This document is the accepted manuscript version of the following article:
Müller, B., Meyer, J. S., & Gächter, R. (2022). Denitrification and nitrogen burial in Swiss Lakes. Environmental Science and Technology.
https://doi.org/10.1021/acs.est.1c07602

Denitrification and nitrogen burial in Swiss lakes

Beat Müller[1*], Joseph S. Meyer[2,3], René Gächter[4]

[1]Eawag, Swiss Federal Institute of Aquatic Science and Technology, CH-6047 Kastanienbaum, Switzerland

[2]Department of Chemistry, Colorado School of Mines, Golden, Colorado 80401 USA

[3]Applied Limnology Professionals LLC, Golden, Colorado 80401 USA

[4]Guest at Eawag

* Corresponding author

Correspondence to: Beat Müller (beat.mueller@eawag.ch) *Phone:* +41 58 765 2149

ORCID: Beat Müller: 0000-0003-3696-9035

Joseph S. Meyer 0000-0001-8097-4187

René Gächter: 0000-0002-7186-8574

Accepted Version of manuscript doi.org/10.1021/acs.est.1c07602

Environ. Sci Technol., Publication date: January 28, 2022

Abstract

Earths' nitrogen (N) cycle is imbalanced due to excessive anthropogenic inputs. Freshwater lakes efficiently remove N from surface waters by transformation of NO_3^- to atmospheric N_2 and/or N_2O (denitrification; DN) and by burial of organic N in sediments (net sedimentation; NS). However, relatively little is known about the controlling environmental conditions, and few long-term measurements on individual lakes are available to quantify conversion rates. We report N-elimination rates in 21 Swiss lakes estimated from whole-lake N budgets covering up to ~20 yr of monitoring. The NO_3^- concentration in the bottom water was the main predictor of DN. Additionally, DN rates were positively correlated with external N load and the area-specific hydraulic loading rate (mean depth/water residence time; Q_s). Net sedimentation of N was strongly related to total phosphorus (P) concentration. Nitrogen removal efficiency (NRE), the fraction of the load of dissolved N to a lake removed by DN and NS, was strongly negatively related to Q_s. This previously unconsidered variable improves predictability of NRE and does not require knowledge of N and P loading rates or concentrations. We conclude that P management alone intended to oligotrophy lakes only slightly increases N export unless it is accompanied by N management.

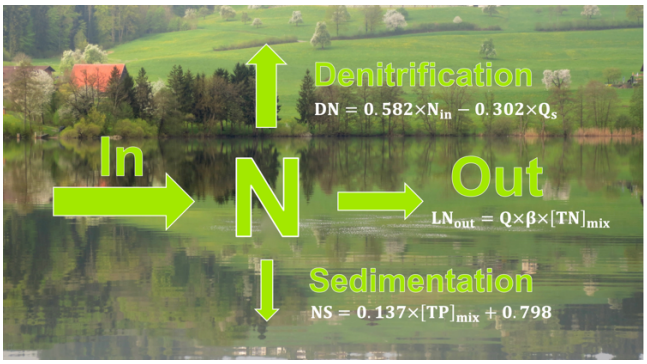
Keywords:

Nitrogen removal efficiency, net sedimentation, nitrogen load, phosphorus management, oligotrophication

Synopsis-statement:

A new quantitative relationship can be used to predict nitrogen removal efficiency from lakes and thereby predict potential eutrophication of nitrogen-limited downstream ecosystems.

TOC-Art:



Photograph by BM

54 # 1. Introduction

55 Freshwater lakes are efficient sinks for reactive nitrogen (N) that is excessively applied as an
56 agricultural fertilizer and adversely affects N-limited downstream costal environments.[1-3] Removal
57 from surface waters is mainly due to (1) deposition and burial of particulate N in lake sediments [i.e.,
58 net sedimentation (NS)], and (2) denitrification [DN; microbial transformation of NO_3^- to N_2 and/or N_2O,
59 among several other lower-magnitude, alternative pathways such as dissimilatory nitrate reduction to
60 ammonium, denitrification via sulfur or iron oxidation, and anaerobic ammonium oxidation
61 (anammox)].[4,5] However, efforts to reverse past eutrophication of lakes (referred to as
62 oligotrophication) that make anaerobic N-removal processes less favorable have led to speculation
63 that "cleaner lakes are dirtier lakes".[6]

64 Increasing concern about the anthropogenically-enhanced natural N cycle has resulted in many
65 studies of N transformation in past decades. However, data are amazingly scarce in view of the
66 importance of the subject, and relatively few studies of N-elimination processes in lakes are
67 available.[7,8] The broad variety of aquatic systems, the variability of processes in time and space, and
68 the limited number of measurements hinder development of a clear picture of the controlling variables
69 on a representative scale. Two compilations of DN rates measured in individual lakes cover a wide
70 range of 5.2 to 21 g N m^{-2} yr^{-1} [(9)] and 0.2 to 38 g N m^{-2} yr^{-1}.[10] In a review of a selected dataset, the
71 latter authors concluded that seasonality and the concentrations of NO_3^- and O_2 explained the
72 majority of variations in DN rates of lakes. Global average DN rates estimated by Harrison et al.[11] are
73 3.6 g N m^{-2} yr^{-1} for small lakes (<50 km^2) and 3.1 g N m^{-2} yr^{-1} for larger lakes, emphasizing the
74 importance of small lakes for the removal of N because the combined area of small lakes is more than
75 twice that of large lakes.[11]

76 More-recent studies supported and extended these quantitative findings. However, most studies that
77 have used the isotope-pairing technique or the acetylene-inhibition method are site-specific and
78 document DN rates determined from isolated sediment cores.[12-15] Àlvarez-Cobelas et al.[8] reported a
79 range of denitrification rates of 3.5 to19 g N m^{-2} yr^{-1} in a chain of flow-through lakes, and Bruesewitz
80 et al.[16] reported a similar range (4.4-15 g N m^{-2} yr^{-1}) at 40 sites in the shallow Gull Lake, Michigan
81 (USA). Müller et al.,[17] investigating benthic nitrate fluxes in an oligotrophic lake and a eutrophic lake,
82 reported a weak seasonality with ~2-fold higher DN rates in spring than in fall, but no significant

differences at various locations within the two lakes. They observed distinctly different average DN rates of 20 ± 6.6 g N m^{-2} yr^{-1} and 3.2 ± 4.2 g N m^{-2} yr^{-1} in the eutrophic and oligotrophic lake, respectively, supporting earlier results (e.g.,[18]) that indicated DN rates were higher in eutrophic than in oligotrophic lakes.

Li and Katsev[19] and Small et al.[20] concluded that in oligotrophic lakes, DN is negligible due to a thick oxygenated sediment layer, in which nitrification exceeds DN and thus the sediment may become a net source instead of a sink for NO_3^-, resulting in a hypolimnetic NO_3^- accumulation above the sediment during lake stratification. On the other hand, meromixis quenches DN in the anoxic part of the water column due to a lack of nitrification.[21] Meromixis is the lack of annual, complete mixing in a lake, an increasingly frequent phenomenon as climate warming increases lake temperatures.[22]

Two empirical relationships for the removal of bioavailable N in freshwater lakes, rivers, wetlands, estuaries, and coastal oceans have been proposed in an overarching attempt to generalize individual observations. First, DN rate increases as areal N load increases[9,23-26] and as in-lake NO_3^- concentration increases.[14,18] Second, the percentage of N removed by a lake increases with increasing water residence time.[23,25,27,28]

Herein, based on whole-lake N budgets extracted from up to ~20 yr of monitoring data collected from 21 Swiss lakes, we estimate annual average DN and NS rates and discuss the controlling factors such as (1) bottom-water NO_3^- concentration above the sediments ($[NO_3^-]_{bottom}$), (2) areal N load (N_{in}), (3) water residence time (τ), (4) mean depth (z_{mean}), and (5) trophic state. Finally, we discuss the consequences of lake oligotrophication on the removal of N. These results provide insight into previously unreported relationships that can be used to predict N removal from seasonally-stratified freshwater lakes.

