6) | 70(60) | 4(6) | 130(90) | 100–400 | 1.1(0.6) |

* Data from www.ivl.se.
† Data from www.internat.environ.se.

Schindler 1977, 1978). The analysis was performed with data from the SRLMP lakes (1996–2001) complemented with published data from different research projects (cf., *Lake databases*). Only unproductive lakes (Tot-P and TOC $\leq$ 25 $\mu$gL$^{-1}$ and $\leq$ 25 mg L$^{-1}$, respectively), sampled during August at water depths 0.5–1 m, were used. August values were chosen to represent the summer conditions in the lakes. The analysis was performed on data from each region where the Tot-P concentrations in lakes varied from 5 $\mu$g L$^{-1}$ to 25 $\mu$g L$^{-1}$. As a whole this data set represents 225 lakes.

*Eutrophication of oligotrophic lakes*—We used the relationship between Chl *a* and Tot-P concentrations (Chl *a* : Tot-P ratio) to indicate if increased N deposition has caused a eutrophication of naturally N-limited lakes. The analysis was performed with the same lakes as the evaluation of nutrient limitation (i.e., the data set of 225 lakes). As mentioned earlier, the atmospheric P deposition in Sweden is low (cf., *Atmospheric deposition*), and P deposition in Sweden does not show the same geographical pattern as the N deposition (Knulst 2001). The choice of Chl *a* : Tot-P ratios as response variable to increased input of N is based on results from whole-lake experiments in northern Sweden (Jansson et al. 1996, 2001) and permits the assessment of a possible eutrophication effect of N deposition independent of eventual differences in P input between lakes in different regions. N-limited lakes should respond to increased input of N with an increase in the Chl *a* : Tot-P ratio. If lakes were P-limited, no increase in Chl *a* : Tot-P ratio could be expected as a result of increased N input. Similarly, if lakes were P-limited and received higher input of P, it should result in higher Chl *a* values but not a change in the Chl *a* : Tot-P ratio.

*DIN transport in Swedish rivers*—The DIN transport in river water for 12 catchments were calculated by using data from the Department of Environmental Assessment Analyses, SLU, Uppsala, Sweden (www.ma.slu.se). Catchments were chosen from regions 1, 2, 4, and 10–13 (Fig. 1). For each catchment, the mean annual river DIN transport was plotted against the mean annual wet DIN deposition (calculated mean values for the time period 1995–2001).

## Results

*Chemical status in unproductive Swedish lakes*—The chemical status (mean concentration with standard deviation) of lakes in different Swedish regions is presented in Table 2. The mean lake pH was very similar between regions, as well as the mean TOC concentration, with the exception of lakes situated in region 13. The mean conductivity was lower in lakes situated in the northern parts of Sweden. This is a reflection of variations in climate (i.e., lower air temperatures and weathering capacities in the north in comparison to the south) (Raab and Vedin 1995), variation in bedrock and soil characteristics (Fredén 1994), and variation in precipitation and atmospheric inputs of ions (Raab and Vedin 1995).

Mean ammonia ($NH_4$-N) and mean nitrite plus nitrate ($NO_2$-N + $NO_3$-N) concentrations increased from the northern part of Sweden to the southwestern parts. Hence, the DIN concentrations increased from 17 $\mu$g L$^{-1}$ in region 13 to 330 $\mu$g L$^{-1}$ in region 1 (Table 2). The pattern of increasing

Table 3. Linear regression analyses between Chl *a* and total phosphorus (Tot-P) concentrations.

