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Reducing Phosphorus to Curb Lake Eutrophication is a Success

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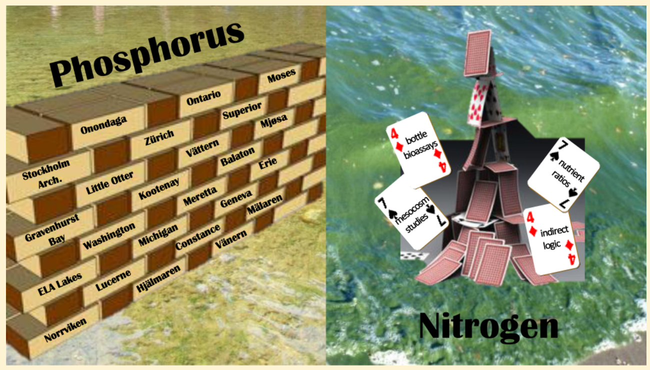
ABSTRACT: As human populations increase and land-use intensifies, toxic and unsightly nuisance blooms of algae are becoming larger and more frequent in freshwater lakes. In most cases, the blooms are predominantly blue-green algae (Cyanobacteria), which are favored by low ratios of nitrogen to phosphorus. In the past half century, aquatic scientists have devoted much effort to understanding the causes of such blooms and how they can be prevented or reduced. Here we review the evidence, finding that numerous long-term studies of lake ecosystems in Europe and North America show that controlling algal blooms and other symptoms of eutrophication depends on reducing inputs of a single nutrient: phosphorus. In contrast, small-scale experiments of short duration, where nutrients are added rather than removed, often give spurious and confusing results that bear little relevance to solving the problem of cyanobacteria blooms in lakes.



INTRODUCTION

Eutrophication, the scientific term that describes algal blooms and associated problems that are caused by the response of natural waters to excessive inputs of nutrients, is one of the greatest environmental problems facing humanity. Growing human populations produce increasing volumes of waste and require increasing areas and intensity of land use to feed the growing population. The effects of higher water temperatures under global warming act in concert with increasing nutrients, to paint a bleak future for a problem that is already acute.[1]

The scientific study of algal blooms dates from the earliest days of the science of limnology (the study of inland waters). Limnologists used quantitative methods to observe increases in the size and duration of algal blooms in Zürichsee, Switzerland as early as 1890.[2] The term *eutrophic*, derived from Greek meaning rich in food, was first applied to lakes with algal blooms by Einar Naumann in the early years of the 20th century. The term *eutrophication* was coined to describe the process by which lakes become nutrient enriched, which was believed to occur naturally as lakes slowly filled with sediment, concentrating nutrients in less and less water.

By the mid-20th century it was clear that human activity was accelerating the eutrophication process. The problem was found to be more widespread and to have occurred much earlier than had been previously realized, in lakes near centers of human activity. Hutchinson et al.[3] were able to deduce from lake sediments that algal blooms developed in Lago di Monterosi as the Romans built and used the adjacent Via Appia. Even in the high arctic, eutrophication occurred at sites where prehistoric Inuit whalers butchered their prey on the shores of freshwater lakes.[4] As human populations increased exponentially and modern intensive agriculture developed in Europe and North America in the early 20th century, the problem reached epidemic proportions. The term *cultural eutrophication* was used to refer to the acceleration of bloom development and other symptoms of eutrophication by human activity.

Until the mid-20th century, the causes of eutrophication were a mystery. By comparing the chemical composition of algae with water chemistry, limnologists were able to deduce that the problem had something to do with nutrient enrichment. Logically, the nutrients with the highest ratio of concentration in algae relative to concentration in lake water were suspected to be the culprits, and that led to an early focus on phosphorus, nitrogen, and carbon. Later studies suggested that the abundance of trace elements, and even major ions also might play a role. Indeed, speakers at a 1967 symposium on the topic sponsored by the U.S. National Academy of Sciences (NAS) presented a wide variety of opinions on the causes of eutrophication and did not reach a consensus as to what nutrients must be controlled to rein in the problem.[5]

The rapid development of modern water chemistry in the latter half of the 20th century allowed limnologists to quickly

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narrow down the precise causes of eutrophication and to formulate remediation measures. In the 1970s, phosphorus reduction was widely adopted as the solution to reducing eutrophication of the North American Great Lakes and other lakes in Europe and North America. This early choice of phosphorus regulation was based largely on three pieces of evidence: an extensive review of the eutrophication problem, whole-lake experiments, and a well-documented successful case history of recovery, as described below.

■ VOLLENWEIDER'S REVIEW

Richard Vollenweider's classic review of the eutrophication problem[6] was already in manuscript form at the time of the 1967 NAS conference, but despite being one of Europe's most senior limnologists, he was not among the invited speakers. Vollenweider reviewed hundreds of studies at all scales to conclude that the most likely causes of eutrophication were increases in the nutrients phosphorus and nitrogen from sources outside the lake. He proposed the first simple models relating inputs of those elements to the magnitude of algal blooms in lakes.

The significance of Vollenweider's review was quickly grasped by North American limnologists concerned about the rapid eutrophication of the Laurentian Great Lakes. In particular, J. R. Vallentyne, the newly appointed head of the Fisheries Research Board of Canada's Eutrophication Section at the Freshwater Institute in Winnipeg, Manitoba, recruited Vollenweider to head a new group devoted to the study of Great Lakes eutrophication at the Canada Center for Inland Waters in Burlington, Ontario, and used[6] to convince the International Joint Commission (IJC) of the need to control phosphorus to decrease eutrophication.

