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OVERVIEW

Nutrients, eutrophication and harmful algal blooms along the freshwater to marine continuum

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Abstract

Agricultural, urban and industrial activities have dramatically increased aquatic nitrogen and phosphorus pollution (eutrophication), threatening water quality and biotic integrity from headwater streams to coastal areas world-wide. Eutrophication creates multiple problems, including hypoxic "dead zones" that reduce fish and shellfish production; harmful algal blooms that create taste and odor problems and threaten the safety of drinking water and aquatic food supplies; stimulation of greenhouse gas releases; and degradation of cultural and social values of these waters. Conservative estimates of annual costs of eutrophication have indicated $1 billion losses for European coastal waters and $2.4 billion for lakes and streams in the United States. Scientists have debated whether phosphorus, nitrogen, or both need to be reduced to control eutrophication along the freshwater to marine continuum, but many management agencies worldwide are increasingly opting for dual control. The unidirectional flow of water and nutrients through streams, rivers, lakes, estuaries and ultimately coastal oceans adds additional complexity, as each of these ecosystems may be limited by different factors. Consequently, the reduction of just one nutrient upstream to control eutrophication can allow the export of other nutrients downstream where they may stimulate algal production. The technology exists for controlling eutrophication, but many challenges remain for understanding and managing this global environmental problem.

This article is categorized under:
Science of Water > Water Quality
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KEYWORDS

aquatic, coastal, dead zones, ecology, estuaries, eutrophication, harmful algal bloom, lakes, nitrogen, nutrients, phosphorus, rivers, streams

1 | INTRODUCTION

One of the facets of the increasing human population and its environmental impacts is an acceleration of the rates at which nitrogen (N) and phosphorus (P) enter the biosphere, and these rates are projected to further increase in coming decades (Glibert, 2017; Sutton et al., 2013). Nutrients are essential to maintaining aquatic food webs and can have positive effects in water bodies such as supporting fish and shellfish production (e.g., Figure 1; Bachmann et al., 1996). Aquaculture ponds are

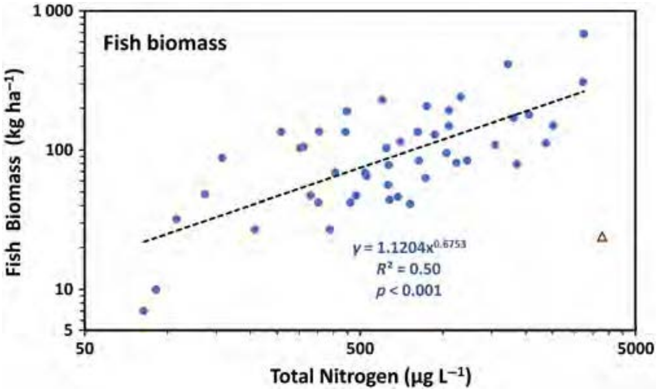


FIGURE 1 Relationship between total nitrogen in water and fish biomass in 48 Florida lakes (USA). One statistical outlier (triangle) was removed from the regression. Fish biomass was less closely correlated with total phosphorus ($r^2 = 0.34$, $p = .009$). Derived from data of Bachmann et al. (1996)

frequently fertilized to increase production of fish (Mischke, 2012) and unproductive (oligotrophic) lakes have been fertilized to increase the production of commercially valuable or endangered salmon (Budy, Luecke, & Wurtsbaugh, 1998; Stockner, Rydin, & Hyenstrand, 2000). However, negative effects of excessive human nutrient inputs (cultural eutrophication) frequently outweigh positive aspects, often leading to declining water quality along the freshwater to marine continuum.

Despite successes in North America and Europe in reducing eutrophication, many eutrophication-related problems remain both in freshwater rivers and lakes, as well as in coastal oceans, particularly in developing countries. In many cases, progress has been slow due to reliance on voluntary nutrient reduction strategies in lieu of legislation (Boesch, 2019), and scientific controversies on the role of N and P in controlling eutrophication (Lewis & Wurtsbaugh, 2008; Paerl et al., 2016; Schindler et al., 2008; Schindler, Carpenter, Chapra, Hecky, & Orihel, 2016). Here, we review the history of eutrophication and its impacts, and address five central issues regarding eutrophication in aquatic systems: (a) What are the impacts of eutrophication? (b) How do algal blooms and nutrient over-enrichment vary spatially as a result of different natural and anthropogenic biogeochemical drivers? (c) What are harmful algal blooms (HABs)?; (d) Do N and P interact to control eutrophication? (e) How can we most effectively manage nutrient over-enrichment along the freshwater to marine continuum?