2. Materials and Methods

2.1. Whole-lake N budgets

We analyzed long-term monitoring data from 21 large Swiss lakes (Figure 1 and Table 1) collected during the past ~20 yr [made available by the Swiss Federal Office for the Environment (FOEN) or the responsible cantonal offices] and estimated annual N-removal rates from whole-lake budgets. We

define DN as including all processes in a lake that convert dissolved N into N_2 and N_2O gases and thus remove N from the lake to the atmosphere.[18] The equation for the whole-lake N budget is:

$$LN_{in} = LN_{out} + LN_{NS} + LN_{DN} \quad (1)$$

where LN_{in} is the annual dissolved N load in the inflowing waters and atmospheric deposition, LN_{out} is the mass of N annually exported from the lake, LN_{NS} is the net mass of N annually deposited in the sediment, and LN_{DN} is the mass of N annually removed by denitrification. LN_{DN} was estimated as the difference between LN_{in} and (LN_{out} + LN_{NS}) in the mass balance equation 1. Because LN_{in} and LN_{NS} were only available for an integrated time range and not annually, we averaged the annual values of LN_{out} and LN_{DN} over the entire time range of ~20 yr for compatibility. The mass of dissolved N in each lake was assumed to be at steady state (i.e., V·dN/dt =0). This assumption is justified because the mass of dissolved N changed little in these lakes (see values of V·dN/dt in Table S-1), and those changes were negligible compared to LN_{in}.

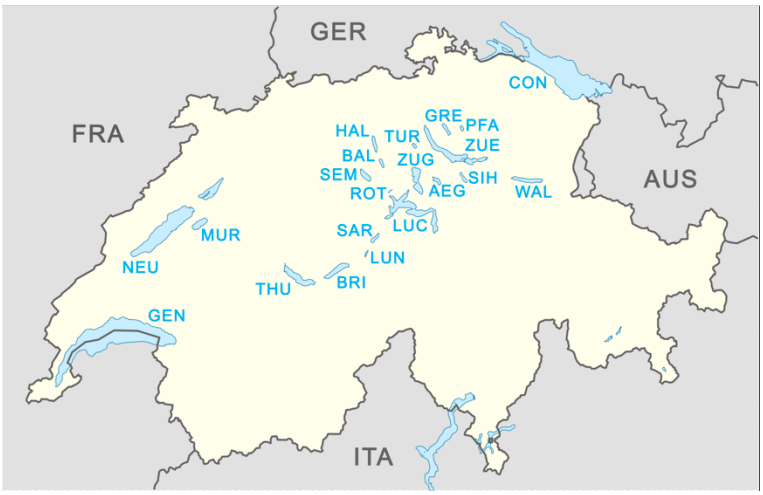


Figure 1: *The 21 Swiss lakes included in the study, with monitoring data available for the past ~20 yr.*

Total N (TN) concentrations for lakes WAL, AEG, NEU, SEM, BAL, ROT and ZUG (abbreviations identified in Table 1) were not available for the entire time span considered for the whole-lake budgets. We estimated TN by multiplying the NO_3^- concentration measured in the water column after winter overturn and before summer stratification ($[NO_3^-]_{mix}$) by a constant factor (1.23) derived from

132 regression of the TN concentration measured in the water column after winter overturn and before
133 summer stratification ($[TN]_{mix}$) vs. $[NO_3^-]_{mix}$ in datasets from Swiss lakes in which both concentrations
134 were measured (Note S-1, Figure S-1). The volume-weighted concentrations of $[NO_3^-]_{mix}$, $[TN]_{mix}$, and
135 the total phosphorus concentration measured in the water column after winter overturn and before
136 summer stratification ($[TP]_{mix}$) were averaged from February to April using monthly monitoring data
137 and bathymetric information (Table 1; see Table S-1 for a characterization of the lakes).

138 We estimated LN_{in} using the model MODIFFUS of Hürdler et al.[29] (Note S-2) that has been calibrated
139 and verified in Switzerland, incorporating land use, agricultural practices, soil type, ground slopes, and
140 precipitation. The model only estimates dissolved N inputs from diffuse sources; inputs of particulate
141 N (PN) are not considered. Thus, LN_{in} underestimates the true N loads because even though the
142 major fraction of PN inputs settles within the delta regions of tributaries, a relatively small fraction may
143 become bioavailable and subsequently may participate in the in-lake N cycling. This may cause some
144 underestimation of the export terms of Equation 1. As most PN does not reach the lakes' profundal
145 zone, its deposition rate in the spatially constrained delta region cannot be determined accurately.
146 Moreover, the temporal distribution of PN may be extremely variable depending on floodwaters and
147 catchment quality and thus difficult to quantify. In conclusion, we think that the use of dissolved N
148 loads considers the participation of N in the lakes biogeochemical cycle more appropriately than if the
149 PN fraction was included. Furthermore, the MODIFFUS model estimates atmospheric N deposition
150 but not N inputs to surface waters from sewage treatment plant discharges. We obtained data on total
151 N export from 854 sewage treatment plants in Switzerland, including estimates of storm-water
152 overflows from FOEN, and allocated the discharges from 652 to their respective lake catchments.
153 Atmospheric N deposition directly to the lake surface was added separately using data available from
154 FOEN (https://map.geo.admin.ch).

155 Only a fraction of the catchment areas of Lakes Geneva (91.3%), and Constance (51.2%) are located
156 in Switzerland. The areal loads known from Switzerland (based on MODIFFUS output) for Lake
157 Geneva were scaled-up for the whole catchment. For Lake Constance, we used a reported
158 estimate.[30]

159 Because N-cycling processes in rivers and streams are very complex, there are many uncertainties
160 when using models such as MODIFFUS to simulate N inputs to lakes. However, we were able to test
161 the accuracy of MODIFFUS for the purposes of the current study, because two of the 21 lakes in our

dataset (Baldegg and Sempach) have systematic long-term monitoring data from tributaries. The close agreement of dissolved N loads estimated by the MODIFFUS model and calculated from the past ~20 yr of chemical monitoring for the two lakes supports the reliability of the modeled N loads (Note S-3).

Nitrogen export data from the lakes by surface outflow includes dissolved NO_3^- and PN bound in algal biomass. We calculated annual values of LN_{out} as:

$$\mathrm{LN_{out}} = \mathrm{Q} \times \beta \times [\mathrm{TN}]_{\mathrm{mix}} \quad (2)$$

and then averaged across all years. In Equation 2, $[TN]_{mix}$ is expressed as g N m^{-3}, Q is the annual water discharge [m^3 yr^{-1}], and β is a unitless stratification coefficient calculated as:

$$\beta = \frac{\overline{[\mathrm{TN}]}_{\mathrm{epi}}}{[\mathrm{TN}]_{\mathrm{mix}}} \quad (3)$$

where $\overline{[TN]}_{epi}$ is the mean annual concentration of TN in the epilimnion.

We calculated τ as the lake volume divided by Q. Outflows of these lakes are measured with automated gauge stations at 10-min intervals and published as daily, monthly, and annual averages by FOEN (available at: https://www.hydrodaten.admin.ch/en/) or the responsible cantonal agencies.

We determined NS (g N m^{-2} yr^{-1}) from dated sediment cores. All cores were dated at Eawag (Swiss Federal Institute of Aquatic Science and Technology). Dating was performed by γ-ray measurements of ^{210}Pb and ^{137}Cs in a CANBERRA Li-Ge borehole detector.[31] Details on dating, literature references, sedimentation rates, and core numbers are provided in Table S-2. No dated cores were available from Lakes Murten, Lungeren, and Sihlsee. For these lakes, we estimated the N net sedimentation rate from a regression of NS vs. $[TP]_{mix}$ for lakes with available data (see Section 3.3).

