Nitrogen retention in lakes in Sweden; a review

by

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Summary

Nitrogen retention was defined as the difference between total nitrogen input to a lake (including microbial nitrogen fixation) and the outlet nitrogen loss. When changes in the nitrogen pool of lake water was neglected the term equilibrium retention was used. It is dominated by gaseous nitrogen losses to the atmosphere through microbial denitrification. Focus is put here on the magnitude and regulation of this process.

Lake sediments offer particularly good denitrification sites if nitrate and easily degradable organic matter is available. At an almost anoxic zone below the sediment surface both nitrification (a microbial production of nitrate under oxic conditions) and denitrification may co-occur in a microzonation that strongly promotes denitrification. Nitrate may also diffuse from oxic superficial sediment layers or oxic lake water. The rate limiting step may be nitrate production, oxygen status or supply of organic matter.

The direct assessment of denitrification suffers from a number of problems that are reviewed. Assessments of nine cases in Sweden indicated higher rates in bottoms above the thermocline as compared to bottoms below. Furthermore the process seemed not to be enhanced during summer in six of the lakes. Obviously, unfavourable denitrification conditions lowered possible rates during this period. The limiting factor(s) was not pinpointed.

Nitrogen retention and denitrification rates may also be obtained through mass balance. Retention is calculated as $\sum$input -$\sum$output and denitrification as retention - permanent sediment accumulation. In the present survey 62 Swedish yearly mass balances have been compiled. Nine of these also gave denitrification by difference (mean 73% of retention). In the complete material a medium equilibrium retention of 40% was noted. 25 and 75 percentiles were 21 and 49% respectively. In absolute numbers the 25, 50 and 75 percentiles corresponded to 1.6, 4.0 and 9.9 g $N{*}m^{-2}$,y. A brief check showed the low-retention group to contain lakes with higher than average water flushing, higher external and internal phosphorus supply as well as low phosphorus retention.

Further reference data on nitrogen retention in Sweden is found in two studies using dynamic models to estimate nitrogen retention in watersheds and lakes over large parts of southern Sweden (ca. 1700 lakes). The retention rates compiled here were found to be higher than those modelled. One reason may be that medium-eutrophic lakes are over-represented in the compiled material.

In an attempt to reveal factors that regulate denitrification, multiple regression was performed on the complete set of data and on subsets of lakes having a) potential for nitrogen fixation or not, b) high phosphorus concentration or not, c) high water turnover time or not and d) high phosphorus retention or not. Nitrogen input was found to explain a major part of the variation (r^2=0,66, log-transformed) in the set of retentions, and further reduced variance and a logical behaviour was found for the subgroups. The functions agreed well with a similar model also based on Swedish data.

When hydraulic loading was added as a factor the variance was further explained (r^2=0,77 for the whole set) and for two of the subsets 81% of the variance was explained. The additional inclusion of a phosphorus factor or a N/P-factor did not give any significant three-variable functions. On the other hand functions containing just nitrogen input and phosphorus in-lake concentration explained a major part of the variance for the total set. The subsets did not behave logically though, and it was hypothesised that nitrogen retention may be described as an optimum function of phosphorus supply. The functions describing nitrogen retention as a function of nitrogen input and hydraulic loading is recommended as predictive models (p .34)

Content

Preface

The present report on nitrogen retention in Swedish lakes was initiated by the Swedish environmental protection agency. The agency had to supply data for negotiations on the EU-directive on treatment of urban sewage water (Directive 91/271/EEG). In the question on nitrogen reduction in sewage works, it was wished from the Swedish side to elucidate the role of natural nitrogen reduction in the many lakes and watercourses protecting the coastal and sea areas declared as "sensitive". In this context the retention in lakes was considered of key importance and the Department of Environmental Assessment of the Swedish University of Agricultural Sciences was asked to supply such data. These data are supplementary to data on nitrogen retention on a landscape scale earlier supplied by other contractors to the agency (Anon. 1999).

The data used in this report was gathered from various written sources. They were frequently made available to the present author upon inquiries to regional authorities and collegues or even calculated by colleagues on available material. This made a broad compilation possible, for which the author wish to thank all kind suppliers of data, as well as suppliers of hints to find data.

No names are mentioned here but as a special tribute to all participants an appendix containing the core of the final database is published including the sources. This is also intended to facilitate further reuse of gathered data and encourage revision of data.

The author is also indebted to Anders Wilander for supplying constructive criticism on the report.

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Gunnar Persson

Introduction

Dissolved nitrogen, on its way through the landscape, suffers from regular losses through denitrification, releasing gaseous nitrogen and through immobilization and accumulation of organic nitrogen in soil and sediments. Both these processes, commonly known as nitrogen retention, are particularly important in lakes, where water residence times (and reaction times) are long, and the environments often favourable for microbial processes leading to loss of gaseous nitrogen. The losses are also fairly easy to assess in lakes as differences between input and output of nitrogen. Such data form a most solid ”ground truth” about nitrogen retention in the majority of cases where nitrogen fixation is minimal or can be accounted for.

Nitrogen retention or denitrification may also be assessed by other methods and at other types of sites, but the present paper is devoted to nitrogen mass balances for lakes in Sweden. Parts of the material has earlier been compiled by Fleischer et al. (1991) and Hellström (1996) but a compilation of the nationally available material has long been lacking.

This survey of nitrogen retention in lakes is also complementary to studies of denitrification i other environments. Graneli (1993) comprehended national and international knowledge about ”Wetlands as nitrogen traps” as part of a joint project focused on nitrogen retention which was later reported in a special issue of the ”Ambio” journal (Jansson et al 1994). Furthermore, Arheimer (1998) treated ”Riverine nitrogen …under Nordic conditions” where she dealt with retention in water courses in particular but also with other sites on a landscape scale. Part of her work was included in a particularly broad study on nitrogen retention on a watershed and landscape scale, initiated by the Swedish Environmental Protection Agency to test different scenarios of nitrogen discharges from agriculture and point sources (Arheimer & Brandt 1998, 1999, Anon. 1997) This study covered the southern part of Sweden (from Stockholm southwards), involved 3725 subwatersheds and1637 lakes. Later the methods were slightly modified and used to model nitrogen behaviour in 1000 watersheds covering the entire Sweden (Brandt & Ejhed 2003). Similar but smaller studies involved up to 1513 subwatersheds and 40 lakes (eg. Johansson & Kvarnäs 1998, Sonesten et al. 2003).

In these landscape studies the approach was to estimate nitrogen retention by dynamic modelling. In the dynamic models temperature, water discharge and nitrogen losses (point and/or diffuse source) are used as inputs. This kind of watershed models considers subwatersheds in separate subroutines and retention may be modelled separately for water passing soils as well as for stream channels or lakes. Retention is thus stepwise modeled and the downstream transport is described through successive addition of the contributions from subwatersheds. Essential for this type of models is the calibration of several factors in the various retention functions against measured concentrations at various points in the watershed. Such of retention models are thus very dependent on time series of data both for the dynamic driving functions and for calibration purposes.

The approach presented in this paper is of a more static descriptive nature, and with the aim to use retention data from environmentally different lakes to analyse which factors are more or less instrumental in the regulation of denitrification. A second aim is to formulate simple functions for prediction of yearly nitrogen retention in different lakes.

However, a basic knowledge of all processes involved and their regulation is essential for both analysis and modelling. This paper therefore starts with a review of processes and their *in situ* assessment.

Retention terminology

The term "retention" appears in calculations of balances between input and output of a substance to a physical site or a theoretical compartment. Retention stands for all losses calculated as the difference between input and output including also a temporarily increased storage in the pool under consideration. Although negative retention (meaning a source term) may appear over shorter periods of time or under very special conditions, the use of that term should be avoided.

In the terrestrial and aquatic nitrogen cycle, the components of retention are i) gaseous losses due to denitrification ii) permanent burial or accumulation in refractory humic matter in soils and accumulation in sediments which build up in aquatic systems and iii) temporary storage or immobilization.

Denitrification, the bacterial transformation of nitrate to nitrogen gas (N_2) – and to a smaller extent dinitrogen oxide (N_2O) – generally is the major retention component whereas permanent accumulation follows next (Seitzinger 1988, Christensen et al 1991). The size of the components may be variable, however (Leonardson,1993).

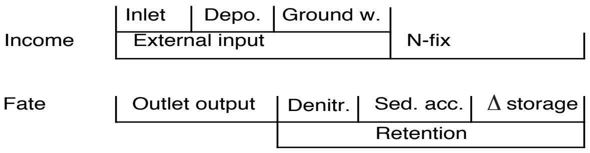
Temporary accumulation in biota, in organic detritus or storage as dissolved compounds (iii, above) may be considered and included in the retention term dependent upon the time scale studied. If, however, the natural cycle of one year is considered, temporary storage in different pools should theoretically be balanced. The remaining changes in storage between years, or rather over a number of years, are in fact those that generally are included as parts of calculated retentions.

The terms gross and net retention are sometimes used to distinguish between temporary storage (= seasonally) and more permanent retention.

In practice, net retention may have significant contributions from accumulation in biomass-expansive vegetative communities (i.e. wetland vegetation, forests). Harvest and translocation to other systems of nitrogen in biomass from the system under consideration may also be included in net retention (Leonardsson, 1993).

Nitrogen retention occurs at many sites in an entire watershed. This review aims at describing the nitrogen retention primarily in lakes.

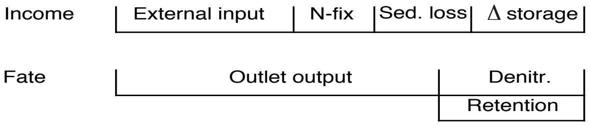
Nitrogen net balance of lake water



Ret=Input-output (in cases with negligible nitrogen fixation)
Ret=Input+fix-output (in cases with significant nitrogen fixation)
Eq.Ret=Input+fix-output-□ storage (in cases with significant change in concentration and/or water level)
%Ret=Ret/Input
%Eq.ret=(Ret-Δstorage)/Input
%Denitr=Denitr/Ret
%Denitr=Denitr/Input

Figure 1. Terminology and elements in the nitrogen balance of lakes. In the analysis of governing factors the term for changed nitrogen pool in the water mass "Δ storage"has been omitted. It is also seldom reported.

Nitrogen balance; atypical cases



The denitrification process

1. Microbial transformations and their ecological optima

Bacterial denitrification, is the dominating pathway of dissimilative nitrate reduction, the process where organic matter is oxidized with nitrate as an electron acceptor instead of oxygen like in "ordinary" respiration. The term considers the processes where nitrate or other oxidized nitrogen compounds are reduced to gaseous end products as originally stated by Gayon & Dupetit (1886). Nitrate is the most frequently used electron acceptor. It is reduced, over nitrite to nitrogen oxide (NO), dinitrogenoxide (N_2O) and/or molecular nitrogen (N_2). The nitrogen gases diffuse into the atmosphere and are lost from the ecosystem; thus denitrification results in the permanent removal of fixed nitrogen from an ecosystem. A great number of common bacteria possess the ability to produce the nitrate reductase enzyme (used in denitrification) in addition to cytochrome oxidase which they may normally possess and use when oxygen is used as the electron acceptor (Nicholas 1963). The switching ability of these heterotrophic bacteria means that they are facultative anaerobic (Brezonik 1975). The biochemistry and physiology of denitrification have been reviewed by Painter (1970), Paine (1973), Focht & Verstraete (1977) and Knowles (1982).

Another kind of denitrification carried out by chemolithotrophic bacteria using sulphuric compounds as energy donors rarely occurs in anoxic hypolimnia of lakes. It is considered to be of low general significance (Wetzel 1983) and is not treated further on. Nitrogen oxide and dinitrogen oxide may also appear as a result of non-respiratory processes in fungi, nitrate assimilatory bacteria and algae (Tiedje 1988) or in a respiratory process (Rosswall et al 1989). These processes may also contribute to retention but they are poorly known and most often neglected.

The denitrification process must not be confused with the dissimilative process often called nitrate-ammonification where nitrate is reduced to ammonium (Pettersson & Boström 1990). This process may occur under similar conditions as denitrification, but is considered significant only in situations with extremely high organic load. It does not affect nitrogen retention since the end product is confined within the system. It is not further treated here.

Denitrification is the most important process used for nitrogen reduction in modern sewage works. This has greatly improved our knowledge of the very special conditions required to optimize the process.

- ❑ The process is heterotrophic and adequate supply of organic material of an easily degradable type is needed.
- ❑ The process proceeds under almost anaerobic conditions (below 0,2–0,3 mg O_2*l^{-1}) and even under anoxic conditions, since nitrate is used as an alternate electron acceptor to oxygen.
- ❑ High oxygen levels energetically favours the use of oxygen as electron acceptor and is also inhibitory to the synthesis of the enzyme nitrate reductase and some cytochromes needed in the process.
- ❑ High to intermediate oxygen levels are on the other hand needed for the bacterial oxidation of ammonium to nitrate – the precursor for the denitrification process.
- ❑ The cell-specific rate or capacity for denitrification is fairly low meaning that high bacterial abundance and long transit time is essential in sewage works.

- The process is highly temperature dependent and is particularly slow below 10°C, approaching zero at 4°C.
- The pH-optimum of the process is at pH 7-8 whereas acid conditions are inhibitory (severely inhibitory below pH 4). The proportions between the end products N_2O and N_2 are also altered in favour of N_2O at low pH (Tirén 1977, Focht 1974)

The above is generally valid also for field conditions meaning that suitable environments should have fairly high organic production or supply of organic material and intensive respiration (incl. ammonification-nitrification) leading to partial oxygen deficiency preferrably at porous floccs or other microsites where bacteria might form dense colonies. According to the "Microzone model" the different favourable environments appears at very close distances e.g. at the in- and outside of an organic particle or floc (Leonardson 1992). The site would further benefit from some water movement, recirculation rather than transit, and raised temperature. There are several indications, however, that sewage work bacteria are to some degree "domesticated"; performing well at their "close to optimum" conditions, whereas bacteria in nature generally perform comparatively better aside of their optimum areas e.g. as regards temperature.

2. Favourable denitrification and retention sites in nature

Looking at retention from a catchment point of view, it is obvious that water discharged at the catchment outlet is a mixture of water with different origins, transport routes and different residence times.