2 | HISTORY OF EUTROPHICATION

The word "eutrophication" derives from the Greek *eutrophos*, or "well-nourished." European limnologists, August Thienemann (1918) and Einar Naumann (1919), first applied the term to lakes, using it to indicate increasing concentrations of phytoplankton in the water, contrasting with "oligotrophic," or poorly nourished systems. Anthropogenic increases in nutrients are technically "cultural eutrophication" to distinguish from natural causes, but here we use the term eutrophication to refer to human-caused enrichment. The first accounts were from Lake Zürich, Switzerland in the late 1800s, when scientists detected the proliferation of new species of cyanobacteria, and subsequently the loss of oxygen in bottom waters. This caused the demise of trout and whitefish and the rise of perch and minnows that were more tolerant of low oxygen conditions (Vallentyne, 1974). In the 1950s and 1960s, nutrients were diverted from the lake and by the 1970s Lake Zurich began recovering.

Like Europe, eutrophication problems in North America grew during the 20th century with population growth and intensification of agriculture. By the 1960s, limnologists understood the warning signs and Dr. Thomas Edmondson realized that sewage from the city of Seattle was causing cyanobacterial blooms in metropolitan Lake Washington. In response to his public warnings in the 1960s, wastewaters were diverted from the lake into nearby Puget Sound and algal blooms decreased and water clarity increased markedly in the lake (Edmondson, 1991). This diversion has likely exacerbated eutrophication and unwanted algal blooms in the Sound (Anderson et al., 2008), a frequent problem when nutrients are passed through linked water bodies across the freshwater to marine continuum (Paerl, Otten, & Kudela, 2018). Another story of degradation and partial recovery is Lake Erie, the 11th largest lake in the world. During the 20th century the population in Erie's watershed grew to over 12 million residents and their sewage was discharged into the relative shallow lake waters. Algae bloomed, dissolved oxygen was depleted by the decay of the algae in deeper water, and fish die-offs were common. Control of P inputs to the lake due to the banning of phosphates in detergents and advanced wastewater treatment decreased algal blooms and improved fish stocks by the 1990s (Schindler & Vallentyne, 2008). However, since 2010 there has been a resurgence of toxic cyanobacterial blooms, plaguing the residents around the lake (Ho & Michalak, 2015).

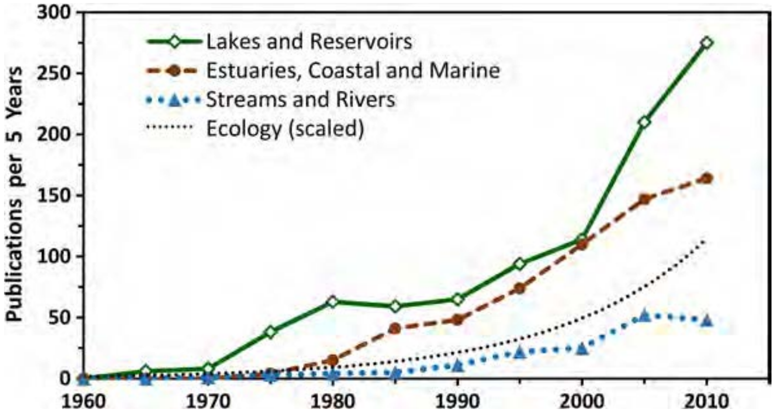


FIGURE 2 Publications referenced in web of science™ for five-year periods beginning in the indicated year. Search terms used in combination with "eutrophication" were: Lakes—"pond*, lake*, reservoir* or lentic"; estuaries and coastal—"marine, coastal, estuar*, ocean, sea"; streams and Rivers—"creek*, stream*, river*, lotic". The black dotted line shows the scaled increase since 1960 in publications found using the search term "ecolog*"

Although pollution studies and controls were implemented in lakes in the last millennium, the study of eutrophication in streams and coastal waters has lagged. Publications on freshwater lakes began accelerating in the 1960s (Figure 2). Estuarine and coastal waters were once thought to be minimally impacted by eutrophication because of their greater size and tidal flushing. However, these nearshore eutrophication problems finally gained recognition in the 1980s (e.g., Boynton, Kemp, & Keefe, 1982; D'Elia, Sanders, & Boynton, 1986; Jaworski, 1981). Publications about stream and river eutrophication only started to accelerate in the 1990s (e.g., Dodds, Jones, & Welch, 1998; Welch, Quinn, & Hickey, 1992) and publications on this topic are increasing slower than the general rate for ecological research.