2.2. Data Analyses

We calculated N removal efficiency (NRE; the fraction of the inflowing N removed by DN and NS) as (NS + DN)/N_{in}. Thus, the percentage of NRE contributed by NS is 100·NS/N_{in}, and the percentage contributed by DN is 100·DN/N_{in}.

We performed linear and nonlinear regressions of DN, NS, or NRE vs. various physical and chemical characteristics of the lakes, using the statistical software program Stata Version 14 (StataCorp LLC,

College Station, Texas, USA). When a linear-regression constant did not differ significantly from zero, we forced the regression through the origin and calculated a corrected R^2 value (Equation 10 in Kozak and Kozak[32]; see also Spiess and Neumeyer[33]). Because in nonlinear regression, R^2 and p values are not reliable indicators of the goodness of a regression fit and the statistical significance of predictor variables,[33] we also calculated Akaike Information Criteria (AIC) and bias-corrected AIC (AIC_c) to aid in model selection.[34] To avoid collinearity, predictor variables were not included in the same regression equation if the absolute value of the nonparametric, Spearman rank correlation coefficient ($r_{Spearman}$) between them was ≥0.9. Because most of the chemical data for the lakes are based on field measurements and thus are subject to more uncertainty than laboratory data, we inferred significant differences when $p > 0.1$.

3. Results and Discussion

3.1. Denitrification is controlled by nitrate concentration in the sediment-overlying water

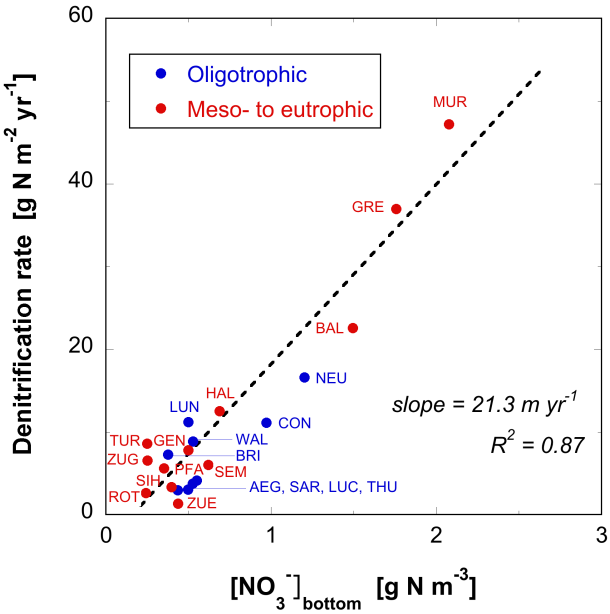
Denitrification rates in our set of 21 lakes were linearly related to the annual average $[NO_3^-]_{bottom}$ (Figure 2; R^2=0.87). Although additional variables that are discussed below can improve the prediction of DN rates, especially in several oligotrophic lakes, the main determinant of DN was $[NO_3^-]_{bottom}$. It indicates that DN within the top sediment layer is a first-order process in the concentration of NO_3^-. The slope s of 21.3 m yr^{-1} is the kinetic rate constant in the following equation (Model 6 in Table S-7):

$$\mathrm{DN}\ [\mathrm{g\,N\,m^{-2}\,yr^{-1}}] = \mathrm{N_{in}} - \mathrm{N_{out}} - \mathrm{NS} = \mathrm{s} \times [\mathrm{NO_3^-}]_{\mathrm{bottom}} - 3.88 \qquad (4)$$

The DN rate is limited by the transport of NO_3^-, which is proportional to the concentration gradient across the diffusive boundary layer and oxic surface sediment layer. Therefore, assuming similar diffusion lengths, DN rate is proportional to $[NO_3^-]_{bottom}$. This linkage has been demonstrated for the consumption of O_2 in lake sediments.[35] Because NO_3^-, unlike O_2, is not consumed immediately at the sediment surface but only in the suboxic zone, the thickness of the oxic sediment layer moderates the proportionality between $[NO_3^-]_{bottom}$ and DN rates. Because a greater thickness of the oxic sediment surface layer lengthens the diffusion path of NO_3^-, DN rates of oligotrophic lakes tend to be lower than for meso- to eutrophic lakes at similar $[NO_3^-]_{bottom}$. The oxic surface sediment layers of the meso- to

217 eutrophic lakes with oxic bottom waters were measured with microelectrodes to be 1 to 3.5 mm thick,
218 while diffusion lengths were about 2-fold greater (2.5 to 6 mm) in oligotrophic lakes.[36] Annual average
219 bottom-water O_2 concentrations of these oligotrophic lakes were all >8 mg L^{-1}, whereas for all other
220 lakes they were between 0.5 and 7 mg L^{-1}.

221



222

223 ***Figure 2:*** *Denitrification (DN) rates in 21 lakes increase as concentrations of NO_3^- in the*
224 *bottom water increase ($DN = 21.3 \cdot [NO_3^-]_{bottom} - 3.88$; $R^2 = 0.88$, $p < 0.0001$). Oligotrophic*
225 *lakes have $[TP]_{mix} \leq 10$ mg P m^{-3}, meso- to eutrophic lakes have $[TP]_{mix} > 10$ mg P m^{-3}. Lake*
226 *abbreviations are listed in Table 1; additional regression statistics are listed for Model 6 in*
227 *Table S-7.*

228

229 A similar regression with a slope of 20.4 m yr^{-1} was reported by Höhener and Gächter[18] for data from
230 10 Swiss lakes. Those 10 lakes are a subset of the 21 lakes in the current study but cover only 1 or 2
231 yr each between the 1950s and 1980s. Mengis et al.[15] determined mass-transfer coefficients for NO_3^-
232 of 21.7 and 21.4 m yr^{-1} for Lakes Baldegg and Zug, respectively (and which are also a subset of the
233 21 lakes), for individual years in the 1990s. Lower values of 7.2 to 9.2 m yr^{-1} were reported by Kelly et
234 al.[28] for acidified lakes in Ontario, Canada. Correlations of DN with NO_3^- concentrations in freshwater
235 lakes have also been reported,[8,10,13,14,37] although no kinetic constants were reported.

The relationship between $[NO_3^-]_{bottom}$ and DN rate has a mechanistic basis in microbial transformations at the oxic-anoxic interface of sediments, for which sufficient mineralizable organic matter is needed. Suboxic conditions are needed for denitrifying microorganisms to switch from O_2 to NO_3^- as an electron acceptor. These organisms are mainly heterotrophic and can use either electron acceptor for the mineralization of organic matter. A threshold of ~0.2 mg O_2 L^{-1} was suggested by Seitzinger et al.[23], below which DN can occur.

Several of the 21 lakes developed bottom-water anoxia toward the end of summer stratification (TUR, MUR, ROT, and GRE; Table S-3). However, DN rates remained unaffected and invariably related to $[NO_3^-]_{bottom}$, although the bottom-water stratification leading to the depletion of O_2 similarly caused depletion of NO_3^-. This is also valid for Lake Zürich, which does not mix to the deepest location annually, and meromictic Lake Zug, which has long-term bottom-water anoxia covering up to one-third of the lake area.

3.2. Nitrogen load and area-specific hydraulic load also predict denitrification rates

Although $[NO_3^-]_{bottom}$ appears to be a major, mechanistically-based factor controlling DN rate in these Swiss lakes, $[NO_3^-]_{bottom}$ is not measured in many lakes. Therefore, we tested if more-commonly-measured physical-chemical variables could also be used to predict DN rate relatively well.

Denitrification rates in coastal oceans, estuaries, rivers, and lakes increase in proportion to the areal TN load [34% of N load removed across 23 freshwater lakes in Saunders and Kalff[25] (R^2 = 0.80); 26% of N load removed across 26 freshwater and marine systems in Seitzinger et al.[23] (R^2 = 0.77)]. That statement is partly mechanistically-based, because $[NO_3^-]_{bottom}$ in our set of lakes is in part related to the areal dissolved N load to a lake (N_{in}) [e.g., $r_{Spearmen}$ between $[NO_3^-]_{bottom}$ and N_{in} in the 21 Swiss lakes is 0.61; Table S-6, Figure S-2]. However, the overlap between the set of oligotrophic lakes and the set meso- to eutrophic lakes in Figure 3a indicates that trophic state (which is influenced by physical characteristics of a lake in addition to N_{in}) alone is not a good predictor of DN rate.