Vollenweider's report was written for the Organization for Economic Cooperation and Development (OECD). In the late 1970s and early 1980s, he headed a global effort to develop broadly applicable eutrophication models.[7] However, his original report was the one that guided most of the world's early attempts to decrease eutrophication, under the philosophy that it was important to get started. Curiously, although it became familiar to most limnologists of the day, the review[6] was never published in the peer-reviewed literature. Vollenweider later received the prestigious Tyler Prize for Environmental Achievement for his contribution to reducing eutrophication.

■ WHOLE LAKE EXPERIMENTS

W.E. Johnson was the first director of the newly formed Freshwater Institute in Winnipeg, Manitoba, which included Vallentyne's Eutrophication Section. He believed that only large-scale experiments in whole lakes would convince policy makers to pass expensive legislation to control eutrophication. Johnson convinced the Fisheries Research Board of Canada to sponsor an area where whole-lake experiments with nutrients could be undertaken to guide policy alternatives. This approach was assigned to Vallentyne's Eutrophication Section. An area east of Kenora, Ontario and south of the Trans-Canada Highway was selected, and control of over 50 small lakes and their watersheds was negotiated with the Ontario government and local logging companies. A road to the Experimental Lakes Area (hereafter ELA) and a year-round campsite complete with scientific facilities and accommodations for about 30 scientific staff were installed during the winter of 1968–69.[8]

Before complete results of ELA experiments were reported, the IJC had already in 1971 recommended control of phosphorus inputs as the most likely way to decrease eutrophication in the Great Lakes.[9] They reasoned that the proportion of the nutrient inputs to Lakes Erie and Ontario supplied by humans was higher for phosphorus than it was for nitrogen, and also that there was proven technology for removing phosphorus inexpensively from wastewater treatment facilities (WWTFs), whereas there were still questions about the feasibility and cost of removing nitrogen.

The IJC also recommended that the phosphorus content of household detergents, which were then about 50% of the anthropogenic phosphorus input to the lower Great Lakes from Canada and 70% of inputs from the USA,[9] should be reduced as much as possible. The detergent industry, fearful that its effective and lucrative cleaning products would be banned, publicized a number of papers that disagreed with the IJC's recommendation to control phosphorus, making the case that carbon was the element causing the problem. These papers were summarized in an issue of *Canadian Research and Development* featuring a hangman's noose on the cover and titled *"We Hung Phosphorus Without a Fair Trial.*[10] The widely publicized claim that carbon control was necessary caused a delay in eutrophication control policy, especially in the U.S., although both governments agreed to reduce phosphorus inputs in the Great Lakes Water Quality Agreement of 1972 http://www.ijc.org/en/activitiesX/consultations/glwqa/guide_3.php#1972. The subject of carbon limitation is discussed at some length in several papers in a 1972 symposium on eutrophication.[11]

ELA's first key experiment, in Lake 227, a lake with extremely low concentrations of dissolved inorganic carbon, showed that increasing inputs of phosphorus and nitrogen would cause algal blooms no matter how limiting carbon was in small-scale experiments.[12] Although bottle and mesocosm bioassays in the lake indicated carbon limitation as publsihed[10] in the lake enough carbon invaded from the atmosphere to maintain algal blooms in proportion to phosphorus inputs. The Lake 227 result disproved the detergent industry's claim that carbon control (which was impossible in large lakes open to the atmosphere) was needed to curb eutrophication, and called into question the small-scale assays that are commonly used to make decisions about nutrient control.

In a second experiment at ELA, Lake 226 was split into two basins with a heavy sea curtain. Nitrogen plus carbon were added to both basins, but phosphorus only to the north basin. Algal blooms formed quickly in the basin receiving phosphorus, but not in the basin receiving only nitrogen and carbon.[13] The results of these experiments further encouraged phosphorus control policies in many jurisdictions. A comprehensive survey of ELA lakes using nutrient enrichment bioassays[14] and similar assays plus mesocosm experiments in Lake 227[15,16] showed that there were a variety of enrichment responses to C, N, P, and Fe in short-term assays, but whole lake experiments showed that these were largely irrelevant to whole-ecosystem responses. The primacy of phosphorus as ultimately limiting the eutrophication of ELA lakes could only be demonstrated by whole-lake experiments. It is unfortunate that this shortcoming of nutrient enrichment bioassays has not been understood by the broader limnological community and regulatory agencies, which still accept small-scale nutrient addition bioassays as reliably informing ecosystems scale questions of nutrient management (Box 1).

■ THE LAKE WASHINGTON CASE HISTORY

The third important influence on early policy decisions to regulate phosphorus was a long-term case history of Lake

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Box 1. Eutrophication Control Cannot Be Reliably Predicted from Short, Small Scale Nutrient Addition Bioassays

Many have claimed that because nutrient addition bioassays or nutrient ratios show that waters are nitrogen limited, nitrogen inputs must be reduced in order to control eutrophication.[51,52] However, this is the very sort of evidence that would have led to erroneous conclusions in the ELA lakes, and we suggest that such assays no longer be used to guide eutrophication management in whole lakes. As shown below, there are now many well-documented case histories in lakes of a large range of sizes and geological settings to show that reducing phosphorus input will reduce eutrophication, although delays in response due to hysteresis caused by internal loading are common.