3 | EUTROPHICATION IMPACTS IN LAKES, COASTAL ZONES, AND RIVERS

Lakes—In lakes, initial research and management targeted point-source reductions, largely by limiting use of phosphate-rich detergents and subsequently chemical removal in wastewater treatment plants. This early work focused on reducing taste and odor problems in drinking water and minimizing loss of dissolved oxygen (hypoxia) as sinking algae decomposed in the deeper strata (hypolimnia) of lakes (Lewis, Wurtsbaugh, & Paerl, 2011 and citations therein). Hypoxia decreases redox potentials in the bottom waters leading to a positive feedback that returns N and P to the lake water from the sediments (Dodds & Whiles, 2010; Mortimer, 1942).

Even without hypoxia, excess nutrients can cause a state change in lake, river, and coastal ecosystems, with a shift in the food web from one dominated by benthic microalgal and macrophyte production, to a phytoplankton-dominated system (Moss, Jeppesen, Sondergaard, Lauridsen, & Liu, 2013; Vadeboncoeur, Vander Zanden, & Lodge, 2002). Since fishes are often highly dependent on benthic invertebrates, these state changes may reduce the efficiency of fish production (Vander Zanden & Vadeboncoeur, 2002). More recently, concerns about lake eutrophication have broadened to include reductions in biodiversity of both autotrophs and heterotrophs (Azevedo et al., 2013), increased production of the greenhouse gases, nitrous oxide and methane (Beaulieu, DelSontro, & Downing, 2019), and the proliferation of HABs (EPA, 2009b, 2017; Paerl et al., 2018).

Estuarine and coastal systems—Eutrophication of estuarine and coastal (jointly termed coastal) ecosystems is particularly important because 45% of the human population is concentrated in their watersheds, thus increasing nutrient loading due to unprecedented agricultural, urban and industrial growth worldwide (Kay & Alder, 2005). Just as in lakes, when nutrient loading and hydrodynamic features combine to produce large phytoplankton blooms that sink, decompose, and deplete oxygen in bottom waters, shellfish and finfish resources are often damaged (Davidson et al., 2014; Rabalais, Turner, & Wiseman, 2002). These "dead zones" impact coastal zones over wide areas (Breitburg et al., 2018; Diaz & Rosenberg, 2008). For example, nutrient and organic matter discharge from the Mississippi River often produces a hypoxic dead zone exceeding 15,000 km^2 in the Gulf of Mexico (Scavia et al., 2017). Increasing phytoplankton production can also reduce light penetration and limit the extent of benthic sea grass habitat, a critical food resource and refuge for a variety of organisms. Eutrophication may also contribute to decline of coral reefs (Kroon, Thorburn, Schaffelke, & Whitten, 2016). Excessive P and N loading can stimulate nuisance dinoflagellate "red tide" and cyanobacterial blooms; these can produce toxic substances that bioaccumulate in shellfish and finfish, posing significant human health concerns. However, low-nutrient toxic blooms also occur, so the relationship between N, P and red tides is not simple (Davidson et al., 2012; Glibert, 2017).

Streams and rivers—Eutrophication impacts in streams and rivers have received far less attention than those in coastal areas and lakes, even though the U.S. Environmental Protection Agency (EPA, 2016) indicates that 41% and 46% of streams or rivers surveyed in 2008–2009 are impaired because of excessive N and P inputs. However, well-defined standards to protect these ecosystems are only beginning to be developed. Dodds (2006) summarized the negative effects of eutrophication in lotic systems (streams and rivers). Unlike lakes, the turbulence in streams usually minimizes anoxia, except in cases of extreme eutrophication or external organic matter loading. Nevertheless, high nutrient levels cause considerable problems in these systems. High concentrations of NH_3 in water are toxic to aquatic animals, especially at higher pHs (EPA, 2013a) and NO_3 above 10 mg/L can cause methemoglobinemia in infants (Fewtrell, 2004). Recent evidence indicates that NO_3 is also an endocrine disruptor in humans (Poulsen, Cedergreen, Hayes, & Hansen, 2018). Stream studies also indicate eutrophication-induced shifts in ecosystem rates and invertebrate community structure (Dodds & Smith, 2016).