Our analysis partly supports a relationship between DN and TN (Figure 3a), but not when six of the deep oligotrophic lakes (THU, WAL, BRI, LUC, CON, and SAR) and Lake Zürich (ZUE) were included in a linear regression of DN rate vs. N_{in} for all 21 lakes (R^2 = 0.65; Model 10 in Table S-7). The DN elimination factor of the remaining 14 lakes (i.e., the slope of DN rate vs. N_{in}) was 0.56 (R^2 = 0.97;

Model 25 in Table S-7), meaning that, on average, 56% of the dissolved N load to those lakes was eliminated by DN.

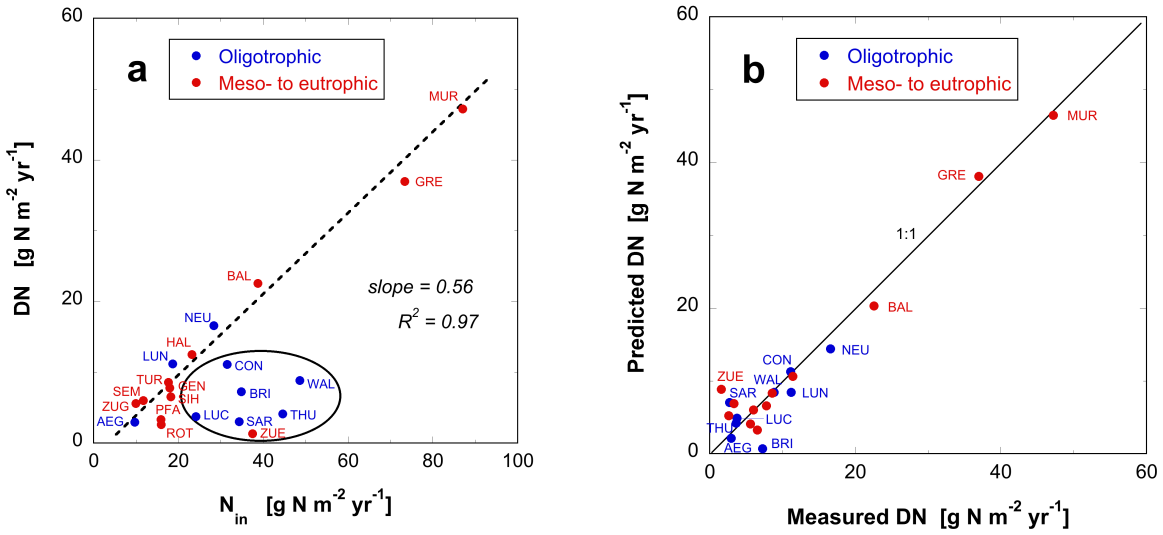


Figure 3: *a) Denitrification (DN) rate increases as areal N load (N_{in}) increases in 21 Swiss lakes. The encircled deep oligotrophic lakes (WAL, THU, SAR, BRI, CON, and LUC) and meromictic Lake Zürich (ZUE) were excluded from the regression calculation. b) Comparison of predicted and measured DN rates in 21 Swiss lakes. Predicted DN = $0.582 \cdot N_{in} - 0.302 \cdot Q_s$, ($R^2_{adjusted} = 0.93$, $p < 0.0001$, $n = 21$). The solid diagonal line is the 1:1 line of perfect agreement. Lake abbreviations are listed in Table 1; additional regression statistics are listed for Model 19 in Table S-7.*

Notably, DN rates of the six deep oligotrophic lakes fit relatively close to the regression line for DN vs. $[NO_3^-]_{bottom}$ (Figure 2), thus supporting that the major process in those lakes was DN at the sediment surface controlled by the NO_3^- concentration. But the combination of the large volume of these lakes relative to their surface area (i.e., large z_{mean}; 34-168 m) and the relatively short τ (1.6-4.2 yr) decreased the probability of the transfer of NO_3^- to the sediment-water interface and the subsequent redox reaction of NO_3^- with dissolved organic matter. Thus, a considerable fraction of the dissolved N load to those six lakes is exported via the lake outlet without interacting with the sediment. This is supported by the contrasting case of the deep oligotrophic Lake Neuchâtel, which has a maximum depth of 153 m but a much longer τ of 9.2 yr than the six outlier deep oligotrophic lakes. The relatively

long τ of Lake Neuchâtel may be the reason why this lake lies close to the DN vs. N_{in} regression line (Figure 3a).

Lake Zürich is also a prominent outlier from the DN vs. N_{in} regression line. Because deep mixing of that lake is infrequent due to warming in recent decades,[38,39] the supply of NO_3^- and O_2 to the sediment surface is impeded by the stratified deep water, thus restricting nitrification and subsequent DN. A similar situation occurs in meromictic Lake Idro in northern Italy.[21] As a consequence, the NO_3^- concentration at the sediment surface has become decoupled from the N load to Lake Zürich.

Lake Zug has a two-basin structure, in which the 198-m-deep south basin is meromictic with permanent anoxia below 120 m, but the north basin has oxic deep water throughout the year.[40] Because the catchment and thus the water flow feeding the south basin is very small, the north basin receives the main N load. Thus, the DN rate in the holomictic north basin dominates the whole-lake DN rate and plots close to the regression line in Figure 3a, along with the other holomictic eutrophic lakes and three of the nine oligotrophic lakes.

Despite their potential importance to DN, τ and z_{mean} alone were poor individual predictors of DN rate across all 21 lakes (R^2 = 0.01 and 0.07, respectively; Models 2 and 3 in Table S-7). However, a linear combination of N_{in} (g N m^{-2} yr^{-1}), τ (yr), and z_{mean} (m) as independent variables in a multiple regression improved the overall prediction of DN rate (g N m^{-2} yr^{-1}) for all 21 lakes:

$$DN = 0.555 \cdot N_{in} + 0.926 \cdot \tau - 0.0824 \cdot z_{mean} - 5.14 \qquad (5)$$

with $R^2_{adjusted}$ = 0.78 and highly significant coefficients for all three predictor variables ($p \leq 0.006$; Model 24 in Table S-7). But four lakes (AEG, BRI, SAR, and ZUE) remained as major outliers (i.e., predicted DN/measured DN ratio was 0.03-0.38 or 3.94-9.51; Figure S-3a).

As an alternative to Equation 5, τ and z_{mean} can be combined into a single variable we define as the area-specific hydraulic loading rate, Q_s (m yr^{-1}):

$$Q_s = \frac{Q}{A_0} = \frac{\frac{V}{\tau}}{\frac{V}{z_{mean}}} \cdot = \frac{z_{mean}}{\tau} \qquad (6)$$

where A_0 is surface area of the lake (m^2), Q is hydraulic flow rate into the lake (m^3 yr^{-1}), and V is the lake volume (m^3). Although Q_s alone was a poor individual predictor of DN rate across all 21 lakes (R^2

= 0.06; Model 1 in Table S-7), a linear combination of N_{in} and Q_s (m yr^{-1}) as independent variables improved the overall prediction of DN rate in the 21 lakes even more than Equation 5:

$$DN = 0.582 \cdot N_{in} - 0.302 \cdot Q_s \quad (7)$$

with $R^2_{adjusted}$ = 0.93 and the lowest AIC_c of all models tested, and the coefficients for both predictor variables were highly significant ($p < 0.001$; Model 19 in Table S-7). We forced that regression through the origin because the intercept in a full model was not statistically significant (Model 18 in Table S-7). However, two lakes remained as major outliers (i.e., predicted DN/measured DN ratio after applying Equation 7 was 0.08 for BRI and 6.50 for ZUE; Figures 3b and S-3b). No other physical-chemical variables significantly increased the percentage of variance accounted for by Equation 7 or resulted in a lower AIC_c (Table S-7).