The dominant part of the swedish landscape is forested and the dominance of forested moraine soil is the general rule within watersheds larger than 10-50 km^2. In forested recharge areas nitrogen retention due to denitrification is small within the root zone and upper mineral soil. The major retention component there is permanent storage in the humic layer and the accumulation horizon of podsols. The build-up of humic nitrogen in forest soils since deglaciation 10000 years ago is generally considered to be ca 0,02 - 0,05 g $N*m^{-2}*y^{-1}$, (Rosén 1991). Since the standing biomass of swedish forests is presently also increasing over several forest generations this might also slightly increase the retention in the layers above the ground-water table.

At or below the groundwater table in recharge areas, significant retention due to denitrification may occur if oxygen levels are low (Fustek et al. 1991, Lowrance 1992, Geyer et al. 1992). Significant amounts of dissolved nitrate are, however, rarely transported to these particular areas in the northern 2/3 of Sweden (Fölster 2000). On theoretical grounds denitrification in recharge areas may be expected mainly in the southernmost part or south-westerly part of Sweden where nitrogen deposition is higher than in the rest of the country. If also recharge areas in cultivated watersheds is considered, the conditions favourable for denitrification should be found over extended areas of the country depending on the supply of excess fertilizer and organic material as well as texture and other site-specific conditions.

More favourable conditions for denitrification are on the other hand expected at discharge areas, riparian zones or wetlands (Leonardson 1993, Lovrance 1992, Vought 1994). The aquatic-terrestrial interface is assumed to be generally favourable for denitrification although methodological shortcomings do not always make it possible to distinguish between denitrification and dilution by deeper ground water. Wetlands offer alternating oxygen concentrations, prolonged residence times as compared to streams and extended colonization or reaction surfaces due to abundant vegetation. Although nitrogen retention occurs in oligotrophic wetlands (Jacks et al. 1994, Seitzinger 1994) it is strongly favoured in the richer types of wetlands (Cirmo & McDonell 1977, Christensen et al. 1991, Seitzinger 1994).

Since rich wetlands have almost all the priority properties for nitrogen retention, there has been a great interest in constructed wetlands for wastewater treatment (eg Hammer 1989, Moshiri 1993,Wittgren 1994) and for the reduction of non-point-source pollution (eg Jörgensen et al. 1989, Brüsch & Nilsson 1990, Mitsch 1992, Emmet et al. 1994, Raisin & Mitchell 1995, Fenessy & Cronk 1997).

However, many wetlands in southern Scandinavia also suffer from drawbacks with respect to nitrogen retention. High flow gives high hydraulic load or short residence times especially during late autumn and winter or early spring when temperature and biological activity are low, leading to very low retention. Since nitrate concentrations are often close to maximum under these conditions large amounts of nitrogen passes under unfavourable retention conditions which severely affects nitrogen retention on a yearly basis (Leonardson 1994, Jansson et al. 1986). This does not fit into the pattern of high winter denitrification described for river corridors further south in Europe (Haycock et al. 1993).

Streams also offer favourable denitrification sites especially if the channels contains organic sediments (in contrast to gravel) and rooted vegetation (Seitzinger 1988, Jansson et al. 1986). However, they also suffer from the same drawback as wetlands regarding retention seasonality. Under favourable seasons, denitrification may be quite high (Christensen et al. 1990, Svendsen & Kronvang 1993) but on a yearly basis less than 5% retention (of the total river load) has been reported in Denmark and southernmost Sweden (Svendsen & Kronvang 1993, Jansson et al. 1994). As a general rule, watercourses therefore must be considered to have low retention and primarily serve as transport channels for nitrogen.

In lakes, finally, retention generally is important if water residence times exceeds one or two weeks. Denitrification is usually highly significant due to long water residence time and favourable conditions in the upper sediment strata. Permanent burial of organic nitrogen in sediments frequently occurs to some extent and may be of significance.

The rain of organic particles supplies the energy to the bacteria dwelling at lake bottoms and the respiration causes microzones of low oxygen levels suitable both for denitrification and supply of nitrate through nitrification. Superficially oxidized sediments are considered as the most favourable sites for both processes and denitrification as a whole. The presence of benthic animals, active through bioturbation and the pumping of water through channels and tubes, is also considered to promote denitrification (Graneli 1979, Andersen 1976, Chatarpaul et al. 1980, Svensson 1997). Under completely anoxic conditions nitrate supply ceases and denitrification stops (unless nitrate is supplied from the water mass).

Further favourable conditions in lakes are the frequently large macrophyte belts. Macrophyte root lacunal systems may by diffusion supply oxygen to deeper sediments thereby increasing the zone favourable for nitrification/denitrification (Christensen & Sørensen 1986).

Denitrification is generally low in the water mass of lakes but may be significant in the water mass in lakes with completely anoxic sediments and benthic water. Denitrification then appears in the water mass at the vertical ecocline where a low concentration of oxygen changes into a completely anoxic benthic water and some nitrate appears as well (Chan & Campbell 1980, Brezonik & Lee 1968, Goering & Dugdale 1966).

3. The role of nitrate supply

Nitrate availability obviously is crucial for denitrification and limited supply may limit both rate and final throughput of nitrate to denitrification. At some sites, nitrate supply is brought about mainly by transport from distant sources. At other sites both nitrate production and reduction are co-localised and occur at microdistances. Under such circumstances it may often be difficult to determine which step is rate-limiting. In many instances it appears to be nitrification, though. Nitrification is basically a kemoautotrophic process carried out first from ammonium to nitrite (NO_2^-) by the genus *Nitrosomonas* and second from nitrite to nitrate (NO_3^- by the genus *Nitrobacter*. The bacteria use the energy obtained from oxidation for the synthesis of organic matter, which however is fairly costly, meaning that much ammonia is processed per unit bacterial growth. For the same reason, oxygen consumption is high per unit body growth. In fact a significant part of lake sediment oxygen consumption is derived from nitrification (Seitzinger et al. 1984). It is also a well known fact that un-nitrified sewage water has a high potential for oxygen consumption due to nitrification which adds to the oxygen demand by respiratory processes. The nitrification process thus contributes to deoxygenation and therefore to the creation of suitable microsites for denitrification. It also promotes denitrification by routing ammonium to denitrification contrary to being conserved within the system.

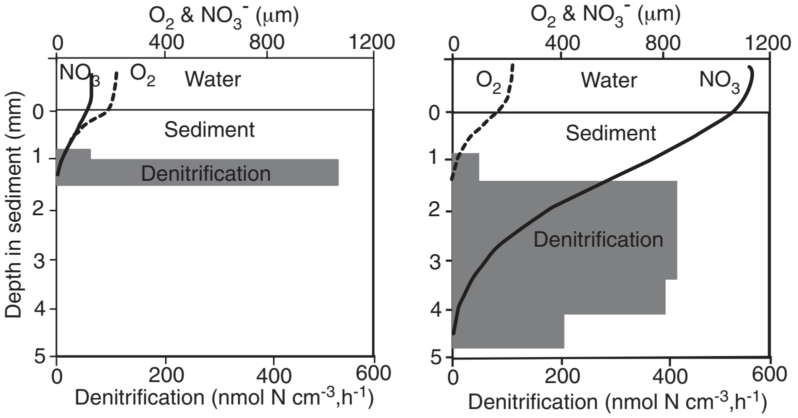


Figure 2. Oxygen and nitrate concentrations at the sediment-water interface in two cores from Århus stream in Denmark supplied with different nitrate concentrations in the water phase. The induced denitrification underlines the limiting role of nitrate and the extreme microzonation. Nitrification (not included in the figure) could proceed in the uppermost mm of the sediment. Redrawn from Christensen et al (1989).

A number of chemoorganotrophic bacteria may also oxidize ammonium but they have lower process rates and are more rare and thus considered unimportant (Fenchel & Blackburn 1979). A brief review of the process conditions for nitrificants shows that:

- ☐ The process is chemoautotrophic and adequate supply of ammonium and carbon dioxide is essential. Typical for the process is low energy gain and body growth as compared to the processed amount of ammonium.
- ☐ Oxygen is necessary for both process steps but the process continues to low oxygen levels (0,02–0,1 mg $O_2 * l^{-1}$ in pure cultures) (Gundersen 1966). The *Nitrobacter* step is more sensitive to low oxygen levels which at times may lead to accumulation of nitrite.

- ❑ The temperature optimum is high, at 25–35°C and growth continues from 3 to 45°C (Vocht & Verstraete 1977). Q10 -values from 2 to more than 3 have been reported.
- ❑ pH-optimum appears between 7 and 8.5 with absolute limits at pH 6 and 9,5 respectively (Vocht & Verstraete 1977). However, in nature a low limit of <pH 6 have been recorded (Schindler et al.. 1985). The upper pH-limit appears to be set by ammonia toxicity.
- ❑ Available surfaces, slow population growth, predation sensitivity and light inhibition are other factors that may regulate nitrification rates in nature.
- ❑ As for denitrificants, bioturbation and burrowing may enhance nitrification in natural sediments. Enhanced nitrification in root zones probably is due to oxygen diffusion from gas vacuoles in the roots.

An example of both favourable site conditions and the importance of nitrate supply is given in figure 2 according to the so called dual-layer model (Christensen et al.. 1989). Denitrification proceeded close below the sediment surface in a 0.7 mm thick layer at normal water concentration of nitrate. When nitrate concentration was raised an extended zone of denitrification was activated showing the high potential ability for denitrification given an adequate supply of nitrate. In this case, the denitrifiction was dependent upon the diffusion of supplied nitrate. When nitrification is significant (of minor importance in the studied cases) it proceeds in the uppermost oxidized mm, which in this case is close enough to compete with nitrate supplied from the water mass if nitrate production is high.

An interesting switch between nitrate sources is also reported from phosphorus rich Danish shallow lakes (Christensen et al. 1991). During winter, nitrate concentrations are high in the circulating waters of the lakes and lake water is suggested as the dominating nitrate source for denitrification. During summer, both nitrate and ammonium may often reach levels below detection limits. Under such conditions nitrogen is brought to the sediments in sedimenting particles, ammonified, nitrified and to a high extent denitrified as shown by mass balance calculations in Lake Søbygård Sø. Nitrogen from direct nitrification thus seems to be the major nitrogen supply for denitrification during summer. This situation is not general, however. In many oligotrophic Swedish lakes nitrate levels of 0,1-0,4 mg NO_3^- N/l persist throughout summer without being denitrified which points to the importance of other important regulatory mechanisms.

Methods for nitrogen retention and denitrification assessment

1. Mass balance

Retention is calculated as the (positive) difference between the total nitrogen input and the loss through the outflow. Measured or calculated input through surface water, ground water, deposition and nitrogen fixation are to be summed.

Since denitrification is determined by difference, the quality of the result is governed by the accuracy of all calculated or assessed terms, which is the most severe and general problem with regard to mass balance calculations.

Two components of the balance, groundwater input and input from the atmosphere through biological fixation, may cause special problems. Generally, groundwater nitrogen concentration has to be approximated. Under Swedish conditions this is a small problem as far as lakes are concerned since lakes generally receive most water through superficial inflows and ground water nitrate concentrations are lower than in superficial water. Ground water nitrogen input may therefore most often be neglected.

Nitrogen fixation, the other problematic term, may be important particularly in eutrophic or hypertrophic environments with an excess phosphorus supply (Howarth et al. 1988, Seitzinger 1988, Hellström 1996). Nitrogen fixation may be assessed in situ by various methods (Howarth et al. 1988) or approximated by counting cyanobacterial heterocyts (the nitrogen-fixing cells) and applying heterocyt-specific nitrogen fixation rates. Lack of data often means that nitrogen fixation is neglected where it should be included.

Mass balance calculations for lakes may be more detailed including also the fractions that are denitrified, buried in sediments and accumulated in the water mass. Phosphorus retention is then also calculated by mass balance and the yearly phosphorus retention in sediments is multiplied by quotients between the nitrogen and phosphorus contents analysed in representative sediments. Denitrification is then estimated by difference:

$$N_2=Q_{in}*C_{in}- Q_{out}*C_{out} - V*C_{diff} - TP_{sed}*TN/TP_{sed}$$

N_2= Denitrification
Q = Water flow
C = Total nitrogen concentration
V = Lake volume
$_{in,\ out}$ = indices for in- and output
C_{diff} = long term concentration change in lake water
TP_{sed}= long term phosphorus accumulation in sediment (from mass balance)
TN/TP_{sed}= Quotient: total nitrogen/total phosphorus as assessed in representative sediments

Direct methods to measure the permanent accumulation of particles or nitrogen in sediments are seldom used with the aim to estimate permanent nitrogen burial. Indirect calculation methods like the mass balance method presented above are More common. Alternatively, dating of sediment horizons in lakes followed by chemical analysis of the specified sediment strata can be used. Calculation of the amount of nitrogen stored during a specified number of years can then be made for representative sediment samples and form an estimate of permanent sediment accumulation of i.e. nitrogen.

2. Dynamic mass balance modelling

Mass balance nitrogen models with high temporal resolution and the possibility to simulate the consequences of changed inputs have become increasingly popular during the last decade. These models frequently include source appointment as an integrated part and find use to pinpoint cost-effective remedial measures. Swedish models of this type and their estimates of nitrogen retention in particular is presented briefly here. Detailed model descriptions are found elsewhere (Arheimer & Brandt 1998, 1999, Sonesten et al. 2003).

Temperature, precipitation, water discharge and nitrogen input (point and diffuse sources) have been used to model nitrogen dynamics of whole watersheds, several coupled watersheds, or landscapes.

The so far most complex model was presented in 1997 (Arheimer et al. 1997, Arheimer & Brandt 1998). The aim was to assess Sweden´s progress towards the 50% nitrogen reduction agreement among states surrounding the Baltic and North Sea.

This study covered the southern part of Sweden (from Stockholm southwards) and involved 3725 subwatersheds and was calibrated at 722 points. The same type of model was later run for 1000 somewhat larger drainage basins covering the whole of Sweden (Brandt & Ejhed 2003). This study supplied data to the "Pollution Load Compilation No 4" within HELCOM. These large studies were based on earlier experiences of modelling at the watershed scale (eg. Brandt 1990, Arheimer &Wittgren 1994). Modelling of N-losses from the root zone of crop land (the SOIL-N-model) constitutes another important component the landscape studies (Johnsson, H. et al. 2002, Johnsson & Hoffman 1997, Johnsson & Mårtensson 2002.).