| Region | Regression | $n$ | $r^2$ | $F$ | SE | $p$ |
|---|---|---|---|---|---|---|
| 1 | log Chl $a$ = (1.24 log Tot-P) − 0.57 | 25 | 0.56 | 29.5 | 0.22 | <0.001 |
| 2 | log Chl $a$ = (1.49 log Tot-P) − 0.82 | 14 | 0.88 | 89.8 | 0.19 | <0.001 |
| 3 | log Chl $a$ = (0.96 log Tot-P) − 0.26 | 22 | 0.62 | 32.5 | 0.15 | <0.001 |
| 4 | log Chl $a$ = (0.94 log Tot-P) − 0.28 | 8 | 0.8 | 25.2 | 0.11 | 0.003 |
| 5 | log Chl $a$ = (0.88 log Tot-P) − 0.26 | 17 | 0.53 | 17 | 0.13 | <0.001 |
| 6 | log Chl $a$ = (1.19 log Tot-P) − 0.46 | 7 | 0.85 | 28.5 | 0.1 | 0.003 |
| 7 | log Chl $a$ = (0.81 log Tot-P) − 0.14 | 7 | 0.81 | 21.1 | 0.10 | 0.006 |
| 8 | log Chl $a$ = (1.12 log Tot-P) − 0.42 | 19 | 0.68 | 38.9 | 0.13 | <0.001 |
| 9 | log Chl $a$ = (1.21 log Tot-P) − 0.68 | 13 | 0.59 | 16.1 | 0.2 | 0.002 |
| 10 | log Chl $a$ = (1.15 log Tot-P) − 0.63 | 5 | 0.94 | 39.9 | 0.06 | 0.006 |
| 11 | log Chl $a$ = (0.99 log Tot-P) − 0.45 | 34 | 0.56 | 39.9 | 0.11 | <0.001 |
| 12 | log Chl $a$ = (0.06 log Tot-P) + 0.23 | 19 | 0.002 | 0.03 | 0.26 | 0.8 |
| 13 | log Chl $a$ = (0.41 log Tot-P) − 0.38 | 35 | 0.16 | 5 | 0.23 | 0.03 |

DIN concentrations in lakes in the north-to-south gradient matches the spatial pattern of the total N deposition, and the mean wet DIN deposition in Sweden (Table 2) (linear regression ($n$ = 13); DIN ($\mu$g L$^{-1}$) = 0.34 wet DIN (N, kg km$^{-2}$ yr$^{-1}$) − 50.93; $r^2$ = 0.85, SD = 38.6; $p$ = < 0.001). Thus, lakes with high DIN concentrations were those in regions receiving high atmospheric N inputs. In addition, the standard deviation, especially for DIN, decreased from the southern parts to the north, indicating that differences in DIN concentrations among lakes were higher in areas of higher N deposition (Table 2).

The mean Tot-N concentration also showed an increasing trend from the northern parts of the country to the south. This trend was due to variation of DIN since organic N (Tot-N minus DIN) varies little between regions (see Table 2). Tot-P concentrations expressed only small and irregular variations between regions (Table 2). Consequently, the DIN : Tot-P ratios were considerably lower in the northern parts than in the southwestern parts of Sweden and increased from 4 in region 13 to 36 in region 1 (Table 2).

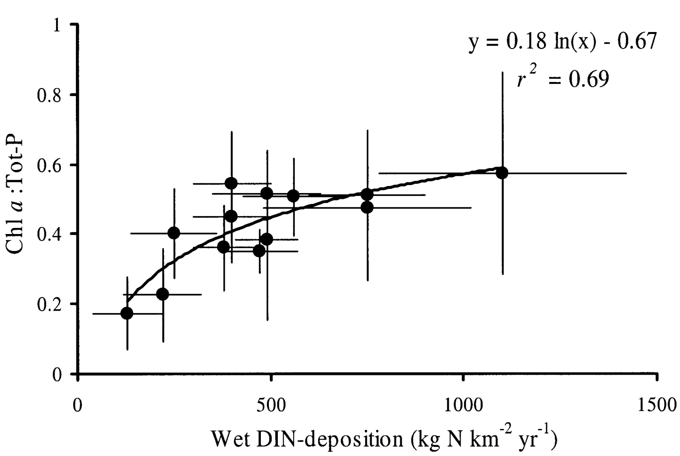


Fig. 2. The relationship between mean ratios between total phosphorus and chlorophyll *a* concentration (Chl *a* : Tot-P) and mean wet inorganic nitrogen deposition (wet DIN deposition) in unproductive lakes in different Swedish regions. The means are calculated for the period 1995–2001.

*Nutrient limitation*—Mean Chl *a* concentrations were higher in the southern parts of Sweden in comparison with the north (Table 2). A significant positive relationship between Chl *a* and Tot-P was found for all Swedish regions, except for regions 12 and 13 (Table 3), which have the lowest atmospheric N deposition (Table 2). The correlation coefficient ($r^2$), when excluding regions 12 and 13, ranged between 0.56 and 0.95.