Eutrophication in rivers increases with nutrient loading. In a study of U.S. streams mean benthic chlorophyll concentrations in U.S. streams increased significantly ($r^2 = 0.40$) from 20 to 180 mg m^{-2} as TN increased from 500 to 3,000 μg L^{-1} (Dodds, 2006; Dodds & Smith, 2016). The relationship between nutrients and suspended phytoplankton in rivers is even stronger (Figure 3; $r^2 = 0.67$; Van Nieuwenhuyse & Jones, 1996). Suspended phytoplankton reduce light penetration and the growth of benthic periphyton and aquatic plants in deep rivers. Eutrophication can also increase growth of epiphytes, thus decreasing the abundance of the macrophyte hosts (O'Hare et al., 2018). Esthetic impairment due to excessive filamentous algae in streams and rivers is increasingly recognized as one of the major results of eutrophication (Figure 4; Dodds & Welch, 2000; Jakus, Nelson, & Ostermiller, 2017; Suplee, Watson, Teply, & McKee, 2009).

Eutrophication in slow-moving rivers can also cause toxic algal blooms such as occurred in a 600-km long segment of the stagnant Murray-Darling River system (Australia), leading to livestock deaths and concerns about impacts on human health (Paerl & Otten, 2013a). Toxic benthic algae in streams can be common and may deliver these toxins to downstream coastal environments (Fetscher et al., 2015). Secondary metabolites of cyanobacteria (e.g., geosmins) in lakes and streams can cause taste and odor problems for drinking water supplies (Antonopoulou, Evgenidou, Lambropoulou, & Konstantinou, 2014) and is a common response to eutrophication.

Stream ecologists have recently emphasized that increased nutrient loading not only increases the production of autotrophs, but also increases the heterotrophic decomposition rate of terrestrial organic matter deposited in streams. Nutrient enrichment can lead to increased release rates of greenhouse gasses from streams (Rosemond et al., 2015a) including nitrous oxide (Beaulieu et al., 2011). Since recent work has shown that most lakes and some marine habitats are also net-heterotrophic

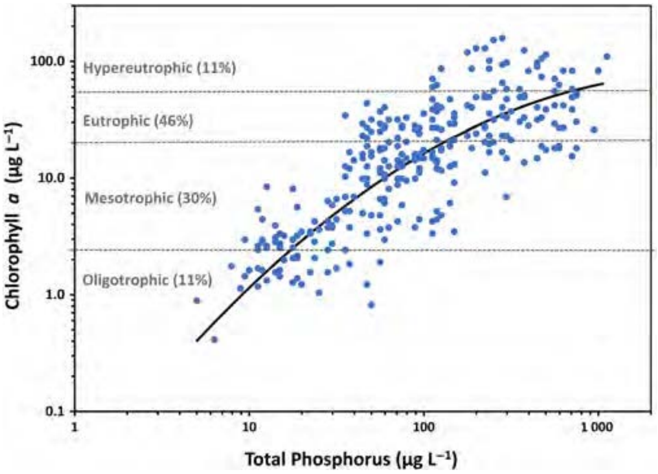


FIGURE 3 Relationship between total phosphorus and phytoplankton concentrations in 292 streams and rivers primarily in North America (79%; especially tributaries of the Missouri and lower Mississippi Rivers) and Europe (16%). Data were derived from the data set of Van Nieuwenhuyse and Jones (1996). Log Chl = −1.65 + 1.99 log TP—0.28 (Log $TP)^2$. Dotted lines show suggested boundaries between oligotrophic, mesotrophic and eutrophic river conditions (Dodds et al., 1998)