Each subwatershed was initially treated separately and then joined to others downstream. Water flow was modelled from daily precipitation and temperature data and monthly evapotranspiration constants for different regions (the HBV model, Bergström 1992). Nitrogen transport and retention was calculated with one separate model for inorganic and one model for organic nitrogen, both merged into the HBV-N-model. Local retention in the soil system was calculated separately starting from root zone loss of nitrogen (including other nitrogen inputs to soil). Each ”local unit” consisted of two compartments with slow and rapid turnover respectively. Nitrogen behaviour and losses in downstream watercourses was modelled separately as was also transport, concentration and retention in lakes. Two subunits with different turnover were also used for lakes. The retention expressions contained among other things water flow, concentration, temperature and a calibration factor that was tuned.

In the second of these broad approaches stream and lake retention were treated together since stream retention was found to be very low. These models contained 10 constants to be calibrated but due to the step by step procedure only 3 factors were open to calibration at the same time. A time step of one day was used, although long-term means were presented as results.

Similar, but simpler approaches have also been brought forward in which retention in soil, stream and lakes have been joined, inorganic and organic nitrogen have been merged, compartments with rapid and slow turnover have been joined and the number of calibration factors reduced to two (Kvarnäs 1996, Kvarnäs 1977, Johansson&Kvarnäs 1998, Wallin et al. 2000, Sonesten et al. 2003). The time step was reduced to one month. These models operate with very small watersheds and originally had ca. 50 basins and 11-34 calibration points. In the latest application, on the Göta Älv drainage basin surrounding Lake Vänern, an area corresponding to 10% of the area of Sweden was covered in 1513 subwatersheds and with 204 calibration points (Sonesten et al. 2003).

Results from these modelling efforts are most often published and used as accumulated retention for entire watersheds. However, the nitrogen retention in lakes is available from two of the most significant studies so far and is discussed further on (p.31).

3. Denitrification assessment

Direct assessments of increases of molecular nitrogen in enclosed samples can normally not be used as an estimator of denitrification due to the high initial concentration of molecular nitrogen gas in water. However, in a few studies sediment cores were incubated in gas-tight chambers where the normal pool of gases dissolved in water were replaced by oxygen, carbon dioxide and helium. The evolved dinitrogen gas could then be directly assessed under otherwise semi-natural conditions (Seitzinger 1988, Seitzinger et al. 1993).

According to the most traditional assay technique, the nitrate decline in the water close above sediments is recorded (Andersen 1977, Robinson et al. 1979). Since processes like nitrate assimilation, nitrification and dissimilative reduction to ammonium may interfere, this method would possibly both over- or underestimate denitrification and is thus unsatisfactory by modern standards. The method has been modified by the use of a stable isotope, so that ^{15}N-NO$_3^-$ is added and the production of ^{15}N-N$_2$ followed which would give values with lower underestimation (Tirén et al. 1976, 1977, Chan & Champbell 1980, Nishio et al. 1983). Nitrate derived from nitrification in microzones is not accounted for, however. In an attempt to also include the freshly nitrified nitrate in the assessment, ^{15}N-NH$_4^+$ have been added to the sediment – water system (Nishio et al. 1983, Jenkins & Kemp 1984) and complementary denitrification directly derived from nitrification have been assessed. However, in these types of ^{15}N-assessments there has been a need to add fairly high amounts of labelled chemicals and to run fairly long experiments to find large enough amounts of ^{15}N$_2$ for detection. Since raised concentrations of ammonium and/or nitrate in most cases will enhance denitrification, the outcome of the assessments have been difficult to transfer to in situ conditions. Lately (during the 1990:ies) a technical evolution is giving some hope that the drawbacks of the method may be reduced (Nielsen 1992, Svensson 1997).

In the mid-70ies it was realized that acetylene blocked the reduction of dinitrogen oxide to molecular nitrogen, which led to a gradual accumulation of dinitrogen oxide. In the corresponding acetylene-inhibition assay the produced dinitrogen oxide was detected with high sensitivity in gas chromatographs (Balderston et al. 1976, Chan & Knowles 1979, Yoshinari & Knowles 1979). However, acetylene also inhibits nitrification and a number of other microbial processes (i.e. nitrogen fixation) which may affect assessments where nitrification is an important source of nitrate for denitrification (Walter et al. 1979, Hynes & Knowles 1978). At concentration levels of several mg N/l surrounding the denitrification site (sediment or soil) it is claimed that the supply of nitrate from the solution exceeds the direct supply from nitrification within sediments (Walter et al. 1979) which would make the acetylene inhibition method more feasible.

Another seldom utilized inhibition method makes use of sodium chlorate to inhibit the dissimilative reduction of nitrate. Since dinitrogenoxide gas production then is supposed to be inhibited the decline of the initially present N_2O-gas can be followed by gas chromatography. Since N_2O-gas is also produced in the nitrification process it was also recommended that nitrapyrine be added to inhibit that production (Miller et al. 1986). Since natural levels of N_2O-gas are very low and changes hard to detect even by gas chromatography this method has never been routinely used.

In the early 90:ies, Seitzinger et al. (1993) were able to further elucidate the capabilities of the different direct assessment techniques. In experiments with sediment cores from a Danish lake, a high flow of $^{15}N_2$ from $^{15}NH_4^+$ and a similarly high flow of N_2 into water where N_2 had been replaced with helium showed high rates of coupled nitrification/denitrification. This flow was not detected when the acetylene inhibition method was used and much lower denitrification rates were consequently measured. In addition it was found that acetylene inhibition of the denitrification step $N_2O \rightarrow N_2$ was not complete and resulted in a 50% underestimate. Clear underestimates of total denitrification rates are thus to be expected both from $^{15}NO_3^-$ and acetylene inhibition assays.

Some other denitrification assessment techniques including analysis of gas composition in gas traps above sediments (Jones 1985) and calculation of denitrification from vertical nitrate profiles in sediments (Oren & Blackburn 1979) never gained any widespread application.

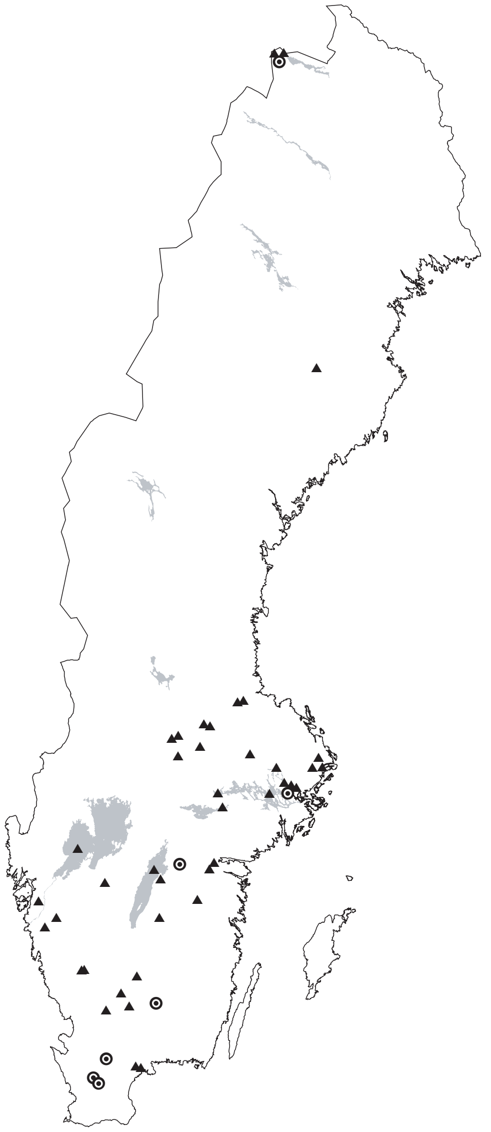


Figure 3. The location within Sweden of lakes from which mass balances (input-output) of nitrogen have been compiled. Triangles give sites from which at least one years´ balance is available. Circles show sites where two cases have been treated, one before nitrogen input reduction and one after input reduction.

The present mass balance survey

1. Material

The history of nitrogen mass balance calculations dates back to 1962 for Swedish lakes (Ahlgren 1967) and denitrification assessments in lakes started 1974 (Tiren et al. 1976). A fair number of mass balances (ca. 25) were calculated during the 70:ies. One major set were calculated for severely polluted lakes for which the phosphorus and organic load were reduced by the introduction of chemical precipitation in the polluting sewage treatment plants. Another significant group of studies derived from basic research. During the 1980:ies and the beginning 90:ies nitrogen reduction in sewage works was high on the agenda. Natural retention in lakes was considered an important aspect affecting the decision whether or not a nitrogen reduction step should be added to the sewage treatment plants. Data for retention calculations were frequently picked from ongoing monitoring programs, many of which were not designed for that purpose, which at times affected the quality of the calculations. During the 90:ies just a few balance studies of research character were performed. Some direct assessments addressing the potential problem of effects on denitrification by acidification were also made.

Mass balance studies from a span of almost 40 years serve here as the primary material to examine nitrogen retention. In total, a number of 62 cases have been gathered from 53 lakes (figure 3). The double (or triple) occurrence of 6 lakes reflects situations with high pollution input followed by decreased input due to introduction of tertiary sewage treatment. Data from a set of 7 created ponds with an extremely high hydraulic load have been excluded in the present compilation. Data may be found elsewhere (Fleischer et al. 1994). The utilized data is found in Appendix 1.

In the compilation of data, the aim has been to gather data with a reasonable quality, as far as that can be discerned. A minimum of ca 75% of the nitrogen load should derive from gauged sources, the rest could rely on reasonable estimates. A major fraction of water discharge should be gauged, not modelled. Outlet water chemistry data should be used for output calculations, chemical data from mid-lake stations were not accepted as substitute. Chemical sampling should be frequent, meaning monthly throughout the

Table 1. Lake characteristics calculated for the 62 cases examined. Internal phosphorus loading leads to "negative" phosphorus retention in 22 cases, Phosphorus data for cases with only positive phosphorus retention are therefore given within brackets (40 lakes representing more normal conditions).

	Mean	Median	Std.err.	Min	Max
Lake area (km^2)	158,2	11,9	95,3	0,014	5650
Mean depth (m)	6,4	4,6	0,86	0,76	40
Residence time (y)	1,92	0,47	0,97	0,008	60
Hydraulic loading (m$*$y^{-1})	16,5	9,2	3,2	0,67	144
Lake surface/drainage area	0,076	0,044	0,01	0,002	0,41
Mean inlet TN conc (μg N$*$l^{-1})	2212	1564	214	132	7784
Mean lake TN conc (μg N$*$l^{-1})	1256	953	119	130	4800
Mean inlet TP conc (μg P$*$l^{-1})	113(124*)	50(116*)	36(51*)	4,5	2160
Mean lake TP conc (μg P$*$l^{-1})	118(86*)	45(29*)	26(29*)	4	1160

year. In some cases with low temporal concentration variability, 6 samplings per year was accepted.

Balances of total nitrogen were the targets in the search for data, implying that other supplementary data were not always of high quality or even available (i.e. input of inorganic nitrogen). This is valid also for inlake N and P concentrations where winter samplings were often rare. Yearly max and min concentrations of inorganic nitrogen therefore in some cases represents approximations. In lakes with dynamic internal phosphorus loading during summer, the samplings may not mirror ambient concentrations correctly.

Rather frequently, water residence times were not specified to the year of assessment but as a mean over a longer period. Obviously, calculated water loadings will then suffer from the same drawback.

At times, changes in storage of nitrogen in the water mass may affect nitrogen retention significantly. Such changes in the water pool of nitrogen were therefore excluded from retention, simulating a situation with no inter-annual concentration changes, termed here "equilibrium" retention. There were 17 cases with information of storage changes. Changes were assumed negligible in the rest of the cases.

A very important aspect when treating these data is the variability between years, which may be quite high. Data from a single year therefore hardly pinpoints the specific character of a certain lake. Consequently the aim was to present mean values for more than three years for each lake, thereby trying to present more typical lake characteristics. Quite frequently, however, only data for one year have been available, and this should be born in mind in the evaluation of general lake behaviour.

The physical characteristics for the studied lakes spans a wide gap; from minimal high-mountain tarns to the largest and deepest Swedish lakes, lakes Vänern and Vättern respectively; from turnover times of a couple of days to 60 years (table 1). Lake volume, lake area and water residence time are well correlated (r>0,94) in a cross correlation. Among the variables describing water flushing (residence time, hydraulic loading, the quotient lake surface/drainage area) there is a weak correlation between residence time and the surface/drainage area ratio (r=0,60) whereas hydraulic loading is not at all correlated to the former two variables.

When it comes to nutrient chemistry a similar wide span in the data is evident (table 1). Phosphorus data for the group of lakes with positive phosphorus retention are given within brackets in table 1, since a high internal phosphorus loading in c. 20 lakes strongly affects and hides the normal retention pattern in the group as a whole.

The data presented are directly comparable to a corresponding compilation from 69 Danish lakes (Jensen et al. 1990). Those lakes are generally smaller (more than one order of magnitude) and more shallow (half the median depth). Residence times are fairly similar, whereas nitrogen concentrations are more than two times higher in the Danish lakes. From Norway a more limited set of 8 lakes is available, characterized primarily by low phosphorus levels (Berge et al. 1997).

2. Nitrogen retention

A wide variation in nitrogen retention between different lakes was seen in the complete material (figure 4). The presented data are ”equilibrium” retentions meaning that changes in the nitrogen pool of the water mass are are neglected in calculations. Ideally these data therefore represent denitrification+permanent sediment accumulation. As seen, the distribution is not even but the dominant part (25-75% percentiles) of the retentions lies in a fairly narrow band between ca 20 and 50% and with a median retention of 40%. Since the corresponding Danish median retention for 69 lakes was 41%, a reasonable general assumption about the fraction retained would then be 20–50% for any lake if nothing else is known.

Expressed in absolute numbers, the retention spans from zero (or slightly negative values) to ca 25 g N*m^{-2},y or expressed in other units 250 kg N*ha^{-1},y (figure 5). This value corresponds to the maximum amount of nitrogen fertilizer applied to Swedish farm crops. The median retention is 4 g N*m^{-2},y. It corresponds to the highest losses of

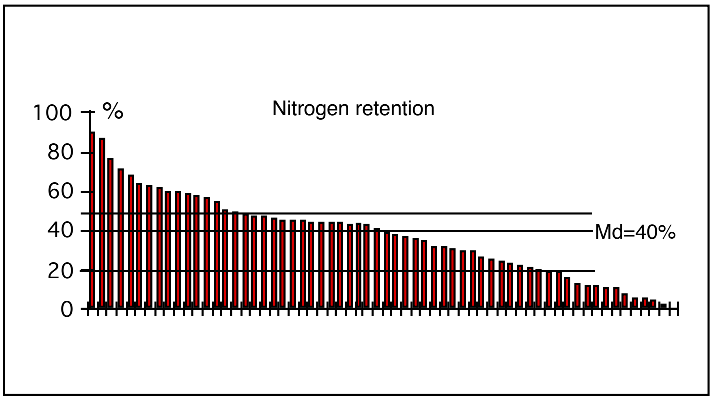


Figure 4. Distribution of relative equilibrium nitrogen retention (retention/input) for the 62 cases studied. 25, 50 and 75% percentiles (lines) correspond to 21, 40 and 49% relative retentions respectively.