Therefore, we conclude that at a macroscale, a combination of external N loading and physical characteristics (e.g., depth and lake flushing rate) contribute to DN rate. But at a microscale, DN is more-directly controlled mainly by $[NO_3^-]_{bottom}$ and the thickness of the oxic upper layer of the sediment, both of which are at least partly manifestations of the macroscale characteristics.

3.3. Total phosphorus concentration predicts net sedimentation of nitrogen

For a subset of 12 of the 21 Swiss lakes, $[TP]_{mix}$ (mg P m^{-3}) was a strong predictor of NS (g N m^{-2} yr^{-1}):

$$NS = 0.137 \cdot [TP]_{mix} + 0.798 \quad (8)$$

with R^2 = 0.96, $p < 0.0001$ (Figure 4 and Model 22 in Table S-8). The four circled oligotrophic lakes in Figure 4 (SAR, WAL, THU, and BRI) were excluded from the regression because they are prone to floodwaters importing high loads of allochthonous erosion material, thus increasing NS excessively (see discussion below). Additionally, three lakes (LUN, MUR, and SIH) could not be included because no sediment data were available. Two other lakes (ZUG and GRE) were excluded from the regression because their $[TP]_{mix}$ were excessively high and thus might have had a low correlation with productivity [i.e., other physical-chemical factors (e.g., limited light penetration because of shading by near-surface algal mats) probably limited production]. Overall, the relationship in Figure 4 suggests that, for most of these lakes, primary production was the main cause for the generation of PN and its

net deposition in the sediment (see Table S-2 for details on sediment analyses, including literature references documenting the values used).

The four oligotrophic lakes circled in Figure 4 receive high loads of allochthonous particulate material from their alpine catchments, especially during stormwater events (SAR[17]; WAL[41]; THU[42]; BRI[43]). The mass accumulation rates of N in the sediments of these lakes (3.5-6.9 g N m^{-2} yr^{-1}) were the highest of all oligotrophic lakes, but only a small fraction of the high allochthonous input of particulate organic material to lakes is bioavailable[44]. The strong relationship between NS and $[TP]_{mix}$ allowed us to estimate the corresponding autochthonous fraction of NS in those four oligotrophic lakes using Equation 8. Additionally, we used this relationship to estimate the unmeasured NS rates of Lakes Murten, Lungeren, and Sihlsee.

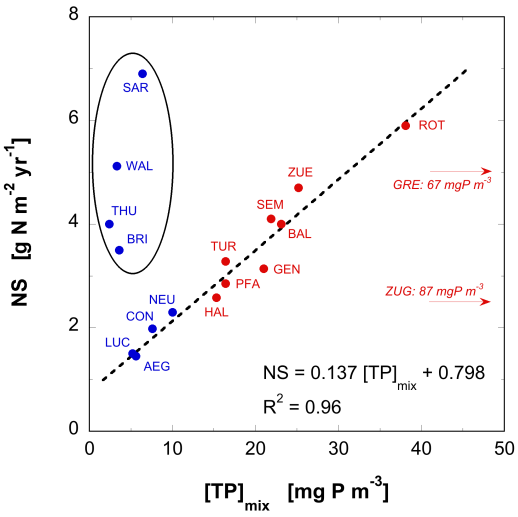


Figure 4: *Net N sedimentation rate (NS) increases as concentration of total phosphorus at lake overturn $[TP]_{mix}$ increases, except when lakes receive high loads of allochthonous material from their catchment (e.g., the four circled lakes), or when $[TP]_{mix}$ exceeds a concentration at which other physical-chemical factors control primary production. The four allochthonously-dominated lakes (SAR, WAL, THU, and BRI) and the hypertrophic Greifensee (GRE) and Lake Zug (ZUG) were excluded from the regression. Lake abbreviations are listed in Table 1; additional regression statistics are listed for Model 22 in Table S-8.*

3.4. Area-specific hydraulic loading rate predicts nitrogen removal efficiency

In compilations of lakes by Finlay et al.[7], Rissanen et al.[13], Seitzinger et al.[23], Saunders and Kalff[25], and Kelly et al.[28], τ was the main predictor of N removal efficiency (NRE; i.e., the fraction of the imported TN removed by DN and NS). The underlying concept is that longer τ increases the probability for the redox reaction of NO_3^- with the sediment organic matter to result in DN; and it also increases generation of PN in the productive zone, thus enhancing subsequent settling and sediment burial.

Supporting those studies, NRE was positively correlated with τ in the 21 Swiss lakes ($r_{Spearman}$ = 0.60; Table S-6). Additionally, NRE was weakly negatively correlated with z_{mean} ($r_{Spearman}$ = -0.34), in part because four deep oligotrophic lakes had much lower DN rates than would be predicted from their N load alone (Figure 3). Therefore, in order to concurrently capture the contributions of τ and z_{mean}, we modeled NRE as a Michaelis-Menton type function of the inverse of Q_s (i.e., as a function of τ/z_{mean}):

$$\mathrm{NRE} = \frac{\frac{1}{Q_s}}{\left(K+\frac{1}{Q_s}\right)} = \frac{1}{K \cdot Q_s + 1} \qquad (9)$$

with K = 0.0705 yr m^{-1} (R^2_{adj} = 0.84, and the second-lowest AIC_c, n = 21; Figure 5 and Model 19 in Table S-9). According to this function, NRE approaches zero when τ approaches zero and/or when z_{mean} approaches infinity, while NRE approaches 1 when τ approaches infinity and/or when z_{mean} approaches zero (consistent with the data in Figure 5).

NRE was not as strongly related with linear models containing any of the other potential predictor variables alone or various combinations of those variables (Table S-9). Linear combinations of $[TP]_{mix}$ or $[NO_3^-]_{bottom}$ with $1/(K \cdot Q_s + 1)$ increased the R^2_{adj} by only 1%, and AIC_c was lowered by only 0.3 AIC units (for $[TP]_{mix}$) or raised by only 0.6 AIC units (for $[NO_3^-]_{bottom}$; Models 23 and 29 in Table S-9). Thus, because the AIC_c values for all three regression models (19, 23, and 29 in Table S-9) are within 2 AIC units of each other, each model has substantial support.[34] However, we recommend the simpler Equation 9 as a predictor of NRE, because it is more parsimonious (i.e., fewer predictor variables) and does not require measurements of $[TP]_{mix}$ and/or $[NO_3^-]_{bottom}$; and little additional predictability is gained by adding an extra variable.

For three reasons, the inverse relationship between NRE and Q_s is conceptually plausible. First, as τ increases, the probability increases that dissolved N is taken up by phytoplankton and then settles as

PN to the lake bottom, where it can be (1) transformed by sequential nitrification and DN or (2) buried as refractory N in the sediment. Second, because z_{mean} relates the lake's volume to its surface area and thus approximately to its sediment area, decreasing z_{mean} is equivalent to increasing the extent of DN and/or NS per unit of lake volume. Thus, as z_{mean} decreases, NRE is likely to increase. Third, assimilatory NO_3^- reduction, the key process for the generation of PN from NO_3^- and subsequent N elimination by burial, is strongly coupled to photosynthesis. Thus, NRE is more likely to decrease as the lake's depth increasingly exceeds the vertical extent of its trophic layer, because transient removal of N from the dissolved phase by photosynthesis is more likely to be reversed as dead organic matter descends farther through a hypolimnion.