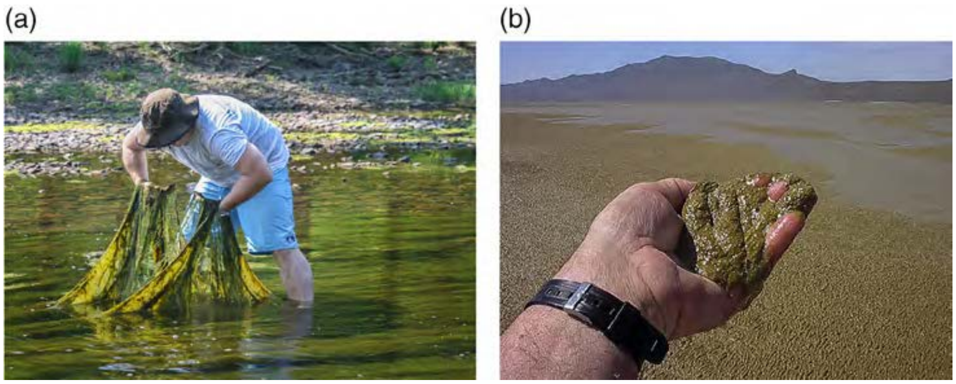


FIGURE 4 (a) Filamentous algae (*Cladophora* sp.) in a nutrient-impacted Greenbrier River, West Virginia. (b). Bloom of the cyanobacterium, *Nodularia spumigena* in Farmington Bay of the Great Salt Lake (Utah, USA). This toxic species creates dangerous blooms in saline estuaries such as the Baltic Sea and Gippsland Lakes, Australia. Photo credits: a) West Virginia Department of Environmental Protection; b) Wayne Wurtsbaugh

(i.e., more respiration of organic carbon than produced by photosynthesis) the importance of nutrients in regulating heterotrophic processes also needs further investigation in lakes and coastal environments (Dodds & Cole, 2007).

Economic costs of eutrophication—Economic costs of these eutrophication problems are substantial, but difficult to estimate. In the freshwater of the U.S. Dodds et al. (2009) estimated costs of approximately \$2.4 billion per year (2015 U.S. dollars), attributable to loss of property values of lake-front homes (49%), costs of purchases of bottled water due to taste and odor problems (25%), losses from recreation (24%) and costs of protecting endangered species (2%). This is an underestimate, because factors such as water treatment plant costs for toxin and taste and odor problems algae were not included. For example, Toledo, Ohio on Lake Erie is spending approximately \$3 million per year to remove cyanotoxins from the city's drinking water (Philpott, 2015), and this cost is being replicated across many cities worldwide. Eutrophication in coastal areas may lead to annual economic costs of near \$100 million in the U.S. and \$1 billion US in the European Union (Davidson et al., 2014). These costs are due to losses related to public health (45%), commercial fisheries (37%), recreation (13%) and monitoring and management (4%). Losses of shellfish and finfish in the Northern Gulf of Mexico's "dead zone" and in the Chesapeake Bay are prominent examples of eutrophication costs (Turner & Rabalais, 2019). Toxic algae and low oxygen levels also cause losses of hundreds of millions of dollars in marine and freshwater aquaculture systems (Proenca & Hallegraeff, 2017).

4 | SPATIAL DIFFERENCES IN NUTRIENTS AND EUTROPHICATION

4.1 | Natural processes influencing nutrient delivery to waterways

The underlying geology and age of watersheds influences the relative releases of N and P into stream water (Sardans, Rivas-Ubach, & Penuelas, 2012). Watersheds with metasedimentary rocks often contain elevated concentrations of N, whereas those with volcanic lithologies or phosphate rock yield waters with relatively high P concentrations (Olson & Hawkins, 2013). Phosphorus leaching decreases with geological age whereas N export can increase due to terrestrial microbial N fixation (Hedin, Vitousek, & Matson, 2003). Glaciation removed soils from many areas of the northern hemisphere 12,000–15,000 years ago, leading to the expectation of more N-limited waters in areas with younger soils, and P-limited waters in geologically older regions. However, climate and vegetation also influence N and P release, and the impacts of soil age, climate, and vegetation on nutrient limitation are not clear (Sardans et al., 2012).

Salinity, and particularly, sulfate concentrations, may also influence N:P ratios along salinity gradients. Sulfur complexes with iron that would otherwise retain P in sediments (Blomqvist, Gunnars, & Elmgren, 2004; Caraco, Cole, & Likens, 1993). In continental waters, sulfur concentrations are most closely related to the lithology of a watershed (Olson & Hawkins, 2012), and hence vary widely between watersheds. In freshwater lakes with low sulfate concentrations, P can remain bound in the sediments, contributing to high N:P ratios and P limitation, whereas in lakes enriched in sulfate, and particularly in salt lakes and coastal systems with abundant sulfate, P is more easily released from the sediments into the water column. Consequently, in these ecosystems, N can more frequently limit algal growth (Howarth & Marino, 2006), particularly if N-fixing cyanobacteria are unable to fix sufficient N (see below).

4.2 | Anthropogenic nutrients and eutrophication

Wastewater discharge, agriculture and other anthropogenic activities generally override natural processes and substantially increase N and P concentrations (Moss et al., 2013). Production of P has increased over 18-fold since 1940 (U.S. Geological Survey, 2014), and that of N over sixfold (Millennium Ecosystem Assessment, 2005a). A portion of these additional nutrients inevitably enters the aquatic environment. Consequently, the annual flux of N to the oceans has nearly doubled from background conditions and the flux of P has tripled (Millennium Ecosystem Assessment, 2005b).