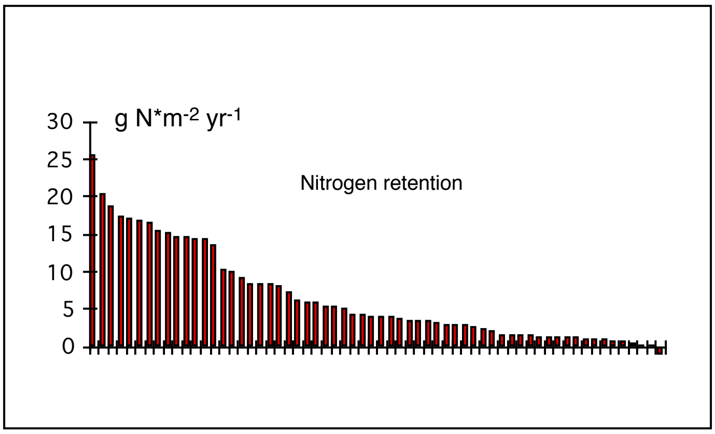


*Figure 5. Distribution of absolute equilibrium retention for the 62 cases studied. 25, 50 and 75% percentiles correspond to 1.6, 4.0 and 9.9 gN*m^{-2},y respectively.*

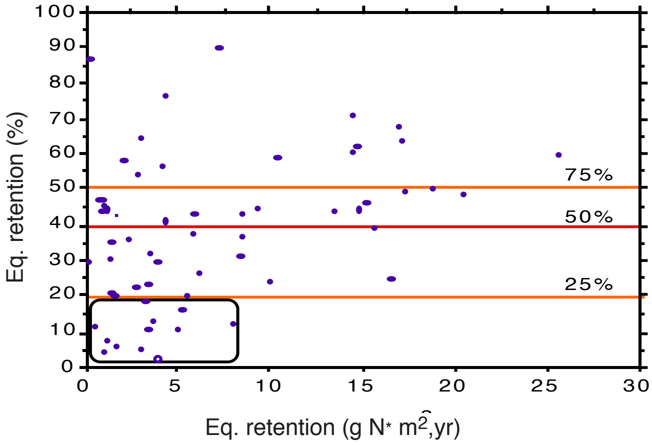


Figure 6. Relative equilibrium retention as a function of absolute retention. Frequency of relative retentions indicated by percentiles (lines). Framed values are the lovest 25% percentile which also have low absolute retention.

nitrogen from farmland or twice the highest nitrogen deposition in Sweden. It is safe to say then, that in general a lake will be able to eliminate the equivalent amount of nitrogen from a heavily polluted area from an at least equal size as the lake. Losses from agricultural areas are generally lower, however, and at losses from farm land corresponding to the national mean (ca 2 kg $N*ha^{-1}*y^{-1}$), a large number of lakes may even eliminate nitrogen corresponding to a five times larger farm land area.

In the corresponding Danish study the mean retention was slightly higher in absolute numbers. However, the median retention was four times the Swedish retention.

3. Absolute vs. relative retention

There is no obvious relation between relative and absolute retention (figure 6). However, it is obvious that the lowest relative retentions coincide with the lowest absolute retentions (framed in figure). Put in another way: high relative retentions are found over the whole spectrum of absolute retentions, whereas no low relative retentions coincide with high absolute retentions. This may be interpreted so that the lowest relative retentions may be limited by some specific factor, but regulation by factors affecting the whole material in some general way may also be responsible.

4. Denitrification vs. retention

For a set of 9 cases denitrification was assessed as a difference using mass balance methods. Nitrogen accumulation in sediments were calculated from phosphorus accumulation and N/P-ratios in the sediments (p. 11) in all but one case. In that case phosphorus was substituted by iron (Ahlgren et al. 1994). Denitrification given as a fraction of retention varied between 54 and 100 % with a mean of 73%. As a fraction of nitrogen input, denitrification varied between 2 and 54% with a mean of 30%. In the corresponding Danish study denitrification was calculated for 58 lakes giving a mean denitrification of 77% of the nitrogen retention (Jensen et al. 1990). Expressed as a fraction of nitrogen input, denitrification accounted for 33%. The corresponding median value was almost similar (30%). The extremely low denitrified fraction of input in one Swedish lake (2%) probably reflected the fact that practically all input nitrogen was organically bound which would strongly reduce the potential for denitrification.

The present compilation also contains in situ or laboratory assessments of denitrification rates in 9 lakes (table 2). They are difficult to extrapolate from site- to lake-representative values and to yearly values. Furthermore, one set of data (Tirén 1977) need correction for enhanced denitrification due to increased $^{15}N\text{-}NO_3$ concentrations in the assessments. Corrected data (0,8 –12,2 g $N*m^{-2}*y^{-1}$) are given in table 2. When the acetylene inhibition method have been used (Gahnström et al. 1993) the immediate supply of nitrate by nitrification is not taken into account. If an arbitrary correction factor of 2 according to Seitzinger et al. (1993) is applied, denitrification rates of 1,5–8 g $N*m^{-2}*y^{-1}$) for littoral bottoms would be obtained. Littoral areas showed higher rates than profundal areas (Gahnström et al. 1993). The lowest denitrification rates (0,3-0,4 g $N*m^{-2}*y^{-1}$) were obtained for the eutrophic or hypertrophic lakes Norrviken and Vallentunasjön where the so far best technique was used. In these two lakes where balance calculations and direct assessments could be compared, denitrification estimates by balance calcula-

Table 2. Direct denitrification assessments in 9 lakes. Assessments were made on undisturbed sediment cores in situ (1) or in the laboratory (2, 3) using three methods. Results obtained by method A correspond to maximum potential denitrification and are given here corrected to ambient conditions. Data according to method C are uncorrected, although a correction factor of ca 2 might be used (Seitzinger et al. 1993). Data sources: 1) Tirén 1977, 2) Ahlgren et al. 1994, 3) Gahnström et al. 1993

Lake	T (y)	Year	DIN-max (μg N*l^{-1})	Tot-P (μg P*l^{-1})	Denitrification (g*m^{-2}*y^{-1})	Method	Ref.
Botjärn	0,3	1972-73	170	14	0,77	A	1
Erken	6,3	1975-77	350	20	1,20	A	1
Norrviken	0,9	1993	1750	210	0,40	B	2
Norrviken	1,5	1974	950	100	8,50	A	1
Vallentunasjön	2	1993	530	100	0,30	B	2
Ramsjön	0,2	1975	2000	300	12,20	A	1
Gaffeln, littoral		1988-89	140	7	0,73	C	3
Gårdsjön, littoral	3,2	1988-89	210	5	1,83	C	3
Stora Skärsjön, litt.	1,1	1988-89	280	13	4,02	C	3
Attavarasjön, littoral		1988-89	270	19	2,45	C	3

A) ^{15}N-NO$_3$ in, ^{15}N-N$_2$ out, high conc.
B) ^{15}N dilution and isotope pairing (Nielsen 1992)
C) Acetylene inhibition method

tions were about two times higher than those derived from assessments. Similar differences have been reported from other studies (Seitzinger 1988).

In figure 7 the fate of external nitrogen input is described for a sample of lakes for which complete balances are available. Total retention obviously varies when given as a fraction of total input. If just the fractions denitrified and accumulated in the sediment are compared they are evidently fairly constant fractions of the equilibrium retention. If, on the other hand, accumulation in lake water is included in the description and analysis of retention, fewer general relationships can be expected. This has been a major reason to exclude nitrogen accumulation in lake water from the analysis of retention. By doing so, the target is set on denitrification despite that the balance calculations at best give equilibrium retention.

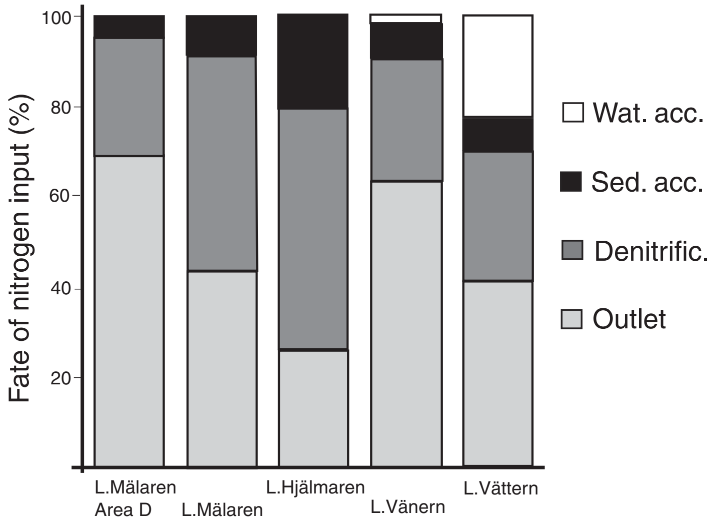


Figure 7 The fate of external nitrogen input in the four largest lakes of Sweden (L. Vänern, L. Vättern, L. Mälaren and L. Hjälmaren) and a deeper than average subbasin of L. Mälaren. Data gathered from mass balances (cf. p. 11) based on input, output and lake monitoring over many (>5) years and sediment analyses of nitrogen and phosphorus.

5. *Nitrogen fixation*

In many lakes nitrogen fixation is a significant source of bound nitrogen. This may add nitrogen to the outflow water of the lake, thereby seemingly decreasing the retention if just external sources are considered. Nitrogen fixation is, however, seldom assessed or calculated. In the present compilation some kind of information on nitrogen fixation was provided in 15 cases but the aim was to discriminate between all cases with or without nitrogen fixation.

Chemical criteria may then be used to indicate the possible occurrence of nitrogen fixation. The most commonly used indication is the in-lake tot N/tot P concentration quotient. It indicates nitrogen deficiency in relation to phosphorus, which offers competition advantages for nitrogen fixing cyanobacteria. This index has been tested in Swedish lakes and indicates nitrogen deficiency when below 10 (or rather an interval 10-17) as a mean during the period June-September (Forsberg et al. 1978).

Hellström (1996) argued that it may be advantageous to calculate this quotient on inflow water that is not affected by the compensatory nitrogen enrichment by the nitrogen fixers in the lake. In the present set of data, several lakes have a high internal phosphorus loading which affects the potential for nitrogen fixation and which is not mirrored in inflow N/P-ratios. Inlake N/P ratios have therefore been preferred.

Morris and Lewis (1988) suggested the ratio dissolved inorganic nitrogen (DIN)/tot-P concentration as a nitrogen deficiency indicator and used values below 0,5 (or rather an interval 0,5-4) as a criterium for nitrogen limitation. Finally, another simple but more crude criterium may be that insignificant DIN is left in the water mass during late summer. Since the sampling frequency is low in the present material (monthly samples) concentration dips are easily missed. Nitrogen deficiency is therefore primarily considered to appear at assessed DIN concentrations lower than 30 µg N/l.

Three indications of nitrogen deficiency were thus used to discriminate between lakes with possible nitrogen fixation and others. The in-lake tot-N/tot-P-quotient was used as a first criterion available from 56 cases (short time fertilization experiments deleted), of which 26 showed quotients lower or equal to 17. The min DIN/tot-P-quotient during summer was available for 44 cases and fell below 4 in all the above cases with low (≤17) tot-N/tot-P and for an additional 11 cases. Assessments indicating minimum DIN concentrations during summer were available for 44 cases and values lower than 30 µg N/l coincided with low tot-N/tot-P-quotients in 16 cases and did not coincide in another 12 cases.

All cases with coinciding indications by all three methods formed the nucleus in a subset of cases where nitrogen fixation could be possible. Cases with measured nitrogen fixation also were assigned to this group. Cases with low tot-N/tot-P-quotients but higher than 30 µg N/l minimum DIN during summer was added to this nucleus, on the grounds that monthly assessments could easily miss concentration dips. Finally some cases with a proven low minimum-DIN but high tot-N/tot-P-quotients were added to the list that ended up with 30 cases where nitrogen fixation might occur. Phytoplankton data from these lakes are rare and a check for the presence of nitrogen-fixing planktonic cyanobacteria did not add any additional information. The stepwise selection method used is considered to include the maximum number of cases, the number of real nitrogen fixation cases most probably falls below the sum of 30 potential cases. In the few cases where nitrogen fixation was measured , it approached 20-30% of the external input in four cases, and consequently must be considered significant.

6. Differences between high- and low-retention lakes

The relative retentions were used to find typical characteristics of high and low-retention lakes. Means of different characteristics were formed for the lower 25% and the upper 25% of the relative retentions as shown in table 3.

Data for the two presented groups give some clues as to the regulation of nitrogen retention. Initially it should be noted that the low-retention lakes typically have higher water flushings compared to the upper quartile lakes. This is well shown by hydraulic loading and related variables. This behaviour follows theory and general experience. It also explains why the low-retention lakes have combined lower inflow nitrogen concentration (=potential concentration) and higher area-specific nitrogen loading by weight compared to the upper quartile lakes. As a group, the low retention lakes have higher external phosphorus supply both calculated by weight and concentration. They also have higher internal phosphorus supply due to low (negative!) phosphorus retention caused by leakage from the sediments.

The richer phosphorus supply to the low-retention lakes may indicate the occurrence of nitrogen fixation that would contribute to an apparently low retention. On the other hand, a richer phosphorus supply would also indicate a richer supply of organic material potentially promoting denitrification and retention. This is, however contradicted by the found low retention.

When the above criteria for classification of nitrogen fixation are applied, 7 cases (of 16) are found in the low-retention quartile and 8 cases in the upper quartile. If these cases are excluded from the respective group and means recalculated for variables according to table 3 the nutrient supply is generally lower in the new subgroups. In particular, the phosphorus supply and concentration is strongly reduced in the new low retention subgroup to levels similar or lower than for the new high retention subgroup.

Table 3. Characteristics for the lowest 25% percentile and for the highest 25% percentile of relative retentions. Means are given for the two quartiles.