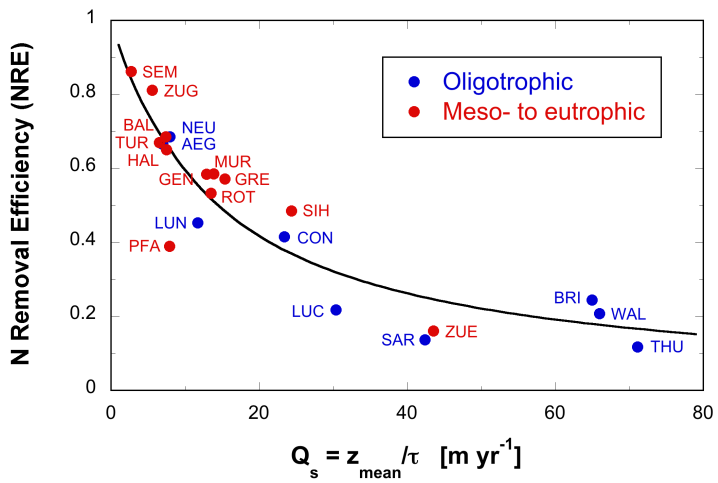


Figure 5: *Nitrogen removal efficiency (NRE, the fraction of the N load removed by denitrification and burial in the sediments) decreases as area-specific hydraulic loading rate (Q_s) increases. The line is a Michaelis-Menton type function [$NRE = 1/(K \cdot Q_s + 1)$], with $K = 0.0705$ yr m^{-1} ($R^2_{adj} = 0.84$, n=21). Z_{mean} is average depth, and τ is water residence time; lake abbreviations are listed in Table 1; additional regression statistics are listed for Model 19 in Table S-9.*

In Figure 5, shallow lakes with long τ tend to be eutrophic (red circles) while deep lakes with short τ tend to be oligotrophic (blue circles). But because those two trophic categories overlap, trophic state does not appear to be the main determinant of NRE. Instead, the combination of τ and z_{mean} as an

410 inverse function of Q_s is a much better individual predictor than traditional determinants of trophic
411 state such as $[TP]_{mix}$. As such, the influence of $[TP]_{mix}$, N_{in}, and $[NO_3^-]_{bottom}$ might be subsumed by Q_s
412 as an overall general predictor of NRE.

413 Finley et al.[7] reported that eutrophic lakes were almost three times more efficient in the removal of TN
414 loads than were unproductive lakes. The results from our set of lakes tend to qualitatively support that
415 observation, because meso- to eutrophic lakes were on average more efficient in removing N from the
416 water column [NRE was 0.62 ± 0.14 (mean ± s.d.), with the exception of Lake Zürich] than
417 oligotrophic lakes (NRE was 0.35 ± 0.22). However, only 29 ± 18% and 21 ± 10% of the total N
418 removal [i.e., mean ± s.d. of 100·NS/(ND + NS) for each of the 12 meso- and eutrophic lakes and 9
419 oligotrophic lakes] could have been influenced by primary production (leading to net sediment burial)
420 in those two trophic categories, respectively. The major percentage of N elimination was by DN, which
421 is mainly controlled at the microscale by $[NO_3^-]_{bottom}$ (Figure 2). Therefore, we conclude that in these
422 holomictic lakes, a combination of τ and z_{mean} is a better predictor of NRE than trophic state alone.
423 However, trophic state might play an underlying mechanistic role that is subsumed within Q_s, because
424 at a macroscale, N_{in} helps to strongly predict DN rate; and $[TP]_{mix}$ is a major determinant of NS.
425 Additionally, the importance of Q_s in predicting DN rates (Equation 7) also helps to explain why it is
426 the best predictor of NRE.

427

428 **3.5. Small decrease of N removal efficiency during oligotrophication**

429 To summarize, two major pathways (DN and NS) are responsible for the removal of N from lakes. In
430 each of the 21 Swiss lakes, and independent of its trophic state, DN was the more efficient pathway
431 [DN accounted for 71-79% (average of 75%) of the removed N, while NS accounted for 21-29%
432 (average of 25%)]. In absolute terms, however, meso- to eutrophic lakes removed more N per unit
433 area via burial in the sediments than oligotrophic lakes (Figure 4). In contrast, DN was directly related
434 to $[NO_3^-]_{bottom}$, which was not related to trophic state (Figure 2) but is partly related to N loading among
435 several other physical characteristics of a lake.

436 Finlay et al.[7] speculated that P management of lakes aimed at decreasing bioavailable P
437 concentrations would lead to a net decrease in NRE and thus an increased N export from lakes. Our
438 results demonstrate a weak tendency for NRE to decrease at lower $[TP]_{mix}$ (R^2 = 0.22; Model 4 in

Table S-9); however, this relationship only occurs because of a proportional decrease of the N burial in the sediments, which in total contributes only 21 to 29% of the total N removal. As a consequence, N export will not increase substantially even if N_{in} remains unchanged, because $[NO_3^-]_{bottom}$ (which is a major determinant of DN rate) tends to decrease as N_{in} decreases (Figure S-2). Because (1) N_{out} equals $(1 – NRE)\cdot N_{in}$ and (2) NRE tends to decrease as a lake undergoes oligotrophication, a decrease or increase in N export from an oligotrophying lake depends on whether the accompanying decrease in N_{in} (if any) can at least compensate for the decreased NRE. That is, P management that only removes P will slightly increase N export to downstream waterbodies. Therefore, we conclude that P management must be concurrently accompanied by N removal (e.g., decreased application of fertilizers and/or manure on agricultural lands in the catchment) for the amelioration of the excessive N loads from catchments to inland surface waters and eventually to coastal ocean waters. That is, clean lakes can be dirtier or cleaner lakes (sensu Bernhardt [6]), depending on the details of the oligotrophication efforts.

Supporting Information

Physical characteristics and general description of the state of the lakes considered in this study, description of the catchment model MOFIFFUS, experimental details on the determination of N net sedimentation rates from sediment cores, comparison of N loads of two lakes estimated with MODIFFUS and calculated from up to ~20 yr of monitoring data, and tables of statistical results.

Acknowledgements

Rosi Siber is greatly acknowledged for the operation of the catchment model MODIFFUS. This work was financially supported by the Swiss National Science Foundation Grant no. 205321_169142.

Author information

Corresponding author: *Phone: +41-58-765 21 49; e-mail: beat.mueller@eawag.ch

Notes: The authors declare no competing financial interest.

Funding sources

This work was partly funded by the Swiss National Science Foundation Grant 205321_169142.

References

1 Grizzetti, B.; Bouraoui, F.; Billen, G.; van Grinsven, H.; Cardoso, A. C.; Thieu, V.; Garnier, J.; Curtis, C.; Howarth, R.; Johnes, P. Nitrogen as a threat to European water quality. In *The European Nitrogen Assessment*; Sutton, M. A., Howard, C. M., Erisman, J. W., Billen, G., Bleeker, A., Grennfelt, P., van Grinsven, H., Grizzetti, B., Eds.; Cambridge University Press: Cambridge 2011; chapter 17, pp. 379-404.

2 Canfield, D. E.; Glazer, A. N.; Falkowski, P. G. The evolution and future of earth's nitrogen cycle. *Science* **2010**, *330*, 192-196; DOI 10.1126/science.1186120.

3 Galloway, J. N.; Townsend, A. R.; Erisman, J. W.; Bekunda, M.; Cai, Z.; Freney, J. R.; Martinelli, L. A.; Seitzinger, S. P.; Sutton, M. A. Transformation of the nitrogen cycle: Recent trends, questions, and potential solutions. *Science* **2008**, *320*, 889-892, 1136674; DOI 10.1126/science.

4 Kuypers, M. M. M.; Marchant, H. K.; Kartal, B. The microbial nitrogen-cycling network. *Nat. Rev. Microbiol.* **2018**, *16* (5), 263-276; DOI 10.1038/nrmicro.2018.9.

5 Burgin, A. J.; Hamilton, S. K. Have we overemphasized the role of denitrification in aquatic ecosystems? A review of nitrate removal pathways. *Front. Ecol. Environ.* **2007**, *5* (2), 89-96; DOI 10.1890/1540-9295(2007)5[89:hwotro]2.0.co;2.

6 Bernhardt, E. S. Cleaner lakes are dirtier lakes. Science 2013, 342, 205-206; DOI 10.1126/science.1245279.

7 Finlay, J. C.; Small, G. E.; Sterner R. W. Human influence on nitrogen removal in lakes. *Science* **2013**, *342*, 247-250; DOI 10.1126/science.1242575.