It therefore seems reasonable to predict that small scale nutrient addition bioassays will also be unreliable indices for eutrophication management in coastal waters. In coastal waters where Cyanobacteria produce nuisance blooms, especially nitrogen fixers or species specializing to thrive at low N:P ratios, phosphorus control is likely to be as effective as it is in freshwater. In the single case where a long-term, whole ecosystem study was conducted in an estuary, reducing phosphorus input effectively reduced the biomass of phytoplankton in the Stockholm Archipelago.[53] There are no similar case histories where nitrogen has been decreased, and despite decades of study, we still do not know enough about coastal ecosystems to ascribe a nutrient control regime with confidence. Before expensive ecosystem-scale nutrient control measures are widely applied, the prescribed management should be verified in a long-term, whole estuary, as has been done many times for phosphorus in lakes.

Washington by W.T. Edmondson.[17] He documented the increased eutrophication as the growing city of Seattle discharged more and more sewage into the lake. He then persuaded the city to divert sewage from the lake into nearby Puget Sound, after which he documented the lake's recovery. Edmondson deduced that the recovery was largely the result of declining phosphorus inputs, because the size of algal blooms declined in direct proportion to declines in phosphorus, while nitrogen accumulated in the lake as nitrate.

Based in these three lines of evidence, reducing phosphorus inputs became the basis for eutrophication policies in the U.S., Canada, and many European countries. While there were several early success stories after phosphorus inputs were decreased,[18,19] little attention was subsequently paid to the overall success of phosphorus management, especially in North America.

■ RENEWED CALLS FOR CONTROLLING NITROGEN INPUTS

In the early years of the new millennium, there were again demands that nitrogen inputs be controlled, fueled by several reports that decreasing phosphorus inputs alone was not reducing eutrophication, and evidence from bottle-scale experiments that nitrogen was often the element limiting production in estuaries and some lakes. A review[20], which included an analysis of early results from four lakes in the ELA, concluded that reducing nitrogen inputs was also necessary to control eutrophication. Despite several flaws in their analysis[21], the "erosion" review[20] became a very popular citation for authors who favored nitrogen or dual nutrient control.

The proponents of nitrogen or dual nutrient control were very persuasive with policy makers. The European Union's Water Framework Directive[22] stipulates removal of nitrogen, suggesting that it will reduce the eutrophication of lakes. The United States Environmental Protection Agency has committed to partnering with states to accelerate the reduction of both nitrogen and phosphorus input to waters, and has recently adopted dual nutrient criteria for lakes, rivers and estuaries.[23] In New Zealand, the controversy is whether to control both nitrogen and phosphorus or only nitrogen.[24] Other countries are expected to soon follow suit.

As several authors have pointed out,[25,26] controlling nitrogen rather than or in addition to phosphorus at point sources is much more costly and technically more difficult than removing only phosphorus. It therefore seems logical to carefully review the evidence for the effectiveness of single and dual nutrient controls before proceeding with more costly eutrophication control policies. This choice, P only or N plus P, and the relative costs will become even more important in the future as burgeoning population growth and urbanization in the developing world increases culturally driven enrichment of receiving waters.

■ EVIDENCE FOR THE SUCCESS OF PHOSPHORUS CONTROL

Evidence that reducing inputs of phosphorus is effective in reducing eutrophication comes from four methods, all long-term studies at ecosystem scales: 1. Long-term case histories, 2. multiyear whole lake experiments, 3. experiments where chemical treatments are used to remove phosphorus from the water column, and 4. chemical additions to inhibit return of phosphorus from the sediments to the water column. Whether or not small-scale nutrient enrichment experiments show nitrogen limitation does not appear to matter, because such methods can only measure short-term (*proximate*) nutrient limitation, whereas controlling eutrophication requires reducing inputs of nutrients that provide long-term (*ultimate*) control.[27]

Case Histories and Whole Lake Experiments Where Phosphorus Input Is Reduced. Reference[21] reviewed both whole-lake experiments and case-histories which had documented the response of lakes to reductions in phosphorus alone. Since that review, several other studies have added to the list of lakes that have been successfully recovered by decreasing phosphorus inputs (Table 1).

Dove and Chapra[28] showed that the Laurentian Great Lakes recovered well following measures agreed to under the 1972 and 1978 water quality agreements, which required control of phosphorus inputs from large WWTPs and in household detergents in the U.S. and Canada. Phosphorus concentrations and algal biomass in all lakes declined to levels equal to or below target concentrations set as the result of the 1978 measures. This is not surprising, because N:P ratios observed in the lakes have remained well above Redfield Ratios (Figure 1).

The remaining eutrophication problem, in Lake Erie, has been shown to be the result of lack of control of nonpoint sources of phosphorus, largely due to increasing intensification of agriculture and other land-use changes, as well as high runoff.[29,30] Increased bioavailability of the incoming phosphorus has also been noted, and is also believed to be caused by intensified agriculture[31] as has been noted in other lakes.[32] Prolonged hypoxia in the central basin of the lake, which promotes internal phosphorus recycling, is also a factor.[33] Currently, the IJC is reviewing its P loading objectives particularly for