Subset	Mean, low 25%	Mean, high 25%
TN retention (%)	11	65
Lake area (km^2)	21	64
Mean depth(m)	7.3	4.3
Residence time (y)	0.43	0.96
Hydraualic loading ($m^3{\ast}m^{-2}{\ast}y^{-1}$)	41.0	6.9
Lake/watershed area (%)	0.018	0.089
TP-loading ($g\ P{\ast}m^{-2}{\ast}y^{-1}$)	1.88	0.38
Potential TP conc. ($\mu g\ P{\ast}l^{-1}$)	184	66
TP-retention (%)	-4.2	29
TP conc. ($\mu g\ P{\ast}l^{-1}$)	138	88
TN-loading ($g\ N{\ast}m^{-2}{\ast}y^{-1}$)	43.2	16,8
Potential TN conc. ($\mu g\ N{\ast}l^{-1}$)	1542	2900
TN output ($g\ N{\ast}m^{-2}{\ast}y^{-1}$)	40	7
TN-retention ($g\ N{\ast}m^{-2}{\ast}y^{-1}$)	3.0	9.9
TN konc ($\mu g\ N{\ast}l^{-1}$)	1310	1180
TN/TP (conc.)	28	23
Pot. TN/pot. TP	31	71

The examination indicates that low apparent nitrogen retention in lakes (and possibly also high retention) does not have one or two single causes like high water flushing + nitrogen fixation but that lakes in the low respective high retention quartiles have heterogeneous characteristics and the reasons for low or high retention may vary between individual cases.

7. Regression models; available factors and approach

The set of factors possibly affecting nitrogen retention, and included in the data set have been grouped:

- ❏. Morfometric: lake area, mean depth, max depth,volume
- ❏ Hydrographic: residence time, drainage area/lake area, hydraulic load (m/y)
- ❏ Nitrogen factors: TN-loading, TN-output, potential TN-conc., TN-conc., max DIN-conc, mean DIN-conc., min DIN-conc., denitrification, N-fixation
- ❏ Phosphorus factors: TP-loading, TP-retention, TP-output, potential TP-conc., TP-conc., TN/TP (loading), TN/TP (conc), TN (loading)/TP (conc)

Several factors are interrelated. All morfometric factors and residence time thus form a particularly strong cluster (r^2=0,71). Several of the nutrient factors form a more loose cluster including TP, TN, max-DIN and mean-DIN as far as in-lake concentrations or calculated outlet concentrations are concerned. Nitrogen and phosphorus inputs and potential concentrations are more loosely coupled to that cluster and phosphorus supply or availability is better mirrored by in-lake or output concentrations (cf above). The same is true for N/P-quotients where in-lake or output quotients still deviate from input quotients. Unfortunately the input of organic matter could not be included due to lack of data.

The most obvious governing factor for nitrogen retention in absolute terms is the input of nitrogen which is used here in regressions. Other nitrogen or phosphorus terms were tested as alternatives but rejected due to poor fit. This means that there is no simple model based only on easily and frequently available assessments like concentrations. Water discharge and transports have to be known.

Representatives for the above four groups were then successively tested in multiple regressions in addition to the nitrogen input term. The aim was to find out the possible role of the group in general. The same procedure was then repeated using transformed values.

8. The role of nitrogen input and concentration

Nitrogen retention was initially plotted as a function of nitrogen input. The plotted data indicated 2 outliers (lakes Aspen and Björkarn) with extremely low (or negative) retention (figure 8A). Since they strongly deviate and have extremely rapid flushing they were deleted in the curve fitting. The fit of the rest of the untransformed data to a linear model gave the equation:

$N_{ret} = 2.0 + 0,24*N_{load}$, *r=0,62, p=0,0001, n=60*

N_{load} = *total nitrogen input* ($gN*m^{-2},y$)

N_{ret} = *corresponding nitrogen retention* ($gN*m^{-2},y$)

In figure 8B the earlier computed median retention of ca 40% (cf figure 4) was inserted

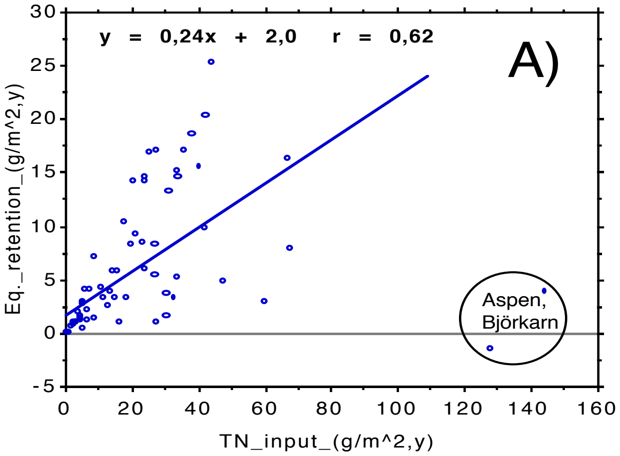


Figure 8 A. Linear fit of equilibrium nitrogen retention as a function of nitrogen input. Two outliers (Lakes Aspen and Björkarn) with extremely low (or negative) retention were deleted from the fitting. They both have extremely rapid flushing.

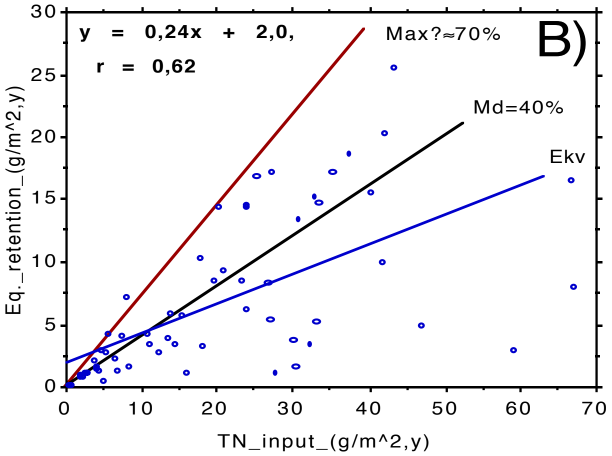


Figure 8 B. Median and hypothetical maximum denitrification added to data according to figure 8 A. Note different X-axis.

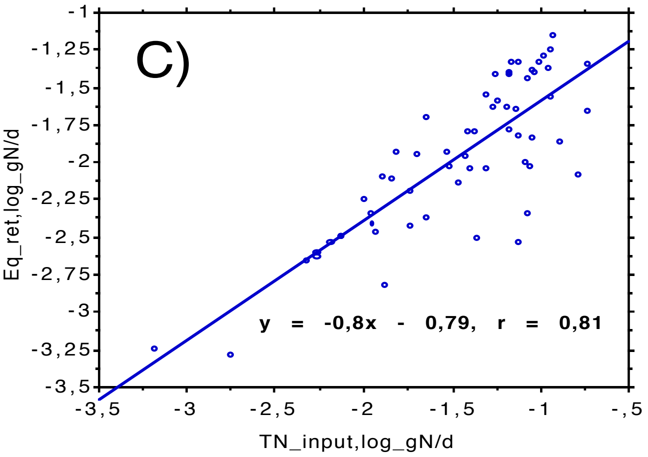


Figure 8 C. Linear fit of log-transformed data according to figure 8 B.

as well as an empirical maximum retention of ca 70%, which seldom is exceeded in the present material. The material also suggests that this limit is not approached at higher loadings.

Similar fitting of the logarithmically transformed data (figure 8C) gave:

$log(N_{ret}) = -0.8 + 0{,}79*log(N_{load}), r=0{,}81, p=0{,}0001, n=60$

$log(N_{load})$ = *total daily nitrogen input;* $log(gN*m^{-2},d)$

$log(N_{ret})$ = *corresponding daily nitrogen retention;* $log(gN*m^{-2},d)$

Data were in this case expressed on a daily basis to facilitate comparisons with other data (p.28)

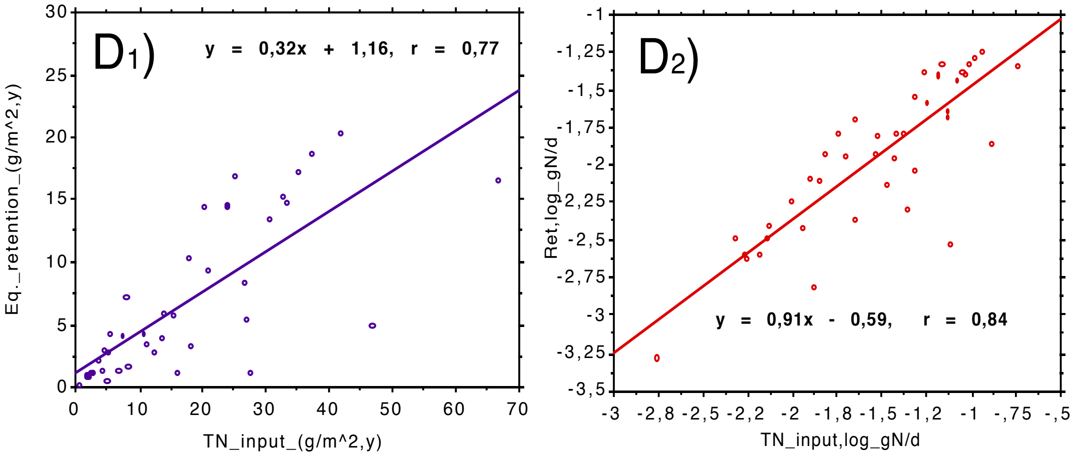


Figure 9. D_1. Linear fit of data from lakes with water turnover time >0,25 y. D_2 :Fit of the same data, log-transformed.

Since water flushing is so decisive a better fit may be obtained if only lakes with higher residence time are considered (figure 9D). For lakes with residence times >0,25 y the corresponding equations are:

N_{ret} = 1,16 + 0,32*N_{load} , r=0,77, p=0,0001, n=42
log(N_{ret}) = -0.59 +0,91*log(N_{load}), r=0.84, p=0,0001, n=42

Hellström (1996) suggested an even longer time for the processes to approach equilibrium. His restriction to lakes with residence times > 1,5 y did improve the fit of the corresponding subset in the present material but the number of cases included was greatly reduced:

N_{ret} = -0,001+0,52 *N_{load}, r=0,82, p=0,002, n=11
log(N_{ret}) = -0.21 –1.06*log(N_{load}), r=0.88, p=0,0003, n=11

Since a low nitrogen retention appeared in many lakes with low or negative phosphorus retention, the fit for a subset, including only lakes with phosphorus retention >5%, was calculated:

N_{ret} = 1,53 + 0,27*N_{load}, r=0,71, p=0,0001, n=39
log(N_{ret}) = -0.75 –0,81*log(N_{load}), r=0.83, p=0,0003, n=39

The subset showed similar coefficients as the complete set but the fit was improved, particularly for the non-transformed data.
Nitrogen retention in the remaining 18 cases of the set (the cases with high internal phosphorus loading) showed a positive but insignificant regression on nitrogen input. In these cases the effect of nitrogen input may thus be overruled by other factors.
When the material was split according to phosphorus level, a better fit was found for phosphorus-rich lakes than for poorer lakes. The slope was also typically steeper for the phosphorus rich lakes (>50 µg P/l):

N_{ret} = 1.6 + 0,34 *N_{load}, r=0,74, p=0,0001, n=27
log(N_{ret}) = -0,49 + 0,95*log(N_{load}), r=0.85, p=0,0001, n=27

The dataset was also split in two groups without and with a potential for nitrogen fixation according to the above criteria (p. 20). The fitting within the groups gave poorer fit for the non-nitrogen-fixing cases as compared to the potentially fixing cases, however. The equations for non-fixing cases were:

$N_{ret} = 2{,}93 + 0{,}12 * N_{load}$, r=0,41, p=0,001, n=25
$\log(N_{ret}) = -1{,}12 + 0{,}61*\log(N_{load})$, r=0.61, p=0,043, n=25

The equations for cases with potential nitrogen fixation were:

$N_{ret} = 2{,}2 + 0{,}26 * N_{load}$, r=0,63, p=0,0001, n=32
$\log(N_{ret}) = -0{,}69 + 0{,}85*\log(N_{load})$, r=0.88, p=0,043, n=32

It was concluded that nitrogen retention was not generally lower in the cases with potential nitrogen fixation as was hypothesised above. The differences indicated by the equations were rather in the opposite direction although differences were not significant due to poor fitting.
With the fairly high inter-annual variation in mind it was also tested if variation was reduced if means of more than 5 years was exclusively used in a subset:

$N_{ret} = 3{,}4 + 0{,}04 * N_{load}$, r=0,74, p=0,0004, n=18
$\log(N_{ret}) = -0{,}98 + 0{,}69*\log(N_{load})$, r=0.82, p=0,0001, n=18

As seen, the long-term means did not specially improve the fitting to these equations.

9. The role of water flushing and other physical factors

In a multiple regression some of the flushing factors (residence time, hydraulic load, and lake/drainage area) were tested in addition to the nitrogen input term. The hydraulic load ($m*y^{-1}$) contributed most to explain the residual variance both for untransformed and logarithmically transformed data and was chosen as the hydraulic factor:

$N_{ret} = 2.6 + 0{,}34*N_{load} - 0.21*Hy_{load}$, r=0,76, p=0,0001, n=60
$\log(N_{ret}) = -0.01 + 1.04*\log(N_{load}) - 0{,}48*\log(Hy_{load})$, r=0.88, p=0,0001, n=60

Hy_{load} = hydraulic load ($m^3*m^{-2}y$)

When different subsets of data were fitted to the two-variable regression following the above scheme, the r-values were generally 0,05-0,10 higher for all subsets. Interestingly enough the exclusion of lakes with turnover times <0,25 y improved the fit also when a

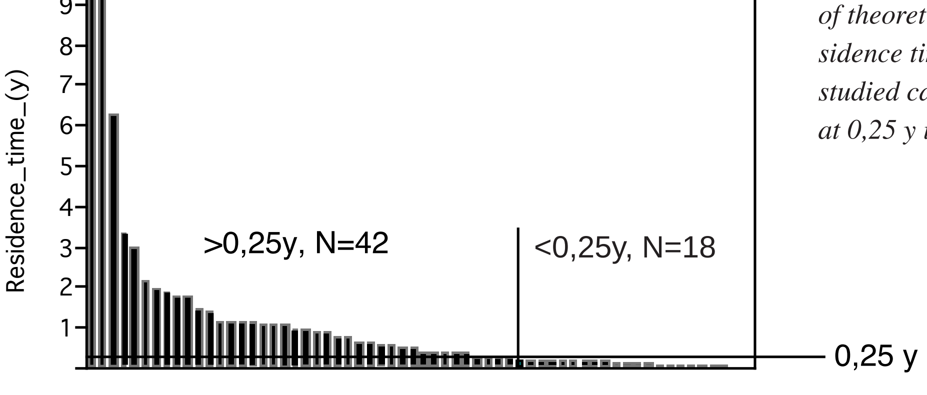


Figure 10. Distribution of theoretical water residence times of the studied cases. Cut-off at 0,25 y indicated.

flushing factor was included in the equation. This indicates that the deviating behavior of some cases with rapid flushing may not be due to the rapid flushing *per se* or have another form of dependence. Figure 10 gives the turnover times for the cases and the cut-off used. The equations for cases with T_W>0,25 y were:

$$Nret = 2,0 + 0,36*Nload - 0.16*Hyload, r=0,83, p=0,0001, n=42$$
$$\log(Nret) = 0,21 + 1.17*\log(Nload)- 0,52*\log(Hyload), r=0.90, p=0,0001, n=42$$

The fit also particularly improved when a subset of cases with "normal" phosphorus retention (>5% retention) was fitted to the two-variable model. Cases with high internal phosphorus loading were thus excluded from the model:

$$Nret = 2,44 + 0,32*Nload - 0.16*Hyload, r=0,81, p=0,0001, n=39$$
$$\log(Nret) = 0,02 + 1.04*\log(Nload)- 0,51*\log(Hyload), r=0.90, p=0,0001, n=39$$

The equations for this set are very similar to the ones for cases with turnover > 0,25 y, in particular for the untransformed data.