8 Álvarez-Cobelas, M.; Piña-Ochoa, E.; Sanchez-Carillo, S.; Delgado-Huertas, A. Spatial variability of denitrification along a nitrate-rich seepage chain of lakes (Ruidera Natural Park, central Spain). *Limnetica* **2019**, *38* (2), 607-621; DOI 10.23818/limn.38.35.

9 Seitzinger, S. P. Denitrification in freshwater and coastal marine ecosystems: Ecological and geochemical significance. *Limnol. Oceanogr*. **1988**, *33* (4, part 2), 702-724.

10 Piña-Ochoa E.; Álvarez-Cobelas M. Denitrification in aquatic environments: A cross-system analysis. *Biogeochem*. **2006**, *81*, 111-130.

11 Harrison, J. A.; Maranger, R. J.; Alexander, R. B.; Giblin, A. E.; Jacinthe, P.-A.; Mayorga, E.; Seitzinger, S. P.; Sobota, D. J.; Wollheim, W. M. The regional and global significance of nitrogen removal in lakes and reservoirs. *Biogeochem*. **2009**, *93*, 143-157; DOI 10.1007/s10533-008-9272-x.

12 Rissanen, A. J.; Tiirola, M.; Ojala, A. Spatial and temporal variation in denitrification and in the denitrifier community in a boreal lake. *Aquat. Microb. Ecol*. **2011**, *64*, 27-40; DOI 10.3354/ame01506.

13 Rissanen, A. J.; Tiirola, M.; Hietanen, S.; Ojala, A. Interlake variation and environmental controls of denitrification across different geographical scales. *Aquat. Microb. Ecol*. **2013**, *69*, 1-16; DOI 10.3354/ame01619.

14 McCrackin, M. L.; Elser, J. J. Atmospheric nitrogen deposition influences denitrification and nitrous oxide production in lakes. *Ecology* **2010**, *91* (2), 528-539; DOI 10.1890/08-2210.1.

15 Mengis, M. M.; Gächter, R.; Wehrli, B.; Bernasconi, S. Nitrogen elimination in two deep eutrophic lakes. *Limnol Oceanogr*. **1997**, *42* (7), 1530-1543.

16 Bruesewitz, D. A.; Tank, J. L.; Hamilton, S. K. Incorporating spatial variation of nitrification and denitrification rates in whole-lake nitrogen dynamics. *J. Geophys. Res*. **2012**, *117*, G00N07; DOI 10.1029/2012JG002006.

17 Müller, B.; Thoma, R.; Baumann, K. B. L.; Callbeck, C. M.; Schubert, C. J. Nitrogen removal processes in lakes of different trophic states from on-site measurements and historic data. *Aquatic Sci*. **2021**, *83* (2), 37; DOI 10.1007/s00027-021-00795-7.

18 Höhener, P.; Gächter, R. Prediction of dissolved inorganic nitrogen (DIN) concentrations in deep, seasonally stratified lakes based on rates of DIN input and N removal processes. *Aquatic Sci.* **1993**, *55* (2), 112-131.

19 Li, J.; Katsev, S. Nitrogen cycling in deeply oxygenated sediments: Results in Lake Superior and implications for marine sediments. *Limnol. Oceanogr*. **2014**, *59* (2), 465-481; DOI 10.4319/lo.2014.0465.

20 Small, G. E.; Cotner, J. B.; Finlay, J. C.; Stark, R. A.; Sterner, R. W. Nitrogen transformation at the sediment-water interface across redox gradients in the Laurentian Great Lakes. *Hydrobiologia* **2014**, *731*, 95-108; DOI 10.1007/s10750-013-1569-7.

21 Nizzoli, D.; Bartoli, M.; Azzoni, R.; Longhi, D.; Castaldelli, G.; Viaroli, P. Denitrification in a meromictic lake and its relevance to nitrogen flows within a moderately impacted forest catchment. *Biogeochem*. **2017**, *137*, 143-161; DOI 10.1007/s10533-017-0407-9.

22 Lau, M. P.; Valerio, G.; Pilotti, M.; Hupfer M. Intermittent meromixis controls the trophic state of warming deep lakes. *Sci. Rep*. **2020**, *10*, 12928; https://www.nature.com/articles/s41598-020-69721-5.

23 Seitzinger, S.; Harrison, J. A.; Böhlke, J. K.; Bouwman, A. F.; Lowrance, R.; Peterson, B.; Tobias, C.; Van Drecht, G. Denitrification across landscapes and waterscapes: A synthesis. *Ecol. Appl.* **2006**, *16* (6), 2064-2090; DOI 10.1890/1051-0761(2006)016[2064:dalawa]2.0.co;2.

24 David, M. B.; Wall, L. G.; Royer, T.; Tank, J. L. Denitrification and the nitrogen budget of a reservoir in an agricultural landscape. *Ecol. Appl*. **2006**, *16* (6), 2177-2190.

25 Saunders D.L.; Kalff J. Nitrogen retention in wetlands, lakes and rivers. *Hydrobiologia* **2001**, *443*, 205-212.

26 Fleischer, S.; Stibe, L. Drainage basin management-reducing river transported nitrogen. *Verh. int. Ver. Limnol*. **1991**, *24*, 1753-1755.

27 Howarth, R. W.; Billen, G.; Swaney, D.; Townsend, A.; Jaworski, N.; Lajtha, K.; Downing, J. A.; Elmgren, R.; Caraco, N.; Jordan, T.; Berendse, F.; Freney, J.; Kudeyarov, V.; Murdoch, P.; Zhu, Z-L. Regional nitrogen budgets and riverine N ; P fluxes for the drainages to the North Atlantic Ocean: Natural and human influences. *Biogeochem*. **1996**, *35*, 75-139.

28 Kelly, C. A.; Rudd, J. W. M.; Hesslein, R. H.; Schindler, D. W.; Dillon, P. J.; Driscoll, C. T.; Gherini, S. A.; Hecky, R. E. Prediction of biological acid neutralization in acid-sensitive lakes. *Biogeochem*. **1987**, *3*, 129-140.

29 Hürdler, J.; Prasuhn, V.; Spiess, E. *Abschätzung diffuser Stickstoff- und Phosphoreinträge in die Gewässer der Schweiz MODIFFUS 3.0* (in German). Report to the Swiss Federal Office for the Environment, 3003 Bern, Switzerland, **2015**. http://link.ira.agroscope.ch/de-CH/publication/35052.

30 Internationale Gewässerschutzkommission für den Bodensee (IGKB). Dem Bodensee in den Abflussjahren 1996 und 1997 zugeführte Stofffrachten. Ber. Int. Gewässerschutzkomm. Bodensee **2000**, *53*, p. 24, Table 7, ISSN 1011-1263; https://www.igkb.org/fileadmin/user_upload/dokumente/publikationen/blaue_berichte/blauer_bericht_53.pdf.

31 Benoit, G.; Rozan T. F. 210Pb and 137Cs dating methods in lakes: A retrospective study. *J. Paleolim.* **2001**, *25*, 455-465.

32 Kozak, A.; Kozak, R. A. Notes on regression through the origin. *Forestry Chron.* **1995**, *71* (3), 326-330.

33 Spiess, A.-N.; Neumeyer, N. An evaluation of R2 as an inadequate measure for nonlinear models in pharmacological and biochemical research: A Monte Carlo approach. *BMC Pharmacol.* **2010**, *10*, 6; DOI 10.1186/1471-2210-10-6 ().

34 Burnham, K. P.; Anderson, D. R. Multimodel inference: Understanding AIC and BIC in model selection. *Sociol. Meth. Res.* **2004**, *33*, 261-304.

35 Müller, B.; Bryant, L. D.; Matzinger, A.; Wüest, A. Hypolimnetic oxygen depletion in eutrophic lakes. *Environ. Sci. Technol*. **2012**, *46*, 9964-9971; DOI 10.1021/es301422r.