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Table 1. Examples of Freshwaters in Nine Countries Where Eutrophication Decreased Following the Control of Phosphorus Inputs[a]

name of water body	country	location
Sacramento River	U.S.	40°N, 121°W
Lake Erie	Canada, U.S.	42°N, 81°W
Lake Onondaga	U.S.	43°N, 76°W
Lake Ontario	Canada, U.S.	44°N, 78°W
Lake Huron	Canada, U.S.	44°N, 82°W
Lake Michigan	U.S.	44°N, 87°W
Little Otter Lake	Canada	45°N, 80°W
Gravenhurst Bay	Canada	45°N, 80°W
Lake Geneva	Switzerland, France	46°N, 6°E
Lago Maggiore	Italy	46°N, 9°E
Moses Lake	USA	47°N, 119°W
Lake Balaton	Hungary	47°N, 18°E
Murtensee	Switzerland	47°N, 7°E
Lake Lucerne	Switzerland	47°N, 8°E
Turlersee	Switzerland	47°N, 8°E
Hallwilersee	Switzerland	47°N, 8°E
Sempachersee	Switzerland	47°N, 8°E
Lake Superior	Canada, U.S.	47°N, 88°W
Lake Zürich	Switzerland	47°N, 9°E
Pfäffikersee	Switzerland	47°N, 9°E
Lake Washington	U.S.	48°N, 122°W
Lake Vättern	Sweden	48°N, 15°E
Lake Constance	Switzerland, Austria, Germany	48°N, 9°E
Rhine River	Europe	49°N, 7°E
Kootenay Lake	Canada	50°N, 117°W
ELA Lakes 226 NE, 303, 304, 261	Canada	50°N, 94°W
Schlachtensee	Germany	52° N, 13°E
Lake Tegel	Germany	53°N, 13°E
Lake Fure	Denmark	56°N, 12°E
Lake Vänern	Sweden	59°N, 13°E
Lake Hjälmaren	Sweden	59°N, 16°E
Lake Mälaren	Sweden	59°N, 17°E
Lake Norrviken	Sweden	59°N, 18°E
Lake Mjøsa	Norway	61°N, 11°E
Meretta Lake	Canada	72°N, 95°W

[a]Recovered lakes range in size from a few hectares to 83 300 km^2 and in depth from 2 to 468 m. Latitudes and longitudes are given, to the nearest degree, to illustrate the wide geographic coverage of case histories. Lakes recovered by using chemicals to precipitate phosphorus are not included. Expanded from ref 21.

Lake Erie, but there is little evidence, again from short-term nutrient enrichment bioassays, to support recent claims that nitrogen control is needed to reduce algal blooms in the Great Lakes, as claimed by some.[34]

Fastner et al.[35] reviewed the case histories of eight lakes in Europe and the U.S., which had been subjected to long-term reductions in phosphorus inputs. In all cases, phosphorus reduction was highly successful, although the response times varied from 5 to 30 years. They noted that algal blooms did not respond until total phosphorus (TP) decreased below some threshold value, usually around 50 μg/L, as would be expected when light, rather than nutrients, limits algal biomass or production. Similarly,[36] found that phosphorus reductions were very effective in reducing algal blooms and the prevalence of Cyanobacteria in European lakes, but not until phosphorus concentrations decreased to <100 μg/L. Müller et al.[37] studied the trajectories of four peri-alpine Swiss lakes that had been closely monitored for 25 years after phosphorus input was reduced. They found that in the latter stages of recovery, the efficiency of sedimentation of phosphorus actually increased, so that recovery accelerated over time. All of these studies suggest a threshold must be reached before phosphorus reductions are effective in controlling eutrophication.

Chemical Precipitation of Phosphorus. In several studies, phosphorus concentrations in lakes were reduced by adding iron, alum, or other compounds to sequester phosphorus in sediments. For example, application of a modified bentonite clay (Phoslock) reduced phosphorus concentrations in Lake Rauwbraken, Netherlands, by 92%, causing this once-hypereutrophic lake to become oligo-mesotrophic.[38] Similarly, Lake Gross-Glienecker, Germany, was treated with iron, combined with oxygenating the hypolimnion using bubblers. Total phosphorus declined rapidly from 500 to 30−40 μg/L, and algal chlorophyll was reduced from an average of 49 to 6.5 μg/L.[39] Such treatments inhibit phosphorus recycling from sediments that otherwise can delay recovery from eutrophication. While chemical treatments are too expensive for large lakes, these examples clearly add weight to the case histories and experiments in whole lakes to demonstrate that controlling phosphorus is the key to reducing eutrophication in freshwaters.

Additions of Nitrate to Maintain High Redox at the Mud−Water Interface. In some lakes, phosphorus concentrations have been reduced by adding nitrate to maintain high redox conditions at the sediment-water interface, preventing mobilization of phosphorus recycled from sediments (internal loading). Such methods have been employed successfully to reduce phosphorus concentrations and algal biomass at several locations in the USA and Europe.[21] The technique has the added benefits of reducing the fluxes of other chemicals strongly influenced by redox conditions, such as methyl mercury and arsenic.[40,41]

In summary, several separate lines at the scale of whole lake investigation show clearly that reduction in phosphorus is very effective at controlling eutrophication of freshwater lakes, over a wide range of lake sizes, geological settings, and latitudes.

EVIDENCE THAT REDUCING INPUT OF NITROGEN DOES NOT REDUCE EUTROPHICATION

There are a number of eutrophic lakes where inputs of nitrogen as well as phosphorus were decreased, including both case histories and whole-lake experiments. Edmondson and Lehman[42] and Welch[43] both deduced that phosphorus was the element to control because after nutrient inputs were reduced, chlorophyll decreased in proportion to phosphorus decline while nitrogen accumulated as nitrate. Similar observations were made in experimental lakes after nutrient loading was terminated.[44] Similar increases in nitrate concentrations during declines in phosphorus and phytoplankton have been observed in the Great Lakes.[28,45] It appears that excess nitrate is denitrified rather rapidly in shallow lakes[21,41] but not in deep ones,[45] although concentrations in the latter have remained well below levels of concern for human health.