Other common methods to estimate in-lake concentration as a function of nitrogen input and water flushing are found among the so called Vollenweider models. Originally Vollenweider (1975) proposed that in-lake concentration was best described by inlet concentration on which a flushing correction was applied. The correction term contained the square root of the water residence time, where the square root may be considered as a transformation. Vollenweider proposed its use both for phosphorus and nitrogen. This model design can also be used for indirect retention estimates:

$$Nret = 0,64 + 0,82*(Nload - Nload/(1+\sqrt{Tw})), r=0,71, p=0,0001, n=60$$
$$\log(Nret) = -0,03+0,89*\log((Nload - Nload/1+\sqrt{Tw})), r=0,83, p=0,0001, n=60$$
$$Tw = \text{water residence time (y)}$$

Also in this case the fitting of different subsets of data to the model lowered the residual variance. A subset of cases with T_w>0,25y gave:

$$Nret = 0,40 + 0,83*(Nload - Nload/(1+\sqrt{Tw})), r=0,79, p=0,0001, n=43$$
$$\log(Nret) = -0,13+1,00*\log(Nload - Nload/(1+\sqrt{Tw})), r=0,85, p=0,0001, n=43$$

In other subsets tested according to the previous scheme the fit was not improved as compared to the complete set, but rather supported the previous conclusions. As a whole, these fittings following the modified Vollenweider model did not explain more of the variance as compared to the above two-variable fittings.

Yet another correction term for water flushing was used by Hellström (1966). He calculated a theoretical concentration increase by using water renewal time and mean depth (actually the square root of mean depth) and fitted retention data to nitrogen load and this variable. The present set of data treated in the same way gave the equation:

$$Nret= 0,82+2,3/((1+1/Tw)*\sqrt{z}), r= 0,74, p=0,0001, n=56$$

This treatment had similarities with the Vollenweider models but showed a poorer fit to the present set of data.

Additional physical variables like mean depth and lake area were added to the above three types of equations containing flushing variables. No further explanation of the variance could be obtained, however.

10. The role of phosphorus and organic supply

Phosphorus may be considered as an indicator of the supply of organic matter to the sediments thereby regulating denitrification. In cases with excess nitrogen (and no potential for nitrogen fixation), nitrogen retention (an estimator of denitrification) may therefore be regulated by phosphorus factors.

In multiple regressions with nitrogen input, hydraulic load and one of several P-factors (cf. above) no phosphorus term was found to be significant when the complete set of data was used (untransformed or log-transformed). Since P-dependence is supposed to interplay with nitrogen fixation, the inclusion of P-factors was also tested on subsets with and without potential for nitrogen fixation. A positive P-dependence might be expected in non-nitrogen fixing cases and a negative otherwise. However, no significance for a phosphorus term was found for these two sets either.

On the other hand, significance was found for equations without a water-loading term, only with nitrogen input and phosphorus terms. This was found for the whole set and the non-nitrogen fixing subset, but not for the subset with potential nitrogen fixing.

Whole set:

$\log N_{ret} = -1.28 + 0,69 * \log(N_{load}) + 0.20 * \log(P_{conc},)$ *r=0,84, p=0,0001, n=58*

Non-fixing subset:

$\log N_{ret} = -1.88 + 0,48 * \log(N_{load}) + 0.43 * \log(P_{conc},)$ *r=0,71, p=0,001, n=29*

P_{conc} = *inlake phosphorus conc (µgP/l)*

It is important to note that total phosphorus apparently affects nitrogen retention positively in these cases, as was expected if the phosphorus level regulates organic supply and denitrification. When N_{load} was omitted, the equation was insignificant.

Further fitting of subgroup data to equations with a phosphorus factor present and with or without water loading and gave very few significances (table 4)

When data from the subgroup of lakes with phosphorus concentrations > 50 µg P/l was fitted to equations with nitrogen input, hydraulic load and one P factor, some P-terms were found significant. The same was true when just nitrogen and phosphorus terms were used. In all these cases it was noted that the P concentration had a negative

Table 4. Significance (yes/no with cut-off at p=0,05) of phosphorus factors (TP-loading, TP conc.,TP output, TN/TP loading and TN/TP conc.) in multiple regressions including nitrogen loading, water loading and a P-term (Nload+Hyload+P-fakt) or only nitrogen loading, and a P-factor (Nload+Pfact). In case of significance the slope for the P-factor is indicated (pos/neg). Subsets of data according to text.

Data set	N_{load}+Hy_{load}+P-fakt	Slope	N_{load}+Pfact	Slope
Total set	no		yes	pos
N-fix	no		no	
No N-fix	no		yes	pos
>50 µgP/l	yes	neg	yes	neg
>50 µgP/l	no		no	
>0,25 y	no		no	
<0,25 y	no		no	
>5% P ret.	no		yes	pos

influence on nitrogen retention. From this it appears that at a given nitrogen loading a higher phosphorus concentration gives a lower N retention in lakes with higher than average phosphorus concentration. Another subgroup characterized by >5% phosphorus retention (which corresponds to"normal or high" phosphorus retention) behaved similarly. However, in this case a significant equation was obtained without the water-loading term.

From this it appears that there is no simple positive dependence of nitrogen retention on the phosphorus level. There may also be an optimum (unidentified here) over which the phosphorus dependence appears to change to negative

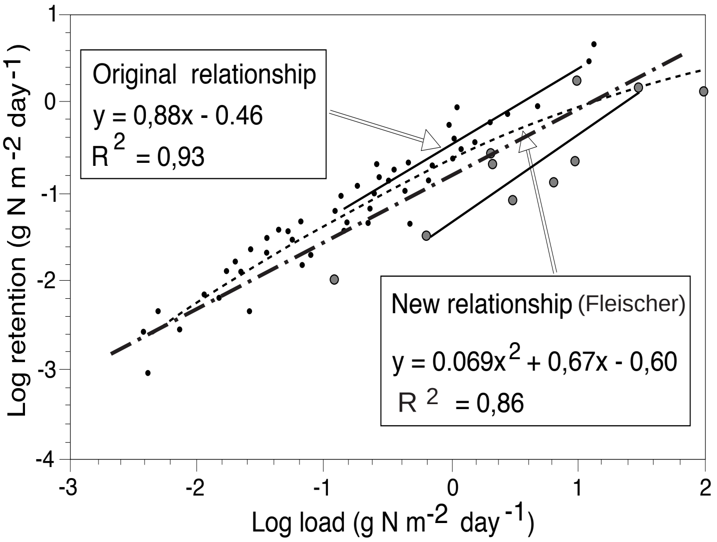


Figure 11. The relation between nitrogen input and retention found by Fleischer & Stibe 1991 (upper line) and their added pond-data as of 1994 (lowest line). The dotted line gives the combination of their data (Redrawn from Fleischer et al. 1994). The relation found in this paper is given as a dot-broken line

Discussion

1. Statistical analysis of equilibrium retention

Four groups of potentially important factors for nitrogen retention were successively tested in multiple regressions. Among nitrogen factors the nitrogen input explained less than half of the variance (r^2=0,4). If logarithmically transformed data were used 2/3 (r^2=0,66) of the variance was explained.

The equations derived from logarithmic data may be directly compared with a corresponding equation for 50 wetlands and lakes in general derived by Fleischer & Stibe (1991). Data came from a variety of environments: lakes (19), constructed reed beds (12), drainage basins (7) and natural wetlands (12). The equation for their combined material was:

$$log(Nret) = -0.46 + 0,88*log(Nload), r=0,97, n=50$$

A higher intercept and higher slope than for the complete present material was thus obtained. In a later paper Fleischer et al. (1994) added data from another 7 constructed ponds. The new data increased the intercept, decreased the slope and reduced the goodness of fit. The fitted function for the present data fell very close to the linepresented by Fleischer et al. 1994 (figure 11).

The Danish study referred to above (Jensen et al. 1990) indirectly describes retention as a function of input by using in-lake concentration as a function of inflow concentration. Inlet concentration explained ca 50% of the variance.

Hellström (1996) considered total nitrogen load, in-lake nitrogen concentration and outlet nitrogen concentration in his analysis of nitrogen retention (or rather nitrogen balance, ΔN) in, at the most, 44 temperate lakes, 10 of which were Swedish. He reported no significant correlation with any of the factors but "a certain degree of correlation with nitrogen load".

The fairly low explanatory power of nitrogen factors alone in the present set of data thus is not surprising seen in the light of other similar attempts. It appears that Fleischer & Stibe's (1991) first attempt was remarkably sucessful (cf fig 11).

Generally, additional factors will have to be considered to explain a larger fraction of the variance. Next in turn were factors related to water flushing and lake morphometry. For the complete present material, multiple regressions with both nitrogen load and hydraulic load increased the explained variance by ca 18% (r^2=0,58) and for logarithmically transformed data to more than 3/4 of the total variance (r^2=0,77). Mean depth was also included in the model but, like all indicators of lake morphometry, seemingly did not affect nitrogen retention.

Further tests on subsets of data also showed that exclusion of lakes with water residence times <0,25 y improved the fit to r^2 values of 0.69 for untransformed data and 0.81 for log-transformed data.

Compared to models with other approaches to consider water flushing this is a high figure. The use of a Vollenweider type of equation with the square root of water turnover time as the flushing factor gave a poorer fit. So did also Hellström's (1996) approach with both water turnover time and mean depth included in the model.

In the Danish study, cited above, the inclusion of water turnover time and mean depth in addition to nitrogen loading in the model of in-lake nitrogen concentration explained less than 2/3 of the total variance. Hellström, in his paper, did not state the r^2 value when nitrogen retention (or rather ΔN) was modelled as proportional to his flushing term. He stated that "the nitrogen balance was found to correlate rather well with this expression".

The final group of factors considered here are indicators of organic loading, primarily phosphorus or N/P-factors. Hellström (op cit) argued for the decisive role of organic supply (or phosphorus) for the nitrogen balance. Using a restricted set of data (8 out of 44 cases) from lakes without observations of cyanobacteria (=no nitrogen fixation) he found no dependence at all on nitrogen input but a strong dependence on total phosphorus concentration and input:

In lakes with cyanobacteria (= presumed or measured nitrogen fixation), the role of total phosphorus changed from being an indicator of organic supply with positive impact on ΔN to be a trigger of nitrogen fixation with apparently inverse impact on ΔN. Nitrogen retention (as estimated by ΔN) may therefore seemingly decrease at higher phosphorus levels giving an inverse relationship to phosphorus. This was the case in lakes with cyanobacteria with the additional condition of long water residence time (>1,5 yr).

Berge et al. (1997) also hypothesized that nitrogen retention under conditions with high N/P-ratio and nitrate accumulation would be low due to low organic supply or production. At medium N/P-ratios, which was supposed to coincide with eutrophic conditions, a high nitrogen utilization was supposed to give optimum nitrogen retention. At low N/P- ratios and high-productive conditions nitrogen fixation would contribute to a seemingly low retention.

The present set of data did not show the same clear-cut dependences of phosphorus factors as stated by Hellström. Yet the principle to separate cases with and without potential of nitrogen fixation was similar and expanded in the present analysis. When the hydraulic load was excluded in the equation based on the subset with non-nitrogen fixing cases a significant and positive dependence on phosphorus concentration was obtained (table 4). A corresponding negative (or positive) dependence was not found for the subset with nitrogen-fixing cases. Subsets with high or low phosphorus concentrations were also tested. In cases with > $50 \mu g\ P*l^{-1}$ in-lake phosphorus concentration at least one phosphorus factor contributed significantly to the explained variance and did so even if the flushing correction was omitted. In this set of more phosphorus-rich lakes the phosphorus factor had a negative influence on nitrogen retention. On the other hand no significance for any phosphorus factor was found in low-phosphorus lakes. This obviously gives no clear-cut answer as to the role of phosphorus in nitrogen regulation. The shift from a positive dependence at low trophic level to an apparent negative dependence at high could just possibly be discerned in the material.

2. Physiological analysis of denitrification

Most methods for the direct assessment of nitrification have drawbacks (cf p. 13). In spite of this, the denitrification rates assessed in 9 lakes fits into the ranges of denitrification found by mass balance (table 2). The records span from 0.8 to 12 g $N*m^{-2}$, y after corrections are applied. They thus spans from below the 25% percentile obtained in mass balances to above the 75% percentile (table 5). The thorough study by Ahlgren et al. 1994 suggests that the physiological assessment of denitrification underestimates, by a factor ca 2, the denitrification estimated by mass balance for the same period.