The N:P ratio entering waterways is useful as a measure of which nutrient may limit algal growth limitation (Redfield, 1958). When TN:TP in the environment is >14:1 (by mass), N may be present in excess and P often limits algal growth: When N:P < 14, N is often the limiting factor (Downing & McCauley, 1992). However, the bioavailability of each nutrient and the nutrient needs of different algal taxa makes this relationship far from precise, and some authors have suggested using different ratios to assess nutrient limitation (Morris & Lewis, 1988; Tank & Dodds, 2003). Understanding which nutrient(s) limit algal growth help managers decide what control measures should be implemented to reduce eutrophication.

Point-sources of nutrients—Efforts to control point sources of nutrient loading to waterways have been successful in developed countries, especially for P. Aggressive removal of P from laundry detergents and sewage effluent has hastened this change (Litke, 1999). van Puijenbroek, Beusen, and Bouwman (2018) estimated that by 2010, tertiary or quaternary treatment to remove P was employed in 56% of North American treatment plants, and 75% of those in Western and Central Europe. In developing countries, however, advanced treatment is rare—for example, in Latin America only 10% of plants have advanced nutrient removal, and in sub-Saharan Africa virtually no nutrients are removed. Nutrient removal from point sources has been most effective in controlling eutrophication in those situations where discharges from large metropolitan cities impacted lakes and estuaries.

Nonpoint loading—Control of nonpoint pollution from agriculture and other sources has been much more difficult. The limited successes in nonpoint control have constrained the overall control of eutrophication because these sources contribute 82–84% of the P and N that flow into our waterways (Carpenter et al., 1998; 1990s estimate). The difficulty in controlling nonpoint pollution stems from the diversity and dispersion of sources. Agriculture contributes approximately 53 and 48%, respectively, of the N and P pollution in the U.S., and loading will often come from hundreds or thousands of farms and ranches along a waterway. About 80% of the nitrogen and 25–75% of phosphorus fertilizers are lost from the fields where they are applied, and enter the environment (Sutton et al., 2013). Accelerating use of N and P in fertilizers since the 1950s has exacerbated this problem (Carpenter et al., 1998; Glibert & Burford, 2017). Urban runoff into storm drains also contributes substantial, and again, from hundreds or thousands of sources. Control of nonpoint pollution has also progressed more slowly than for point sources because governments have largely relied on voluntary compliance for the former, whereas laws have usually been enacted to require point-source compliance (Boesch, 2019).

Land use categories influence the concentrations and proportions of N and P that move into our waterways (Figure 5; U.S. Geological Survey, 2000). Croplands in temperate developed countries yield the highest concentrations of total nitrogen (TN), primarily as oxidized nitrate, with moderate amounts of ammonium and organic N (Figure 5a). Runoff from urban landscapes can also yield surprisingly high concentrations of both nitrate and ammonium. In contrast, forested lands not subject to high amounts of atmospheric deposition yield low quantities of TN and insignificant amounts of ammonium. Urea, which is increasingly used as a N fertilizer (Glibert, Maranger, Sobota, & Bouwman, 2014), is not often measured in waterways and is