None of the above cases provides evidence that dual nutrient control reduced eutrophication of lakes either more effectively or rapidly than controlling phosphorus alone. Welch[43] concluded from his study of the recovery of Moses Lake, Washington: "*Targeting both N and P may not only be much costlier than necessary, but may even promote blooms of N-fixing cyanobacteria, especially in cases of high internal P loading.*"

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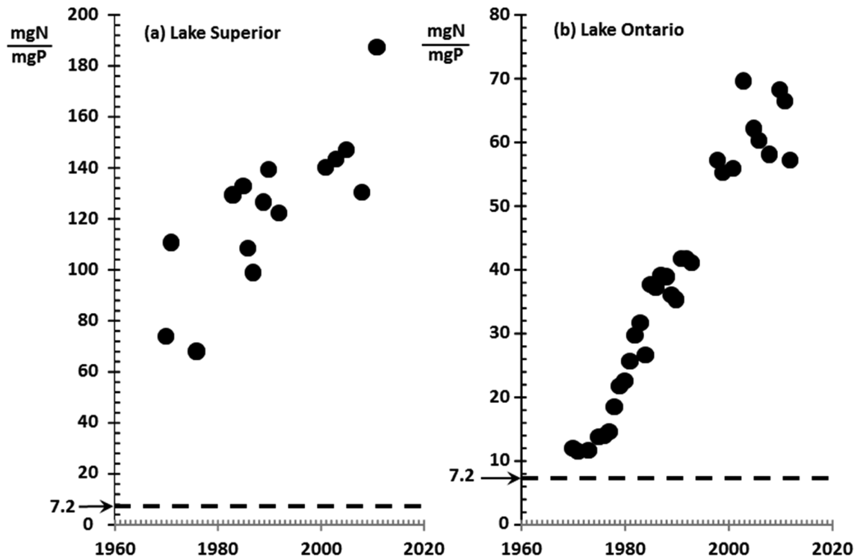

Figure 1. Trends of open lake, spring ratios of median nitrogen-to-phosphorus ratio (as nitrate/total phosphorus) for lakes (a) Superior and (b) Ontario over the past four decades. The Redfield nitrogen-to-phosphorus ratio (horizontal dashed line) is superimposed as an approximate estimate of the level above which the lake would tend to be phosphorus limited. Phosphorus controls were instituted in the Great Lakes beginning about 1973.

In only one case has nitrogen input been decreased without reducing phosphorus: Lake 227 at the ELA. The Lake 227 experiment has now been carried out for 47 years, with two modifications to the N:P ratio in loading to the lake. In 1975, the N:P ratio was decreased from 14:1 to 5:1 by weight. This caused a change from an algal community containing many of the original taxa in the lake to one dominated by nitrogen-fixing Cyanobacteria.[46] While there are other reasons such as selective zooplankton grazing that can cause similar shifts to nitrogen fixing Cyanobacteria, decreased N:P ratios are the most common cause.[21]

In a second change, nitrogen was eliminated entirely from fertilizer added to Lake 227, beginning in 1990. Since then the lake has been fertilized only with phosphorus. Nitrogen fixation increased as Cyanobacteria became increasingly dominant, compensating for the loss of fertilizer input, and the elimination of nitrogen fertilizer has not caused a long-term reduction in eutrophication in the lake.[47] This conclusion was criticized by,[48] who pointed to a slight decline in total nitrogen and phytoplankton biomass for several years following elimination of nitrogen fertilizer. They predicted that declining nitrogen would eventually cause long-term declines in phytoplankton, but that has not happened.[49] In a thorough study of the nitrogen sources and sinks in the lake in 2011, nitrogen fixation in 2011 exceeded the sum of fertilizer plus nitrogen fixation in any year before 1990, and the net income of nitrogen to the lake was positive https://www.youtube.com/watch?v=i1KUSzBRt7U&feature=youtu.be

Nitrogen fixation and phytoplankton biomass have increased over time, and the lake is correcting its nitrogen imbalance. Once nitrogen fixers appear in summer, the phytoplankton community becomes phosphorus limited.[50] Altogether, the long-term record for Lake 227 shows that even the total elimination of anthropogenic nitrogen input has no significant impact on decreasing eutrophication.

To be useful, any assays of nutrient limitation must be able to account for long-term adjustments in a lake's internal biogeochemical cycling.[21] Clearly, bottle bioassays and short-term experiments in small mesocosms are of little value in predicting the long-term results of changes to nutrient inputs. If decreasing eutrophication of freshwaters is the objective, policies must focus on reducing inputs of phosphorus. Although there are many reasons why large regional and global sources of nitrogen should be controlled (Box 2), there is simply no ecosystem-scale evidence that removing nitrogen is effective in reducing algal biomass.

Box 2. Control Nitrogen Where It Is Needed

We agree with most ecologists that increasing anthropogenic emissions of nitrogen must be controlled. Emissions of nitrogen play a role in greenhouse warming,[54] formation of smog and peroxyacetyl nitrate,[55] soil and water acidification,[56] and biodiversity loss.[57] Also, increases in nitrate to concentrations approaching human health guidelines in groundwater near feedlots and intensively fertilized crops clearly justify reductions to nitrogen inputs, primarily from nonpoint sources.