In spite of this, the best physiological methods (cf Ahlgren et al. 1994) are very useful for studies of spatial and temporal variability. Useful insight into the governing factors may thereby be gained. In this respect the Swedish studies have pointed out the epilimnetic bottoms as very important denitrification sites. Even during autumn, winter and spring, i.e. in cold water, denitrification may proceed there at fairly high rates. These rates may occasionally be higher during summer but expected clear-cut increases due to raised temperature during summer are seldom seen (Gahnström et al. 1993, Ahlgren et al. 1994). This is an information of particular interest since the retention models cited here utilize temperature functions to describe daily or monthly nitrogen retention. Peaks will thus be modelled for summer situations despite the fact that four out of six studied lakes did not show summer peaks in denitrification. Although the additional retention component – net sedimentation – may be highest during summer, these data certainly deserve attention in modelling work.

The generally lower denitrification in the hypolimnion appears to be less an effect of low temperature but more an effect of low nitrate access or low nitrification. The redox potential thus seems to stay low enough to severely retard nitrification. This is important since nitrate production may be the rate limiting mechanism for denitrification in lakes with low nitrate levels. It also suggests that lakes with potential for nitrogen fixation (low access to inorganic nitrogen as compared to available phosphorus) also may have unusually low nitrification and denitrification, particularly if they have anoxic hypolimnia.

3. Retention estimates by dynamic modelling

Results from the above modelling efforts (p. 12) are mostly published and used as the summed retention for complete watersheds. For the whole area of southern Sweden, for instance, a total retention of 45% of the gross load was computed (Arheimer & Brandt 1998).

However, the retention in individual lakes is available from two of the most significant studies so far (p.12). Arheimer & Brandt (1998) give absolute retentions (kg N ha^{-1}, y^{-1}) for lakes in 1637 subbasins in southern Sweden. This is a sample of all lakes in the area but the selection principles and representativity are not stated.

Their retention data have been redrawn here and compared to the compiled measured retentions (figure 12). The distribution of data up to the median values is fairly similar. Higher values, up to the maximum, are more evenly distributed for the compiled data

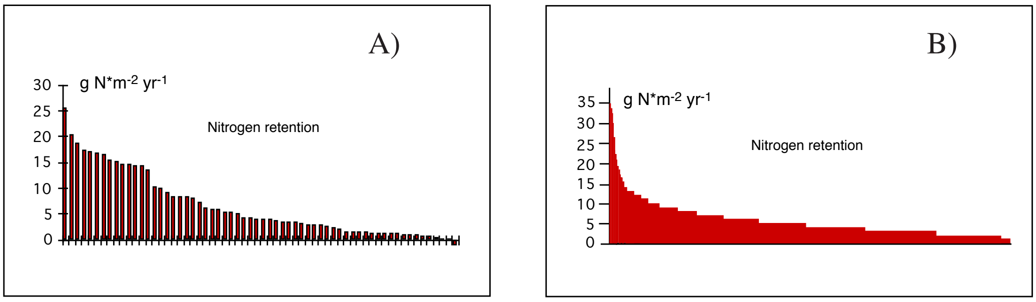


Figure 12. Comparison of retention data (g Nm^{-2}*y^{-1}) from the present input-output measurements (A) and the data derived from large scale dynamic modelling (B) of Arheimer & Brandt (1998). Set (A) consists of 62 cases and set (B) of 1637 modeled lake retentions (redrawn frtom Arheimer & Brandt 1989).Note the different scales.*

Table 5. Comparision of retention data from the compiled input-output measurements (1), the data derived from large scale dynamic modeling (2) of Arheimer & Brandt (1998) and (3) Sonesten et al. (2003). Nitrogen retention/input (%) is given as indented text (lacking from Arheimer et al.)

	Mean	25% percentile	50% percentile	75% percentile	N
1) Ret. (g N/m2,y)	6,6	1,5	4,1	10,0	62
2) Ret. (g N/m2,y)	10,6	1,9	3,4	6,1	1637
3) Ret. (g N/m2,y)	2,43	0,95	1,34	1,82	40
1) Ret./Input (%)	*37,6*	*20,9*	*40*	*49,2*	*62*
3) Ret./Input (%)	*15,1*	*4,2*	*11,6*	*21*	*40*

than for the modelled. Consequently, statistics for these distributions (table 5) show small differences up to the median value (modelled retention 83% of the compiled measurements), higher differences thereafter. Since retention as a percentage of input was not given by Arheimer & Brandt (1998) further comparisons of retention efficiency could not be made.

Data is also available for 40 named lakes in the south-western part of Sweden (Sonesten et al. 2003). Arheimer & Brandt (1998) did not treat the majority of these lakes (west of Lake Vänern). Both absolute and relative retention are now given (table 5). This set also gives lower retention than the compiled set of yearly balances (modelled median retention 39% of the compiled measurements). The difference is larger at the high ends of the data-sets and the 75% percentile of the modelled set is only 20% of the compiled balances.

Nitrogen retention relative to input is also higher according to the present compilation as compared to the modelled data given by Sonesten et al. (2003). The median of their data is ca. 1/3 of the median of the present compilation.

Data on measured nitrogen retention from eight lakes in southern Norway may also be complementary to these data (Berge et al. 1997). In the Norwegian lakes retention efficiency spans 0-28% (one basin with negative retention excluded). These lakes had a wide range of different characteristics but no factors of major influence could be pointed out in regression analysis. Low or zero retention was seen in the most oligotrophic lakes. In these lakes high concentrations of nitrate, due to airborne pollution, and low production (low phosphorus level) as well as low denitrification, were typical. Many of the 40 lakes in south-western Sweden with modelled low retention may also fulfil these requirements; they are mostly low-productive, they are large with supposedly long turnover times, TOC-poor water, and they have excess nitrate. These characteristics would offer an explanation to the low nitrogen retention in these lakes. Since these data are derived from modelling there is also the possibility that model input data are not representative of the true conditions.

It has not been possible to include also oxygen concentration or redox potential in the regressions but direct denitrification assessments (p. 9) also points to the possibility that denitrification is reduced with increasing productivity due to lowered redox-potential and reduced nitrification (Ahlgren et al. 1994). The review of direct assessments also indicates that the processes occur at many sites and at different rates which makes the regulation of denitrification not easily comprehended.

It must also be stressed that in the type of regression analysis presented above total nitrogen retention is used as a surrogate for denitrification. This would be a problem if the balance between denitrification and sedimentation is less constant than previously assumed. The regulation of net sedimentation may well be governed by other factors than denitrification and thus interfere with the denitrification analysis to an unknown extent.

4. Representativity and future development

The question of representativity is a matter of serious concern in the present compilation. Many of the reported yearly retentions are derived from "problematic" lakes, mostly meaning that they suffer from eutrophication, which is evident from table 1. As far as known, less than 800 or 0,8% of the Swedish lakes have phosphorus concentrations above 25 $\mu g\ P*l^{-1}$ (Johansson & Persson 2001). In the present material 40 lakes have phosphorus concentrations higher than 25 $\mu g.P*l^{-1}$. They are thus over-represented in this survey (figure 13). This is important only if the phosphorus level influences nitrogen retention, however which cannot be clearly proven at the present state of knowledge. The equations built on nitrogen input and phosphorus factors do not incorporate flushing factors and give no conclusive evidence for a tentative retention optimum. Whereas flushing factors behave logically for most subsets of data, the phosphorus factors do not. Therefore equations containing nitrogen loading and hydraulic loading must be given priority compared to equations containing only nitrogen loading and a phosphorus factor.

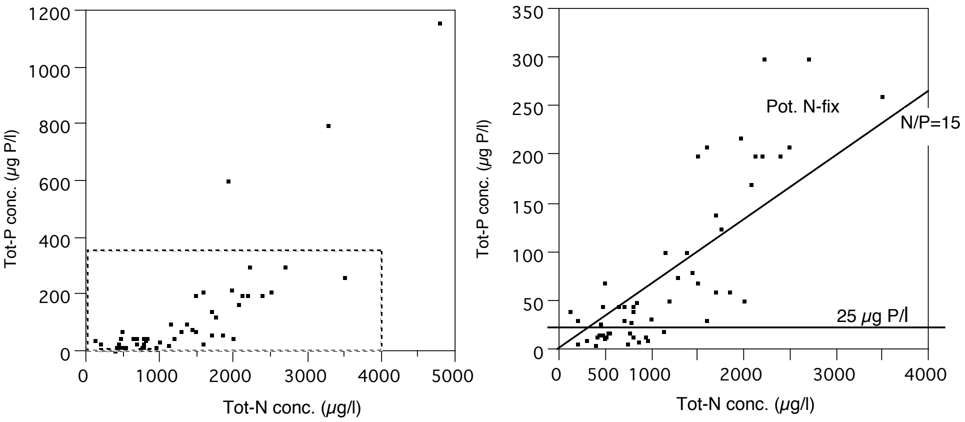


Figure 13. Concentrations of total phosphorus and total nitrogen in the complete set of lakes (A) and in a subset (B) with concentration restrictions according to (A). Limits for eutrophic conditions (25 $\mu gP*l^{-1}$) and nitrogen fixation (tot-N/tot P<15) marked.

The most important driving variable, nitrogen loading, is also higher than average in the eutrophicated lakes in the examined set of data, whereas the hydraulic load may be fairly representative as judged from the lakes´ location (figure 3). Corrections to lower nitrogen loadings typical for most Swedish lakes would reduce retention in absolute terms, but as far as can be judged from figure 6, relative retention stays over 20% until absolute retention passes below 5-10 g $N*m^2*y^{-1}$. This is a rough estimate, though.

For calculation of nitrogen retention in a "normal" lake, input data for nitrogen and water are needed. Coefficients for area-specific nitrogen loss from soil, for atmospheric nitrogen deposition and for sewage nitrogen may be used. Drainage area size, lake surface area and area-specific runoff is also needed. For calculations different equations containing the flushing term may be used, the most general being:

$log(N_{ret}) = -0.01 + 1.04*log(N_{load}) - 0,48*log(Hy_{load}), r=0.88, p=0,0001, n=60$

It is based on all data, but if water turnover time is higher than 0.25 years an equation with slightly better fit could be used:

$log(Nret) = 0,21 + 1.17*log(Nload) - 0,52*log(Hyload), r=0.90, p=0,0001, n=42$

Using these equations as predictive models it is possible to apply them to a representative random sample of Swedish lakes for which adequate data may be gathered. In that way representative data on nitrogen retention in Swedish lakes would be obtained. The data needed could be found in "The national inventory of lakes" with complementary environmental statistics information.

A comparison of the outcome of dynamic modelling and the present approach is also high on the agenda. Input data for the 62 cases treated here is available for dynamic modelling (Appendix 1) but need additional temperature and runoff or precipitation data with high frequency.

The input data for these predictive functions are very similar to input data needed to run dynamic retention models. This also means that a comparison between the two approaches in previously dynamically modelled lakes could be made based on already gathered data for dynamic models but with a time step of one year.

Conclusions

According to the EU-directive on treatment of urban sewage water (Directive 91/271/EEG) a major part of the Swedish coast has been declared "sensitive" for nutrient pollution, and demands on nitrogen reduction in sewage works have been raised. However, for nitrogen there is great natural losses when water passes through the landscape to the sea. In this respect lakes and watercourses protect the Swedish coastal zone from inland nitrogen loading, including sewage nitrogen. This may be an argument for differential nitrogen reduction among inland sewage works, but the magnitude of natural nitrogen retention has been poorly known until the last few years.

In the present review, recent work with retention modelling on a landscape scale is briefly summarised with special attention to nitrogen retention in lakes (Arheimer & Brandt 1998, 1999, Anon. 1997, Brandt & Ejhed 2003. Johansson & Kvarnäs 1998, Sonesten et al. 2003). However, the main issue has been to compile input-output nitrogen mass balances for Swedish lakes. This is considered a secure estimate of nitrogen retention. A total of 62 such cases are discussed. Since retention generally is dominated by denitrification (73% of the total retention in 9 cases here, 77% in 58 Danish lakes) total retention also mirrors denitrification.

Nitrogen retention spanned from zero to c. 25 g $N*m^{-2}*y^{-1}$ with a median of 4.0 g $N*m^{2}*y^{-1}$ in lakes studied with mass balances (figure 10). Retention efficiency spanned from zero to c.80% of input nitrogen with a median of 40%. Low retention efficiency appeared primarily in lakes with low absolute retention but there was no correlation between absolute and relative retention (figure 6).

The range of retention in the present compilation compares favourably with modelled retentions in c. 1640 lakes (figure 10) but there are more high-retention lakes in the present study. Modelled retentions in another study of 40 lakes coincided with the lower quartile of that recorded in the present study.

It has not been possible to compare assessments made by the different methods on individual objects. Since there is a proportionality between nitrogen loading and nitrogen retention it is assumed that the compilation data generally show high retentions because high-loading lakes are over-represented. At present this cannot be checked since data on nitrogen loading for individual Swedish lakes is not available. It can be stated, however, that lakes with high phosphorus concentrations are over-represented which supports the idea of high nitrogen loading.

The compiled data contain a wide range of potentially governing factors in addition to a wide retention range (Appendix 1). A multiple regression analysis was therefore performed to analyse and possibly predict nitrogen retention. Both nitrogen loading and hydraulic loading were instrumental in regulating nitrogen retention but no clear-cut dependence on phosphorus factors could be found. The dependence on nitrogen loading compared favourable with earlier published functions (Fleischer & Stibe 1991, 1994) whereas phosphorus factors, found to be instrumental by Hellström (1996) did not behave logically in the present material and were omitted in predictive functions.

It may be concluded that during recent years the knowledge of nitrogen retention has greatly improved and that on a landscape scale dynamic models with reasonably high resolution may now describe losses of wastewater nitrogen. For individual lakes or lake chains the nitrogen retention may be predicted based on functions derived from the retention data compiled in this paper and with the precision given for these estimates (p. 34).

On a scale comprising all Swedish bodies of standing water (c. 60,000 lakes > 0.04 km^2) data cannot yet be presented. However, different samples within this lake-population have given the magnitude of retention variation, the span of retention efficiency and functions to predict nitrogen retention once area-specific water and nitrogen input for individual lakes are given.

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Appendix 1.