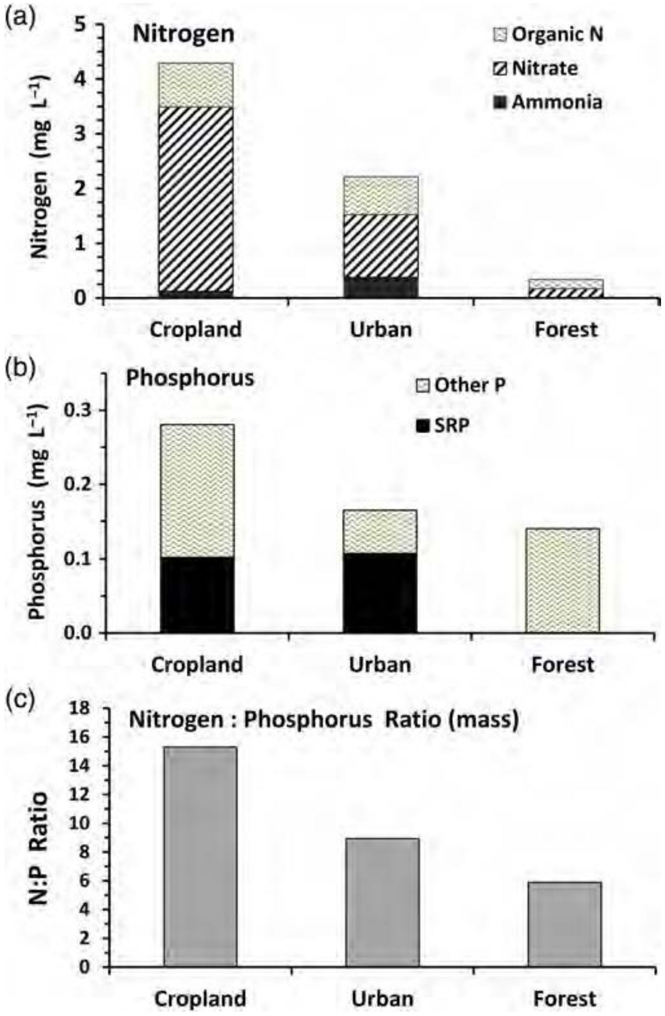


FIGURE 5 Flow-weighted mean nutrient concentrations in rivers draining cropland ($n = 104$), urban ($n = 38$) and forest ($n = 36$) watersheds. (a). Nitrogen, showing the most readily bioavailable forms of ammonia and nitrate as well as organic nitrogen. (b). Phosphorus concentrations in readily bioavailable soluble reactive phosphorus (SRP) and other less bioavailable forms. (c). Total N:Total P ratio (by weight). The nutrient data were derived from a U.S. Geological Survey NAWQA survey (USGS, 2000)

a component of the organic N pool shown in Figure 5a. Phosphorus losses from land follow a similar pattern to that of N (Figure 5b), with particularly high concentrations in agricultural and urban effluents (Tromboni & Dodds, 2017). Both cropland and urban landscapes yield large quantities of biologically available soluble phosphate, whereas forested landscapes yield primarily refractile forms of P (U.S. Geological Survey, 2000; Uusitalo, Yli-Halla, & Turtola, 2000).

The N:P in receiving streams varies markedly with land use (Figure 5c, USGS, 2000). Drainage from agricultural lands has an average ratio of over 14:1 (by mass), often producing P-limited conditions for algal growth. The average ratio of urban runoff (9:1) suggests limitation by N, whereas the mean low N:P ratio of forested ecosystems (6:1; Figure 5c) indicates that algae in receiving waters may be N limited.

Atmospheric deposition of nutrients—Atmospheric deposition of N and P is another type of nonpoint pollution that has been difficult to control. Atmospheric deposition over land has increased threefold, and twofold over the oceans (Kanakidou et al., 2016). High amounts of atmospheric N deposition have led to N-saturation of watersheds with high nitrate exports into streams, lakes, and estuaries. For example, in old-growth forests in the eastern United States, where atmospheric deposition is high, nitrate concentrations in stream water are near 550 μg N L $^{-1}$, whereas pristine streams in old-growth forests of