However, our review of the evidence suggests that removing nitrogen at WWTFs is not an effective way to address the harmful ecological impacts of nitrogen. Waste streams to lakes account for only a few percent of the estimated 221 Tg/y of nitrogen emitted by global human activities.[58] Instead, much larger sources of nitrogen that have been demonstrated to cause ecological damage should be the targets of choice for reducing emissions. Targeting fossil fuels (25 Tg/y), industrial nitrogen fixation (136 Tg/y), or human-induced biological fixation (60 Tg/y) is necessary to protect the ecosystems that are being damaged by excessive inputs of nitrogen.

■ ADDING NITROGEN

In contrast to the lack of evidence for any effect of reducing nitrogen inputs, there are several cases where *adding* nitrogen has actually reduced symptoms of eutrophication, either by increasing the N:P ratio to values that allow more desirable species to outcompete cyanobacteria, or by acting as an electron acceptor that inhibits the release of phosphorus release from sediments, as cited above. Schindler[21] reviewed several studies

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where nitrate was deliberately added to lakes in order to maintain noncyanophyte species from becoming dominant. In Lake Onondaga, where nitrogen inputs have been kept deliberately high, the formation of cyanobacteria blooms has been suppressed, phosphorus release from sediments has been reduced, and formation of methyl mercury has decreased.[40]

■ CONCLUSIONS

Wastes from a growing human population and increasing agricultural production are likely to increase P inputs and eutrophication in coming decades. Decisions about mitigating eutrophication will be made in the context of multiple environmental threats due to changing climate, land use and other factors. These decisions should use the best scientific information relevant to the scale of the problems. There is no evidence that eutrophication can be managed by controlling N inputs. In contrast, multiple lines of evidence at the whole-lake scale of management show that P control works to mitigate eutrophication.

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Notes

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■ REFERENCES

(1) Schindler, D. W. The cumulative effects of climate warming and other human stresses on Canadian freshwaters in the new millennium. *Can. J. Fish. Aquat. Sci.* **2001**, *58*, 18−29.

(2) Schindler, D. W.; Vallentyne, J. R. *The Algal Bowl: Overfertilization of the World's Freshwaters and Estuaries*; University of Alberta Press: Edmonton, Canada, 2008.

(3) Hutchinson, G. E.; Bonatti, E.; Cowgill, U. M.; Goulden, C. E.; Leventhal, E. A.; Mallett, M. E. Iamula: an account of the history and development of the Lago di Monterosi, Latium, Italy. *Trans. Am. Philos. Soc.* **1970**, *60*, 1−178.

(4) Douglas, M. S. V; Smol, J. P.; Savelle, J. M.; Blais, J. M. Prehistoric Inuit whalers affected Arctic freshwater ecosystems. *Proc. Natl. Acad. Sci. U. S. A.* **2004**, *101*, 1613−1617.

(5) National Academy of Sciences USA. *Eutrophication: Causes, Consequences, Correctives*; National Academy Press: Washington, DC, USA, 1969.

(6) Vollenweider, R. A. *Scientific Fundamentals of the Eutrophication of Lakes and Flowing Waters, With Particular Reference to Nitrogen and Phosphorus As Factors in Eutrophication*; OECD Tech. Rep. DAS/CS/68.27, Paris, France, 1968.

(7) Vollenweider, R. A.; Kerekes, J. J. Background and Summary Results of the OECD Cooperative Program on Eutrophication. In *Restoration of lakes and inland waters. International Symposium on Inland Waters and Lake Restoration*, EPA440/5−81-11.; Portland, Maine, **1981**; pp 25−36.

(8) Schindler, D. W. A personal history of the Experimental Lakes Project. *Can. J. Fish. Aquat. Sci.* **2009**, *1847*, 1837−1847.

(9) International Joint Commission. *Pollution of Lake Erie, Lake Ontario and the International Section of the St. Lawrence River.*; Ottawa, Canada, 1971.

(10) Canadian Research and Development. *We Hung Phosphates without a Fair Trial*; Maclean Hunter Publishers, 1970.

(11) Likens, G. E., Ed. *Nutrients and eutrophication*. Amer. Soc. Limnol. Oceanogr Spec. Symp. 1. *328* p. 1972.

(12) Schindler, D. W.; Brunskill, G. J.; Emerson, S.; Broecker, W. S.; Peng, T.-H. Atmospheric carbon dioxide: its role in maintaining phytoplankton standing crops. *Science* **1972**, *177*, 1192−1194.

(13) Schindler, D. W. Eutrophication and recovery in experimental lakes: implications for lake management. *Science* **1974**, *184*, 897−899.

(14) Sakamoto, M. Chemical factors involved in the control of phytoplankton production in the Experimental Lakes Area, northwestern Ontario. *J. Fish. Res. Board Can.* **1971**, *28*, 1763−1782.

(15) Schindler, D. W.; Armstrong, F. A. J.; Holmgren, S. K.; Brunskill, G. J. Eutrophication of Lake 227, Experimental Lakes Area, northwestern Ontario, by addition of phosphate and nitrate. *J. Fish. Res. Board Can.* **1971**, *28*, 1763−1782.

(16) Schindler, D. W. Carbon, nitrogen, and phosphorus and eutrophication of freshwater lakes. *J. Phycol.* **1971**, *7*, 321−329.

(17) Edmondson, W. T. Phosphorus, nitrogen, and algae in Lake Washington after diversion of sewage. *Science* **1970**, *169*, 690−691.

(18) Michalski, M. F. P.; Conroy, N. The oligotrophication of Little Otter Lake, Parry Sound District. In *Proc. 16th Conf. Great Lakes Res.*; 1973; pp 934−948.