1. The data-base

Lake	X-coord	Y-coord	Area (km^2)	Volume (10^6_m^3)	Mean depth(m)	Max depth(m)	Watershed area(km^2)	Year
Norrviken 1	659728	161988	2.70	14.30	5.4	12.5	99.50	1961/62
Norrviken 2	659728	161988	2.70	14.30	5.4	12.5	99.50	1972
Norrviken 3	659728	161988	2.70	14.30	5.4	12.5	99.50	1992-93
L. Vallentuna	659771	162546	6.10	15.40	2.5	4.5	44.20	1992-93
Edssjön	660010	161773	1.02	3.00	3.0	5.4	127.00	1972
Oxundasjön	660637	161566	1.60	5.30	3.3	6.0	264.00	1972
Björkarn	662689	165052	0.37	0.29	0.8		155.00	1987-90+1992
Limmaren	662767	166446	5.50	28.00	4.6	7.8	17.00	1991-92
Mälaren A	659180	152170	61.00	210.00	3.4	19.0	8642.00	1981-85
Mälaren D	662709	160136	94.10	1080.00	11.5	50.0	3979.00	1981-85
Mälaren C	659072	159203	500.00	8450.00	16.9	60.0	11848.00	1981-85
Hjälmaren	657240	152792	477.80	2893.00	6.1	22.0	3340.20	1981-85
Vänern	651574	132967	5650.00	153000.00	27.0	106.0	46800.00	1982-92
Vättern	648694	143413	1856.00	74000.00	40.0	128.0	4503.00	1975-85
Glaningen	666693	145838	0.75	1.13	1.5	3.0	34.70	1974
Ramsjön	664569	156575	0.39	0.70	1.8	3.5	12.30	1974
Ryssbysjön	630069	140009	2.73	4.91	1.8	3.1	99.10	1974
Gåran	668629	150257	0.15	0.23	1.5	3.1	26.00	1988
S. Bergundasjön 1	630406	143665	4.30	10.20	2.4	5.4	45.10	1973
S. Bergundasjön 2	630406	143665	4.30	10.20	2.4	5.4	45.10	1975-76
Fegen	635040	133900	23.60	181.70	7.5	36.0	165.30	1974
Kalvsjön	634991	133494	7.20	44.90	6.2	21.0	466.70	1974
Mjörn	642138	130063	55.50	850.00	15.0	48.0	1053.50	1990
Boren 1	649283	146898	28.00	170.00	6.0	12.0	6437.00	1973
Boren 2	649283	146898	28.00	170.00	6.0	12.0	6437.00	1976
Roxen	648779	150974	97.00	748.00	5.0	8.0	13233.00	1985-89
Glan	649686	151617	75.60	470.00	9.9	22.5	14879.00	1985-89
Bolmen	629511	136866	184.00	1070.00	5.0	36.0	1640.00	1980-84
Ringsjön	620062	135224	39.57	184.20	4.7	17.5	347.00	1991
Ö. Ringsjön+Sät 2	619626	135565	24.73	137.60	5.6	17.5	98.50	1990
V. Ringsjön2	620062	135224	14.80	46.60	3.1	5.5	122.30	1990
Ringsjön	620062	135224	39.57	184.20	4.7	17.5	347.00	1980
Ö. Ringsjön+Sät 1	619626	135565	24.73	137.60	5.6	17.5	98.50	1980
V. Ringsjön 1	620062	135224	14.80	46.60	3.1	5.5	122.30	1980
Finjasjön	622731	136920	11.10	42.80	3.8	12.2	249.50	1976-78
Finjasjön 1	622731	136920	11.10	42.80	3.8	12.2	249.50	1976-77
Finjasjön 2	622731	136920	11.10	42.80	3.8	12.2	249.50	1978
Finjasjön 3	622731	136920	11.10	42.80	3.8	12.2	249.50	1988-90
Ivösjön	621669	141629	51.95	558.29	10.7	48.0	995.90	1990-93
Oppmannasjön	621816	140914	12.73	49.65	3.9	12.5	91.00	1989/90
Stugsjön	759760	160858	0.02	0.02	1.2	4.5	0.11	1971-75
Hymenjaure+P	759728	160850	0.02	0.03	1.3	4.5	0.21	1974
Magnusjaure+N	759637	160677	0.01	0.03	2.2	5.5	0.12	1974
Hymenjaure+N+P	759728	160850	0.02	0.03	1.3	4.5	0.21	1975
Gunillajaure+N+P	759660	160818	0.02	0.14	6.0	13.7	0.20	1979
Gårdsjön	644432	127626	0.31	1.50	4.9	18.5	2.11	1979-81
Örträsket	717326	165656	7.30	160.00	23.0	64.0	2174.00	91/92
Erken	664060	165948	23.70	214.00	9.0	21.5	117.00	1984
V. Storsjön	671610	154970	39.20	181.50	4.5	15.0	1156.70	1991-96
Ö. Storsjön	671880	155660	31.50	89.35	31.5	15.0	2137.20	1991-96
Botjärn	664244	146726	0.10	0.32	3.3	14.4	2.94	1972-73
Rusken	634172	141113	35.81	136.53	3.5	16.4	843.10	1985-89
Vidöstern	631841	138929	43.98	211.23	4.4	35.0	1277.20	1990-98
Ralången	642136	144141	5.30	13.41	2.4	5.0	595.00	1989-90
Brunnsjön	668374	150912	1.48	3.46	2.3	3.6	57.87	1989/90-93/94
Tåkern	647411	144338	45.00	35.00	0.8	2.0	344.00	1983-96
Väsman	667085	146552	38.60	410.00	10.6	53.0	1149.00	1978-80
Södra Barken	665545	149734	11.00	63.00	5.7	24.0	2191.00	1978-80
Åsunden	644635	149350	19.50	293.00	15.0		559.50	1989-91
Åsunden+Yttre Ås.	644635	149350	30.00	358.00	11.9		646.00	1989-91
Aspen	640873	128461	4.80	79.60	16.6	31.5	1392.00	1981-90
Hornborgasjön	646918	136672	33.00	25.00	0.8	1.6	585.00	1984-96

Duration(y)	Residence time (y)	TP-input (g/m^2.y)	TP-retention(g/m2.y)	TN-input (g/m^2.y)	TN-output (g/m^2.y)	TN-retention(g/m^2.y)	Δ_storage (g/m^2.y)	Eq.retention(g/m^2.y)	Eq. retention(%)
2	0.62	4.03	49.00	66.50	50.00	16.50		16.50	24.81
1	0.90	0.45	-125.00	6.62	9.22	-2.60	-3.99	1.39	21.00
2	0.94	0.32	-32.90	8.15	6.58	1.57		1.57	19.50
2	1.99	0.09	6.65	2.62	1.45	1.17		1.17	44.86
1	0.12	2.84	-10.00	29.90	31.17	-1.27	-5.00	3.73	12.47
1	0.11	2.56	14.60	41.50	33.88	7.62	-2.30	9.92	23.90
5	0.01	5.03	3.50	144.20	142.42	1.79	0.00	3.95	2.25
2	3.00	0.06	-24.00	1.96	1.54	0.92		0.92	46.94
5	0.10	2.53	-1.60	59.13	57.00	3.01		3.01	5.09
5	1.20	1.17	34.80	26.56	18.25	8.33		8.33	31.36
5	1.80	0.58	44.20	13.81	8.07	5.95		5.95	43.08
5	3.40	0.14	55.00	5.56	1.30	4.26		4.26	76.62
10	9.30			4.18	2.79	1.39	0.13	1.26	30.24
10	60.00	0.04	90.00	1.72	0.52	1.20	0.40	0.80	46.74
1	0.08	6.36	-35.00	39.90	31.30	8.60	-7.00	15.60	39.10
1	0.20	1.42	2.00	27.20	11.90	15.30	-1.90	17.20	63.24
1	0.13	0.49	-40.00	43.00	24.20	18.80	-6.70	25.50	59.30
1	0.02	1.70	-26.00	67.00	59.00	8.00		8.00	11.94
1	0.58	8.80	52.00	26.85	19.38	7.47	1.98	5.49	20.45
2	1.10	0.32	-493.00	10.90	7.41	5.74	2.27	3.48	31.90
1	1.80	0.10	60.00	2.66	1.24	1.42	0.25	1.17	44.10
1	0.25	0.56	38.00	16.00	14.18	1.82	0.67	1.15	7.19
1	1.40	0.54	76.00	15.38	9.57	5.81		5.81	37.79
1	0.20	1.80	67.00	23.30	14.80	8.50		8.50	36.48
1	0.40	0.40	50.00	17.60	7.20	10.40		10.40	59.09
5	0.16	0.77	-27.00	33.00	27.70	5.30		5.30	16.06
5	0.25	1.79	20.60	46.90	41.93	4.97		4.97	10.60
5	1.00	0.06	60.00	23.91	9.56	14.35		14.35	60.02
1	1.11	0.22	20.00	20.20	5.07	15.13	0.75	14.38	71.19
1	0.80	0.23	-31.79	23.74	9.10	14.64		14.64	61.67
1	0.30	0.33	19.75	20.74	11.42	9.32		9.32	44.95
1	1.11	0.84	50.40	24.96	8.06	16.90		16.90	67.70
1	0.80	1.13	30.00	33.30	18.52	14.78		14.78	44.38
1	0.30	2.48	64.00	41.90	21.50	20.40		20.40	48.69
3	0.42	1.43	30.00	32.80	17.62	15.18		15.18	46.28
2	0.42	1.71	41.00	30.60	17.15	13.45		13.45	43.95
1	0.42	0.89	-10.00	37.40	18.70	18.70		18.70	50.00
3	0.42	0.46	-8.60	35.25	17.99	17.26		17.26	48.96
4	1.90	0.13	39.00	7.30	3.17	4.13		4.13	56.58
1	2.16	0.09	78.00	8.08	0.82	7.26		7.26	89.85
5	0.25	0.03	23.00	0.64	0.48	0.19		0.19	29.61
1	0.20	0.31	89.00	0.24	0.17	0.21		0.21	87.08
1	0.20	0.01	57.00	4.08	0.53	3.55	2.13	1.42	34.80
1	0.20	0.31	73.00	3.88	0.92	2.96	1.31	1.65	42.53
1	1.50	0.51	92.00	6.53	0.65	5.88	3.52	2.36	36.09
3	1.00	0.02	22.00	2.36	1.45	0.91	-0.15	1.07	45.17
1	0.28	0.96	14.00	27.39	26.30	1.09		1.09	3.98
1	6.30	0.06	29.00	1.99	1.12	0.87		0.87	43.67
6	0.65	0.23	-0.16	5.21	2.38	2.83		2.83	54.32
6	0.18	0.56	-0.11	14.43	11.06	3.37		3.37	23.35
2	0.29	0.11	2.00	3.62	1.53	2.09		2.09	57.73
5	0.54			10.60	6.26	4.34		4.34	40.97
9	0.52	0.31	31.80	12.23	9.52	2.71		2.71	22.16
2	0.11	0.69	-41.00	32.00	28.60	3.40		3.40	10.63
5	2.40	0.92	-15.00	23.70	16.82	6.88		6.88	29.03
14	0.67	0.16	70.00	4.60	1.64	2.96		2.96	64.25
3	1.15	0.20	33.00	4.78	4.23	0.55		0.55	11.51
3	0.10	1.09	12.00	30.24	28.57	1.67		1.67	5.52
3	1.20	0.27	7.00	17.94	14.61	3.33		3.33	18.56
3	1.20	0.20	22.00	13.55	9.56	3.99		3.99	29.46
10	0.12	2.15	16.60	127.70	129.00	-1.30		-1.30	-1.02
13	0.15	0.23	37.00	19.53	11.06	8.47		8.47	43.37

Lake	N-fixation) (g/m^2.y)	Denitrific. (g/m^2.y)	TN_conc. (μgN*l^-1)	Min_DIN_ conc.(μgN*l^-1)	Mean_DIN conc.(μgN*l^-1)	Max_DIN conc.(μgN*l^-1)	TP_conc. (μgP*l^-1)	Ref. (nr)
Norrviken 1		24.07	3500	300	1500	3500	260	1
Norrviken 2			2500	10	1000	1750	210	2
Norrviken 3	0.00	0.92	1150	20	427	950	100	3
L. Vallentuna	0.00	0.64	1383	13	167	533	100	4
Edssjön			2700	20	250	2000	300	5
Oxundasjön			1700	20	400	1600	140	6
Björkarn	0.00		1700					7
Limmaren	0.50		1450	10	450		80	8
Mälaren A	0.88		600	10	280	750	48	9
Mälaren D	0.02		820	2200	1200	1000	60	10
Mälaren C	0.21		400	250	350	550	29	11
Hjälmaren			798	20	150	450	46	12
Vänern			870	550	555	575	9	13
Vättern			740	370	390	420	7	14
Glaningen			1930	250	600	1200	600	15
Ramsjön			2220	100	700	2000	300	16
Ryssbysjön			1500	5	400	1200	200	17
Gåran			810		380		40	18
S. Bergundasjön 1			4800	210	1330	3200	1160	19
S. Bergundasjön 2	2.25		3300	20	1200	3000	800	20
Fegen			420		92		14	21
Kalvsjön			500		145		12	22
Mjörn			950	580	650	740	14	23
Boren 1			450	49	149	320	26	24
Boren 2			440	16	145	275	15	25
Roxen	0.00		700				30	26
Glan	0.00		650	0	300	800	45	27
Bolmen			522				14	28
Ringsjön			1300				75	29
Ö. Ringsjön+Sät 2			1700	0	800	1600	60	30
V. Ringsjön 2			1500	0	250	700	70	31
Ringsjön			2400	0	150	800	200	32
Ö. Ringsjön+Sät 1			2200	0	200	800	200	33
V. Ringsjön 1			2500	10	50	800	210	34
Finjasjön			2080	160	1060	2250	170	35
Finjasjön 1			2130	110	1090	2400	200	36
Finjasjön 2			1970	270	770	1780	219	37
Finjasjön 3			1600		300		210	38
Ivösjön			800	110	260	370	14	39
Oppmannasjön			1000	5	120	340	32	40
Stugsjön	0.03		197		12		7	41
Hymenjaure+P	0.14		200		10		30	42
Magnusjaure+N			400		260		4	43
Hymenjaure+N+P		0.70	500				70	44
Gunillajaure+N+P	0.00	0.65	130		700		40	45
Gårdsjön	0.00	1.25	400	80	150	210	5	46
Örträsket		0.62	300	5	40	80	11	47
Erken			770	5	150	350	18	48
V. Storsjön			470	8	100	275	45	49
Ö. Storsjön			700	3	100	180	45	50
Botjärn			420	40	90	170	14	51
Rusken								52
Vidöstern			553	28			17	53
Ralången			1200	10	300	700	50	54
Brunnsjön			1617		462		124	55
Tåkern			2000	1500	1500	3100	50	56
Väsman			480	80	180	200	15	57
Södra Barken			529	30	180	320	18	58
Åsunden			1130				20	59
Åsunden+Yttre Ås.								60
Aspen			956				10	61
Hornborgasjön			1600				30	62

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