*Eutrophication of oligotrophic lakes*—The mean Chl *a* : Tot-P ratios increased more than three times from low N deposition areas in region 13 (0.173) to high N deposition areas in region 1 (0.574) (Fig. 2) (regression: $n$ = 13; $r^2$ = 0.69; SD = 0.2; $p$ = < 0.001), indicating that N deposition and increased input of DIN (Table 2) have contributed to eutrophication (Table 4). The standard deviations for Chl *a* : Tot-P ratios and DIN deposition were larger (Fig. 2) for the regions situated in the south (Table 4).

Table 4. The Chl *a* : Tot-P ratios in lakes, the wet DIN deposition, the precipitation, and the inorganic nitrogen concentration in rain (DIN precipitation) in different Swedish regions (mean values, with standard deviations within parentheses, calculated for the period 1995–2001).

| Region | Chl $a$ : Tot-P | Wet DIN deposition (kg N km$^{-2}$ yr$^{-1}$) | Precipitation (mm) | DIN precipitation (mg L$^{-1}$) |
|---|---|---|---|---|
| 1 | 0.57(0.29) | 1100(320) | 882(229) | 1.3(0.3) |
| 2 | 0.48(0.21) | 7500(270) | 952(260) | 0.8(0.2) |
| 3 | 0.51(0.18) | 7500(150) | 872(164) | 0.8(0.2) |
| 4 | 0.51(0.13) | 490(140) | 631(80) | 0.8(0.2) |
| 5 | 0.45(0.13) | 400(100) | 477(123) | 0.8(0.1) |
| 6 | 0.51(0.11) | 560(130) | 814(166) | 0.7(0.1) |
| 7 | 0.38(0.23) | 490(80) | 808(148) | 0.7(0.1) |
| 8 | 0.54(0.15) | 400(100) | 477(123) | 0.8(0.1) |
| 9 | 0.36(0.12) | 380(80) | 890(148) | 0.4(0.1) |
| 10 | 0.35(0.06) | 470(100) | 786(96) | 0.6(0.1) |
| 11 | 0.40(0.19) | 250(110) | 814(213) | 0.3(0.1) |
| 12 | 0.23(0.13) | 220(100) | 723(157) | 0.3(0.2) |
| 13 | 0.17(0.10) | 130(90) | 521(170) | 0.2(0.1) |

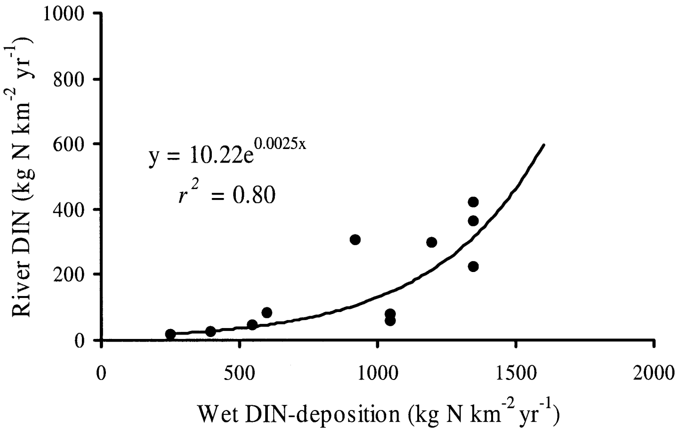


Fig. 3. The relationship between mean river transport of inorganic nitrogen (river DIN) and mean wet inorganic nitrogen deposition (wet DIN deposition) for different river catchments in Sweden (mean values from 1995–2001).

*DIN transport in Swedish rivers*—DIN transport in rivers was lower than atmospheric DIN input (Fig. 3), which shows that a large part of the DIN deposition is trapped or transformed to dissolved organic nitrogen in the terrestrial system. The DIN transport was similar in catchments with DIN deposition $<$1,000 kg km$^{-2}$ yr$^{-1}$ but increased considerably in catchments with higher deposition. Therefore, in the northernmost rivers, river transport represented only 6% of the atmospheric DIN input, whereas this number increased to 25–33% on the Swedish southwest coast.