(19) Ahlgren, I. Role of sediments in the process of recovery of a eutrophicated lake. In *Interactions between Sediments and Freshwater*; Golterman, H. L., Ed.; Pudoc: Wageningen, The Netherlands, 2008; pp 371−377.

(20) Lewis, W. M., Jr.; Wurtsbaugh, W. A. Control of lacustrine phytoplankton by nutrients: Erosion of the phosphorus paradigm. *Int. Rev. Hydrobiol.* **2008**, *93*, 446−465.

(21) Schindler, D. W. The dilemma of controlling cultural eutrophication of lakes. *Proc. R. Soc. London, Ser. B* **2012**, *279*, 4322−4333.

(22) European Union. Water Framework Directive https://circabc.europa.eu/sd/a/9060bdb4-8b66-439e-a9b0-469_a5cfd8db2217/Guidance_document_23_Eutrophication.pdf (accessed March 30, 2016).

(23) United States Environmental Protection Agency.. *Preventing eutrophication: Scientific support for dual nutrient criteria*. EPA-820-S-15-001. 6p. 2015.

(24) Abell, J. M.; Özkundakci, D.; Hamilton, D. P. Nitrogen and phosphorus limitation of phytoplankton growth in New Zealand lakes: Implications for eutrophication control. *Ecosystems* **2010**, *13*, 966−977.

(25) Håkanson, L.; Bryhn, A. C. Controlling Eutrophication in the Baltic Sea and the Kattegat. In *Eutrophication: Causes, Consequences and Control*; Ansari, A. A.; Gill, S. S.; Lanza, G. R.; Rast, W., Eds.; Springer: Dordrecht, Netherlands, 2010; pp 17−67.

(26) Szoke, N.; Yuan, Q.; Oleszkiewicz, J. A.; Celmer, D.; Schindler, D.; Stainton, M.; Paterson, M.; Lockhart, L. Environmentally sensible effluent nitrogen limits. In *Proceeding: 2nd 480 IWA Nutrient Management in Wastewater Treatment Process*; Krakow, Poland, **2009**; pp 1221−1224.

(27) Vitousek, P. M.; Porder, S. P.; Houlton, B. Z.; Chadwick, O. A. Terrestrial phosphorus limitation: mechanisms, implications, and nitrogen − phosphorus interactions. *Ecol. Appl.* **2010**, *20*, 5−15.

(28) Dove, A.; Chapra, S. C. Long-term trends of nutrients and trophic response variables for the Great Lakes. *Limnol. Oceanogr.* **2015**, *60*, 696−721.

(29) Michalak, A. M.; Anderson, E. J.; Beletsky, D.; Boland, S.; Bosch, N. S.; Bridgeman, T. B.; Chaffin, J. D.; Cho, K.; Confesor, R.; Daloglu, I.; et al. Record-setting algal bloom in Lake Erie caused by agricultural and meteorological trends consistent with expected future conditions. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110*, 6243−6244.

(30) Kane, D. D.; Conroy, J. D.; Peter Richards, R.; Baker, D. B.; Culver, D. a. Re-eutrophication of Lake Erie: Correlations between tributary nutrient loads and phytoplankton biomass. *J. Great Lakes Res.* **2014**, *40*, 496−501.

(31) Baker, D. B.; Confesor, R. B.; Ewing, D. E.; Johnson, L. T.; Kramer, J. W.; Merryfield, B. J. Phosphorus loading to Lake Erie from