36 Steinsberger T.; Schwefel R.; Wüest A.; Müller B. Hypolimnetic oxygen depletion rates in deep lakes: Effect of trophic state and organic matter accumulation. *Limnol. Oceanogr*. **2020**, *65* (12), 3128-3138; DOI 10.1992/lno.11578.

37 Hasegawa, T.; Okino, T. Seasonal variation of denitrification rate in Lake Suwa sediment. *Limnology* **2004**, *5*, 33-39; DOI 10.1007/s10201-003-0109-y.

38 Yankova, Y.; Neuenschwander, S.; Köster, O.; Posch, T. Abrupt stop of deep water turnover with lake warming: Drastic consequences for algal primary producers. *Sci. Rep*. **2017**, *7*, 13770; DOI 10.1038/s41598-017-13159-9.

39 Knapp, D.; Castro, B. F.; Marty, D, Loher, E.; Köster, O.; Wüest, A.; Posch, T. The red harmful plague in times of climate change: Blooms of the cyanobacterium Planktothrix rubescens triggered by stratification dynamics and irradiance. *Front. Microbiol*. **2021**, *12*, 705914; DOI 10.3389/fmicb.2021.705914.

40 Schwefel, R.; Müller, B.; Boisgontier, H.; Wüest, A. Global warming affects nutrient upwelling in deep lakes. *Aquatic Sci*. **2019**, *81*, 50; DOI 10.1007/s00027-019-0637-0.

41 Lambert A.; Hsü K.J. Non-annual cycles of varve-like sedimentation in Walensee, Switzerland. *Sedimentology* **1979**, *26*, 453-461; DOI 10.1111/j.1365-3091.1979.tb00920.x.

42 Wirth S.; Girardclos S.; Rellstab C.; Anselmetti F. The sedimentary response to a pioneer geo-engineering project: Tracking the Kander River deviation in the sediments of Lake Thun (Switzerland). *Sedimentology* **2011**, *58*, 1737-1761; DOI 10.1111/j.1365-3091.2011.01237.x.

43 Anselmetti, F.; Bühler, R.; Finger, D.; Girardclos, S.; Lancini, A.; Rellstab, C.; Sturm, M. Effects of hydropower dams on particle transport and lacustrine sedimentation. *Aquatic Sci*. **2007**, *69*, 179-198; DOI 10.1007/s00027-007-0875-4.

44 Sobek, S.; Durisch-Kaiser, E.; Zurbrügg, R.; Wongfun, N.; Wessels, M.; Pasche, N.; Wehrli, B. Organic carbon burial efficiency in lake sediments controlled by oxygen exposure time and sediment source. *Limnol. Oceanogr*. **2009**, *54* (6), 2243-2254; DOI 10.4319/lo.2009.54.6.2243.

Table 1: Physical characteristics and chemical monitoring data of the 21 Swiss lakes. [a]

Lake	Abbrev.	Time range	z_{mean}	τ	$[TP]_{mix}$	$[NO_3^-]_{mix}$	$[NO_3^-]_{bottom}$	$[TN]_{mix}$	LN_{in}	LN_{out}	N_{in}	DN	NS	NRE	Q_s
			m	yr	mg P m^{-3}	g N m^{-3}	g N m^{-3}	g N m^{-3}	t N yr^{-1}	t N yr^{-1}	g N $m^{-2}yr^{-1}$	g N $m^{-2}yr^{-1}$	g N $m^{-2}yr^{-1}$	-	m yr^{-1}
Aegeri	AEG	2000-2018	48	4.1	5.6	0.409	0.435	*0.503*	71	39	44.1	2.94	1.45	0.453	11.7
Baldegg	BAL	2000-2018	33	4.5	23.1	1.593	1.494	*1.959*	202	64	48.7	22.6	4.00	0.686	7.39
Brienz	BRI	2000-2017	168	2.6	3.6	0.374	0.377	0.431	1039	784	34.9	7.24	*1.30*	0.245	65.0
Constance	CON	2000-2016	99	4.2	7.6	0.959	0.971	0.959	15176	8861	24.1	11.1	1.98	0.416	23.4
Geneva	GEN	2000-2016	153	11.9	21.0	0.564	0.500	0.641	10454	4335	9.7	7.78	3.14	0.585	12.9
Greifensee	GRE	2000-2017	18	1.1	67.1	1.590	1.758	2.176	620	266	18.6	37.0	5.00	0.572	15.4
Hallwil	HAL	2000-2018	29	3.8	15.3	0.800	0.689	1.204	231	81	34.1	12.5	2.58	0.651	7.48
Lucerne	LUC	2000-2020	107	3.5	5.2	0.609	0.525	0.691	2683	2099	31.5	3.75	1.50	0.218	30.4
Lungeren	LUN	2003-2015	33	4.2	5.7	0.537	0.500	0.784	37	12	28.4	11.2	*1.59*	0.686	7.93
Murten	MUR	2001-2017	23	1.7	21.4	2.483	2.076	2.770	2002	830	18.2	47.2	*3.78*	0.586	13.9
Neuchâtel	NEU	2000-2018	64	9.2	10.0	1.230	1.201	*1.513*	6185	2068	22.1	16.6	2.30	0.666	6.97
Pfäffikon	PFA	2000-2017	19	2.4	16.4	0.801	0.397	1.268	51	31	15.9	3.35	2.85	0.390	7.89
Rotsee	ROT	2000-2015	8.8	0.65	38.1	0.499	0.242	*0.614*	7.5	3.5	17.7	2.60	5.90	0.533	13.5
Sarnen	SAR	1994-2018	34	0.79	6.4	0.542	0.497	0.699	245	212	18.0	3.02	*1.69*	0.137	42.4
Sempach	SEM	2000-2018	46	16.8	21.9	0.540	0.619	*0.664*	165	23	87.0	6.02	4.10	0.862	2.71
Sihlsee	SIH	2014-2016	9.0	0.37	10.9	0.232	0.251	0.423	203	104	11.7	6.53	*2.31*	0.485	24.4
Thun	THU	2000-2017	135	1.9	2.4	0.563	0.550	0.645	2135	1883	38.7	4.13	*1.13*	0.118	71.1
Türlersee	TUR	2000-2017	13.1	2.0	16.4	0.618	0.250	0.975	8.8	2.9	37.8	8.58	3.28	0.670	6.50
Walensee	WAL	2000-2018	104	1.6	3.3	0.528	0.527	*0.649*	1173	929	16.0	8.85	*1.25*	0.208	66.0
Zug	ZUG	2000-2016	84	15.1	87.0	0.304	0.352	*0.374*	381	72	73.4	5.62	2.46	0.812	5.56
Zürich	ZUE	2000-2017	49	1.1	25.2	0.665	0.437	0.803	2498	2096	9.95	1.33	4.70	0.161	43.5

[a] Time range of monitoring data selected, mean depth (z_{mean}), water residence time (τ), volume-weighted mean concentrations of total phosphorus after winter mixing ($[TP]_{mix}$; calculated as an average of the years 2010-2018), nitrate concentrations after winter mixing ($[NO_3^-]_{mix}$), nitrate concentration above sediment surface ($[NO_3^-]_{bottom}$), total nitrogen after winter mixing ($[TN]_{mix}$), annual load of dissolved N[29] plus the load from sewage treatment plants and

atmospheric deposition on the lake surface (LN_{in}), area-specific annual load of dissolved N (N_{in}), mean annual export of total N (LN_{out}), denitrification rate (DN), net N sedimentation rate (NS), nitrogen removal efficiency (NRE), and area-specific hydraulic loading rate ($Q_s = Q/A_0$, where Q = hydraulic loading rate and A_0 = surface area of lake). Italicized numbers for $[TN]_{mix}$ were estimated as described in Note S-1 and Figure S-1. Italicized numbers for NS are regression-estimated NS values according to Figure 4 (measured values are listed in Table S-2). Additional orographic and hydrologic data are listed in Table S-1.