## Discussion

There was an almost 20-fold increase in lake DIN concentrations from the northern parts of the country to the Swedish southwest coast that was related to the pattern of atmospheric DIN input (Table 2). A similar pattern was found for river transport of DIN (Fig. 3). The relationship between river transport of DIN and DIN deposition was not linear and indicates, similar to previously reported results by Dise and Wright (1995) and Moldan et al. (1995), that a larger share of atmospheric N deposition is leached to freshwater at deposition levels greater than 1,000 kg km$^{-2}$ yr$^{-1}$ than at lower deposition values. In Sweden these levels are only exceeded in the southernmost part, regions 1–4 (Table 2 and Fig. 1). This geographical variation in soil retention/leaching in Sweden has also been stressed by Binkley and Högberg (1997). We therefore conclude that atmospheric deposition of N in excess of natural levels since the middle of the 20th century has caused an increased input of N to streams and lakes in large parts of Sweden. Increased input is due both to direct deposition on the lake surface and increased terrestrial export from catchments receiving high deposition. Excess N-loading to lakes has been particularly pronounced in high-deposition areas in southern Sweden.

The effect of increased N input to lakes on the nutrient balance is obvious. Tot-N concentrations during autumn/winter were considerably higher in high-deposition areas, which is almost entirely a result of high DIN concentrations (Table 2). Tot-P concentrations showed some variations between different regions but with no geographical pattern, which clearly indicates that P input was not affected by atmospheric deposition. Consequently, the atmospheric N deposition was reflected in higher DIN : Tot-P ratios and to some extent in higher Tot-N : Tot-P ratios. The inorganic nutrient concentration during winter is little affected by uptake for biological production and can be seen as a measure of the potential nutrient availability for summer phytoplankton production.

To assess possible effects of altered nutrient balance on nutrient limitation of phytoplankton during summer we examined the relationship between Tot-P and Chl *a* in lakes within each region (Table 3). This relationship indicated that lakes were P-limited from the southern parts (region 1) up to the northern central parts of Sweden (region 11). For lakes in the northern parts (regions 12 and 13), variations in Chl *a* concentrations were not related to Tot-P concentrations, indicating that some other substance than P limited the growth of phytoplankton. Experiments and field studies on nutrient limitation in Sweden have shown that unproductive lakes situated in regions 12 and 13 are N-limited rather than P-limited during summer (Jansson et al. 1996 and 2000; Karlsson 2001). Moreover, nutrient enrichment experiments from different parts of Sweden show a switch from P limitation in the southwestern parts of Sweden (regions 2 and 7) (Jansson et al. 1986; Blomqvist et al. 1993) over N + P limitation in central–eastern parts of Sweden (regions 8 and 10) (Blomqvist et al. 1993, 1995), to N limitation in northern parts of Sweden (regions 12 and 13) (Jansson et al. 1996 and 2000; Karlsson 2001). We therefore interpret the nonexisting correlation between Chl *a* and Tot-P in the northern parts of Sweden to be a consequence of N limitation.

Our data also offer support for the picture, which evolves from the nutrient enrichment experiments referred to above, of a transition from N limitation via N + P limitation to P limitation in a north–south gradient. Thus, we obtained a significant relationship between Chl *a* and Tot-P in all regions except the two most northern ones (regions 12 and 13), which can be expected if phytoplankton in regions 1–11 are limited by P or N + P. When we plotted Chl *a* : Tot-P ratios for different regions against N deposition (Fig. 2) we observed an increase in this ratio with increasing N deposition, i.e., an increase in the use of P for buildup of phytoplankton biomass, which was proportional to N supply. This pattern probably reflects a gradual shift from clear N limitation to clear P limitation. We suggest that phytoplankton in unproductive Swedish lakes are limited mainly by N in areas little affected by deposition of DIN from the atmosphere, and that DIN deposition causes a shift from N limitation in areas with low deposition in the north to P limitation in areas with high deposition in the south.

Our result supports the suggestion of Goldman (1988) that P limitation in unproductive lakes can be a derived character induced by atmospheric N deposition. Similarly, N limitation has been recognized to be rather common among unproductive North American lakes (cf., Elser et al. 1990), especially in areas with low N deposition, e.g., Alaska (Levine and Whalen 2001), northern Minnesota (Axler et al. 1993), and elevated western U.S. lakes (Morris and Lewis 1988; Fenn et al. 2003 and references therein; Nydick et al. 2003). Thus, N limitation can be a more common feature in unproductive natural lakes than earlier believed.