DOI: 10.1021/acs.est.6b02204
Environ. Sci. Technol. 2016, 50, 8923−8929

the Maumee, Sandusky, and Cuyahoga Rivers: The importance of bioavailability. *J. Great Lakes Res.* **2014**, *40*, DOI: 50210.1016/j.jglr.2014.05.001.
(32) Prestigiacomo, A. R.; Effler, S. W.; Gelda, R. K.; Matthews, D. A.; Auer, M. T.; Downer, B. E.; Kuczynski, A.; Walter, M. T. Apportionment of Bioavailable Phosphorus Loads Entering Cayuga Lake, New York. *J. Am. Water Resour. Assoc.* **2016**, *52*, 31−47.
(33) Scavia, D.; Allan, J. D.; Arend, K. K.; Bartell, S.; Beletsky, D.; Bosch, N. S.; Brandt, S. B.; Briland, R. D.; Dalo, I.; Depinto, J. V.; et al. Assessing and addressing the re-eutrophication of Lake Erie: Central basin hypoxia. *J. Great Lakes Res.* **2014**, *40*, 226−246.
(34) Paerl, H. W.; Gardner, W. S.; McCarthy, M. J.; Peierls, B. L.; Wilhelm, S. W. Algal blooms: Noteworthy nitrogen. *Science* **2014**, *346*, 175.
(35) Fastner, J.; Abella, S.; Litt, A.; Morabito, G.; Matthews, D.; Phillips, M. G.; Chorus, I. Combating cyanobacterial proliferation by avoiding or treating inflows with high P load — experiences from eight case studies. *Aquat. Ecol.* **2015**, DOI: 10.1007/s10452-015-9558-8.
(36) Carvalho, L.; McDonald, C.; de Hoyos, C.; Mischke, U.; Phillips, G.; abor Borics, G.; Poikane, S.; Skjelbred, B.; Lyche Solheim, A.; Van Wichelen, J.; Cardoso, A. C. Sustaining recreational quality of European lakes: minimizing the health risks from algal blooms through phosphorus control. *J. Appl. Ecol.* **2013**, *50*, 315−323.
(37) Müller, B.; Gáchter, R.; Wüest, A. Accelerated water quality improvement during oligotrophication in peri-alpine lakes. *Environ. Sci. Technol.* **2014**, *48*, 6671−6677.
(38) Lürling, M.; Van Oosterhout, F. Controlling eutrophication by combined bloom precipitation and sediment phosphorus inactivation. *Water Res.* **2013**, *47*, 6527−6537.
(39) Wolter, K.-D. Restoration of eutrophic lakes by phosphorus precipitation, with a case study on Lake Gross-Glienicker. In *Restoration of Lakes, Streams, Floodplains, And Bogs in Europe: Principles and Case Studies*; Eiseltová, M., Ed.; Wetlands Ecology Conservation and Management; Springer: Dordrecht, Netherlands, 2010; Vol. 3, pp 85−99.
(40) Matthews, D. A.; Babcock, D. B.; Nolan, J. G.; Prestigiacomo, A. R.; Effler, S. W.; Driscoll, C. T.; Todorova, S. G.; Kuhr, K. M. Whole-lake nitrate addition for control of methylmercury in mercury-contaminated Onondaga Lake, NY. *Environ. Res.* **2013**, *125*, 52−60.
(41) Hemond, H. F.; Lin, K. Nitrate suppresses internal phosphorus loading in an eutrophic lake. *Water Res.* **2010**, *44*, 3645−3650.
(42) Edmondson, W. T.; Lehman, J. T. The effect of changes in the nutrient income on the condition of Lake Washington. *Limnol. Oceanogr.* **1981**, *26*, 1−29.
(43) Welch, E. B. Should nitrogen be reduced to manage eutrophication if it is growth limiting? Evidence from Moses Lake. *Lake Reservoir Manage.* **2009**, *25*, 401−409.
(44) Levine, S. N.; Schindler, D. W. Phosphorus, nitrogen, and carbon dynamics of experimental lake 303 during recovery from eutrophication. *Can. J. Fish. Aquat. Sci.* **1989**, *46*, 2−10.
(45) Finlay, J. C.; Small, G. E.; Sterner, R. W. Human influences on nitrogen removal in lakes. *Science* **2013**, *342*, 247−250.
(46) Schindler, D. W. Evolution of phosphorus limitation in lakes. *Science* **1977**, *195*, 260−262.
(47) Schindler, D. W.; Hecky, R. E.; Findlay, D. L.; Stainton, M. P.; Parker, B. R.; Paterson, M. J.; Beaty, K. G.; Lyng, M.; Kasian, S. E. M. Eutrophication of lakes cannot be controlled by reducing nitrogen input: Results of a 37-year whole-ecosystem experiment. *Proc. Natl. Acad. Sci. U. S. A.* **2008**, *105*, 11254−11258.
(48) Scott, J. T.; McCarthy, M. J. Nitrogen fixation may not balance the nitrogen pool in lakes over timescales relevant to eutrophication management. *Limnol. Oceanogr.* **2010**, *55*, 1265−1270.
(49) Paterson, M. J.; Schindler, D. W.; Hecky, R. E.; Findlay, D. L.; Rondeau, K. J. Comment: Lake 227 shows clearly that controlling inputs of nitrogen will not reduce or prevent eutrophication of lakes. *Limnol. Oceanogr.* **2011**, *56*, 1545−1547.
(50) Hendzel, L. L.; Hecky, R. E.; Dindlay, D. L. Recent changes of nitrogen fixation in Lake 227 in response to reduction of the N:P loading ratio. *Can. J. Fish. Aquat. Sci.* **1994**, *51*, 2247−2253.
(51) Paerl, H. W. Controlling eutrophication along the freshwater − marine continuum: Dual nutrient (N and P) reductions are essential. *Estuaries Coasts* **2009**, *32*, 593−601.
(52) Howarth, R. W.; Marino, R. Nitrogen as the limiting nutrient for eutrophication in coastal marine ecosystems: Evolving views over three decades. *Limnol. Oceanogr.* **2006**, *51*, 364−376.
(53) Brattberg, G. Decreased phosphorus loading changes phytoplankton composition and biomass in the Stockholm Archipelago. *Vatten* **1986**, *42*, 141−153.
(54) U.S. Environmental Protection Agency. Overview of greenhouse gases http://www3.epa.gov/climatechange/ghgemissions/gases/n2o.html (accessed February 17, 2016).
(55) U.S. Environmental Protection Agency. Photochemical smog - what it means for us www.epa.sa.gov.au/files/8238_info_photosmog.pdf (accessed February 17, 2016).
(56) Watmough, S. A.; Aherne, J.; Alewell, C.; Bailey, S.; Clair, T. O. M.; Dillon, P.; Duchesne, L.; Eimers, C.; Fernandez, I.; Foster, N. Sulphate, nitrogen and base cation budgets at 21 forested catchments in Canada, the United States and Europe. *Environ. Monit. Assess.* **2005**, *109*, 1−36.
(57) Dise, N. B.; Ashmore, M.; Belyazid, S.; Bleeker, A.; Bobbink, R.; De Vries, W. Nitrogen as a threat to European terrestrial biodiversity. In *The European Nitrogen Assessment. Sources, effects and policy perspective*; Cambridge University Press: Cambridge, United Kingdom, 2011.
(58) Schlesinger, W. H.; Bernhardt, E. S. *Biogeochemistry: An Analysis of Global Change*, 3rd ed.; Academic Press: Waltham, United States of America, 2013.

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Environ. Sci. Technol. 2016, 50, 8923−8929