If N-limited lakes receive an increased input of inorganic nitrogen, this input should increase the phytoplankton production and the phytoplankton biomass (cause eutrophication). Lakes in high N-deposition areas in our study had clearly higher biomass of phytoplankton (expressed as Chl *a*) relative to the Tot-P concentrations in comparison with lakes situated in low-deposition areas (Fig. 2), which indicates eutrophication caused by elevated N inputs. The Chl *a* : Tot-P ratios increased almost three times from region 13 to region 1 (Table 4). The relationship in Fig. 2 does probably not reflect a linear relationship. A linear correlation could be expected only if lakes in all regions in Fig. 2 were N-limited. Since clear N limitation is at hand only in the regions with the lowest N deposition (see discussion above) we interpret the relationship in Fig. 2 so that increased N deposition has a clear gradual eutrophication effect in the deposition interval below 500 kg $km^{-2}$ $yr^{-1}$ and that phytoplankton have become more or less saturated with N in areas with higher deposition. It is also obvious that nonlinear regression better explains the correlation in Fig. 2 than a linear regression (i.e., nonlinear regression [$n = 13$]: Chl *a* : Tot-P [$\mu$g $L^{-1}$] = 0.18 ln wet DIN [kg $km^{-2}$ $yr^{-1}$] − 0.16; $r^2 = 0.69$; and linear regression [$n = 13$]: Chl *a* : Tot-P = 0.0003 wet DIN [kg $km^{-2}$ $yr^{-1}$] + 0.25; $r^2 = 0.55$). Further increases in biomass numbers of phytoplankton in high DIN deposition areas (Fig. 2) were most likely hindered by low P availability (Table 3). Naturally low P concentrations, therefore, set the limits for eutrophication caused by atmospheric DIN input and lead to accumulation of excess amounts of DIN in lakes (Table 2).

We conclude that our results agree well with our hypotheses, i.e., that increased N deposition has caused a higher input of DIN to unproductive lakes in Sweden, and that high N input is reflected by increased lake concentrations of DIN, and a shift from natural N limitation of phytoplankton to P limitation. Increased N input has also caused a eutrophication with higher phytoplankton biomass as the result.

## *References*


ANTTILA, P., P. PAATERO, U. TAPPER, AND O. JÄRVINEN. 1995. Source identification of bulk wet deposition in Finland by positive matrix factorization. Atmos. Environ. **29:** 1705–1718.

AXLER, R. P., C. ROSE, AND C. A. TIKKANEN. 1993. Phytoplankton nutrient deficiency as related to atmospheric nitrogen deposition in northern Minnesota acid-sensitive lakes. Can. J. Fish. Aquat. Sci. **51:** 1281–1296.

BERNES, C. 1991. Acidification and liming of Swedish freshwaters (Monitor 12-SEPA). Schmidts Boktryckeri AB.

BINKLEY, D., AND G. P. HÖGBERG. 1997. Does atmospheric deposition of nitrogen threaten Swedish forests? Forest Ecol. Manage. **92:** 119–152.

BLOMQVIST, P., M. JANSSON, S. DRAKARE, A.-K. BERGSTRÖM, AND L. BRYDSTEN. 2001. Effects of additions of DOC on pelagic biota in a Clearwater system: Results from a whole lake experiment in Northern Sweden. Microb. Ecol. **42:** 383–394.

———, T. B. RUSSEL, H. OLOFSSON, U. STENSDOTTER, AND K. VREDE. 1993. Pelagic ecosystem responses to nutrient additions in acidified and limed lakes in Sweden. Ambio **5:** 283–289.

———, T. B. RUSSEL, H. OLOFSSON, U. STENSDOTTER, AND K. VREDE. 1995. Plankton and water chemistry in Lake Njupfatet before and after liming. Can. J. Fish. Aquat. Sci. **52:** 551–565.

DISE, N. B., AND R. F. WRIGHT. 1995. Nitrogen leaching from European forests in relation to nitrogen deposition. Forest Ecol. Manage. **71:** 153–161.

ELSER, J. J., E. R. MARZOLF, AND C. R. GOLDMAN. 1990. Phosphorus and nitrogen limitation of phytoplankton growth in freshwaters of North America: A review and critique of experimental enrichments. Can. J. Fish. Aquat. Sci. **47:** 1468–1477.

FENN, M. E., AND OTHERS. 2003. Ecological effects of nitrogen deposition in the western United States. BioScience **53:** 404–420.

FREDÉN, C. 1994. Geology. National Atlas of Sweden. Almqvist and Wiksell International. 208 p.

GALLOWAY, J. N., AND E. B. COWLING. 2002. Reactive nitrogen and the world: Two hundred years of change. Ambio **31:** 64–71.

GOLDMAN, C. R. 1988. Primary productivity, nutrients, and transparency during the early onset of eutrophication in ultra-oligotrophic Lake Tahoe, California-Nevada. Limnol. Oceanogr. **33:** 1321–1333.

JANSSON, M., A.-K. BERGSTRÖM, S. DRAKARE, AND P. BLOMQVIST. 2000. Nutrient limitation of bacterioplankton and phytoplankton in humic lakes northern Sweden. Freshw. Biol. **46:** 653–666.

———, P. BLOMQVIST, A. JONSSON, AND A.-K. BERGSTRÖM. 1996. Nutrient limitation of bacterioplankton, autotrophic and mixotrophic phytoplankton, and heterotrophic nanoflagellates in Lake Örträsket. Limnol. Oceanogr. **41:** 1552–1559.

———, G. PERSSON, AND O. BROBERG. 1986. Phosphorus in acidified lakes: The example of Lake Gårdsjön, Sweden. Hydrobiologia **139:** 81–96.

JASSBY, A. D., C. R. GOLDMAN, AND J. E. REUTER. 1995. Long-term change in Lake Tahoe (California-Nevada, USA.) and its relation to atmospheric deposition of algal nutrients. Arch. Hydrobiol. **135:** 1–21.

KARLSSON, J. 2001. Pelagic energy mobilization and carbon dioxide balance in subarctic lakes in northern Sweden. PhD thesis, Umeå University.

KNULST, J. C. 2001. Phosphorus in precipitation—results from measurements during the 1990′s. Report B1442. IVL Swedish Environmental Research Institute.

LEVINE, M. A., AND S. C. WHALEN. 2001. Nutrient limitation of phytoplankton production in Alaskan Arctic foothill lakes. Hydrobiologia **455:** 189–201.

MEILI, M. 1992. Sources, concentrations and characteristics of organic matter in softwater lakes and streams of Swedish forest region. Hydrobiologia **229:** 23–41.

MOLDAN, F., H. HULTBERG, U. NYSTRÖM, AND R. F. WRIGHT. 1995. Nitrogen saturation at Gårdsjön, southwest Sweden, induced by experimental addition of ammonium nitrate. Forest Ecol. Manage. **71:** 89–97.

MORRIS, D. P., AND W. M. LEWIS. 1988. Phytoplankton nutrient limitation in Colorado mountain lakes. Freshw. Biol. **20:** 315–327.

NÜRNBERG, G. K., AND M. SHAW. 1998. Productivity of clear and humic lakes: Nutrients, phytoplankton, bacteria. Hydrobiologia **382:** 97–112.

NYDICK, K. R., B. M. LAFRANCOIS, J. S. BARON, AND B. M. JOHNSON. 2003. Lake-specific responses to elevated atmospheric nitrogen deposition in the Colorado Rocky Mountains, U.S.A. Hydrobiologia **510:** 103–114.

RAAB, B., AND H. VEDIN. 1995. Climate, lakes and rivers. National Atlas of Sweden. Almqvist and Wiksell International. 176 p.
REDFIELD, A. C. 1958. The biological control of chemical factors in the environment. Am. Sci. **46:** 205–221.
SCHINDLER, D. W. 1977. Evolution of phosphorus limitation in lakes. Science **195:** 260–262.
———. 1978. Factors regulating phytoplankton production and standing crop in the world's freshwaters. Limnol. Oceanogr. **23:** 478–486.
———, AND E. J. FEE. 1974. Experimental Lakes Area: Whole-lake experiments in eutrophication. J. Fish. Res. Bd. Can. **31:** 937–953.
SOBEK, S., G. ALGESTEN, A.-K. BERGSTRÖM, M. JANSSON, AND L. J. TRANVIK. 2003. The catchment and climate regulation of $pCO_2$ in boreal lakes. Glob. Change Biol. **9:** 630–641.
VOLLENWEIDER, R. A. 1968. Scientific fundamentals of the eutrophication of lakes and flowing waters, with particular reference to nitrogen and phosphorus as factors in eutrophication. Paris, Rep. Organisation for Economic Cooperation and Development.
WETZEL, R. G. 2001. Limnology, 3rd ed. Academic Press.
WILANDER, A. 1997. Referenssjöarnas vattenkemi under 12 år; tillstånd och trender (in Swedish). Nat. förlag.

*Received: 8 June 2004*
*Amended: 25 November 2004*
*Accepted: 15 December 2004*