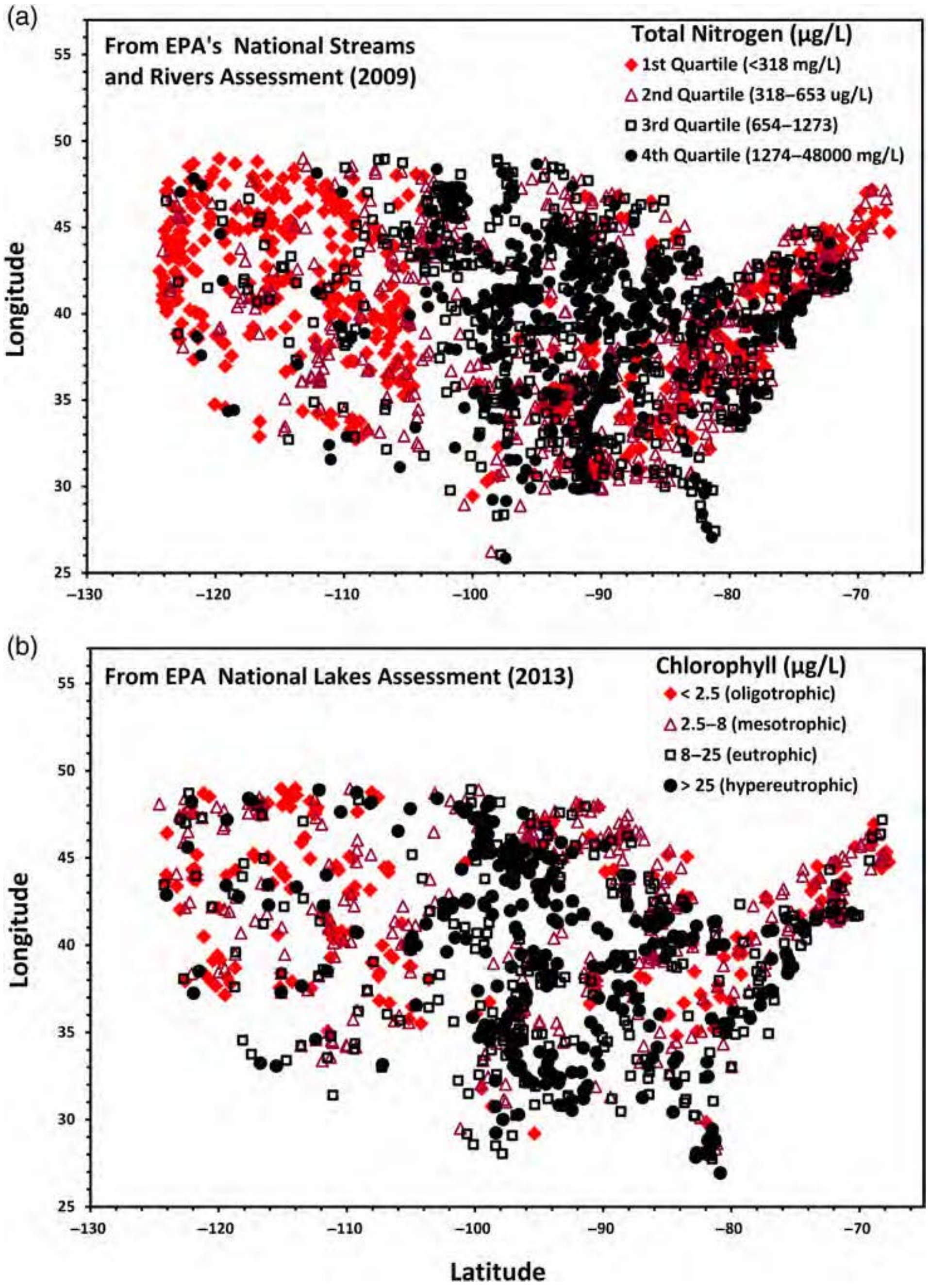


FIGURE 6 (a) Total nitrogen concentrations in 1894 streams and rivers in the United States (derived from EPA, 2013a, 2013b). Total nitrogen can be used as one index of eutrophication (Dodds, 2006). The pattern of total phosphorus (not shown) in stream waters was similar to that for nitrogen, although more western streams had high phosphorus concentration. (b). Chlorophyll *a* levels in the in 1264 United States lakes studied in the Environmental Protection Agency's National Lakes Survey (EPA, 2009a). Only lakes from the EPA database where TP was ≥3 μg L^{-1} were used in the analysis. The trophic state classifications were based on Caspers (1984) for annual mean chlorophyll *a* levels

temperate South America have mean nitrate concentrations <10 μg N L^{-1} (Perakis & Hedin, 2002). Atmospheric deposition of Fe, N and P in dust can also be important, either from local disturbed arid areas, or from continental-scale transport to the oceans (Brahney et al., 2015; Jickells & Moore, 2015).

Spatial distribution of eutrophication in the U.S.—Nutrient patterns of US rivers measured in an extensive study by the Environmental Protection Agency (EPA, 2013b) demonstrate how concentrations vary widely across the U.S. (Figure 6a). Streams and rivers in the Midwestern agriculture belt of the United States have particularly high levels of nutrients, as do many Eastern coastal waterways. In contrast, most rivers in forested areas of the Western U.S., Appalachia and the upper Northeast have relatively low nutrient levels. In most Ecoregions of the U.S., N concentrations in rivers have increased more than P in comparison to historic concentrations causing the N:P ratio to increase (Figure 7; Dodds & Smith, 2016). Many streams in agricultural areas or those receiving wastewater discharges with high concentrations of both N and P exhibit abundant growths of filamentous algae (Figure 4), and larger, deeper rivers with low flushing rates are subject to planktonic blooms as are lakes and estuaries (Van Nieuwenhuyse & Jones, 1996).

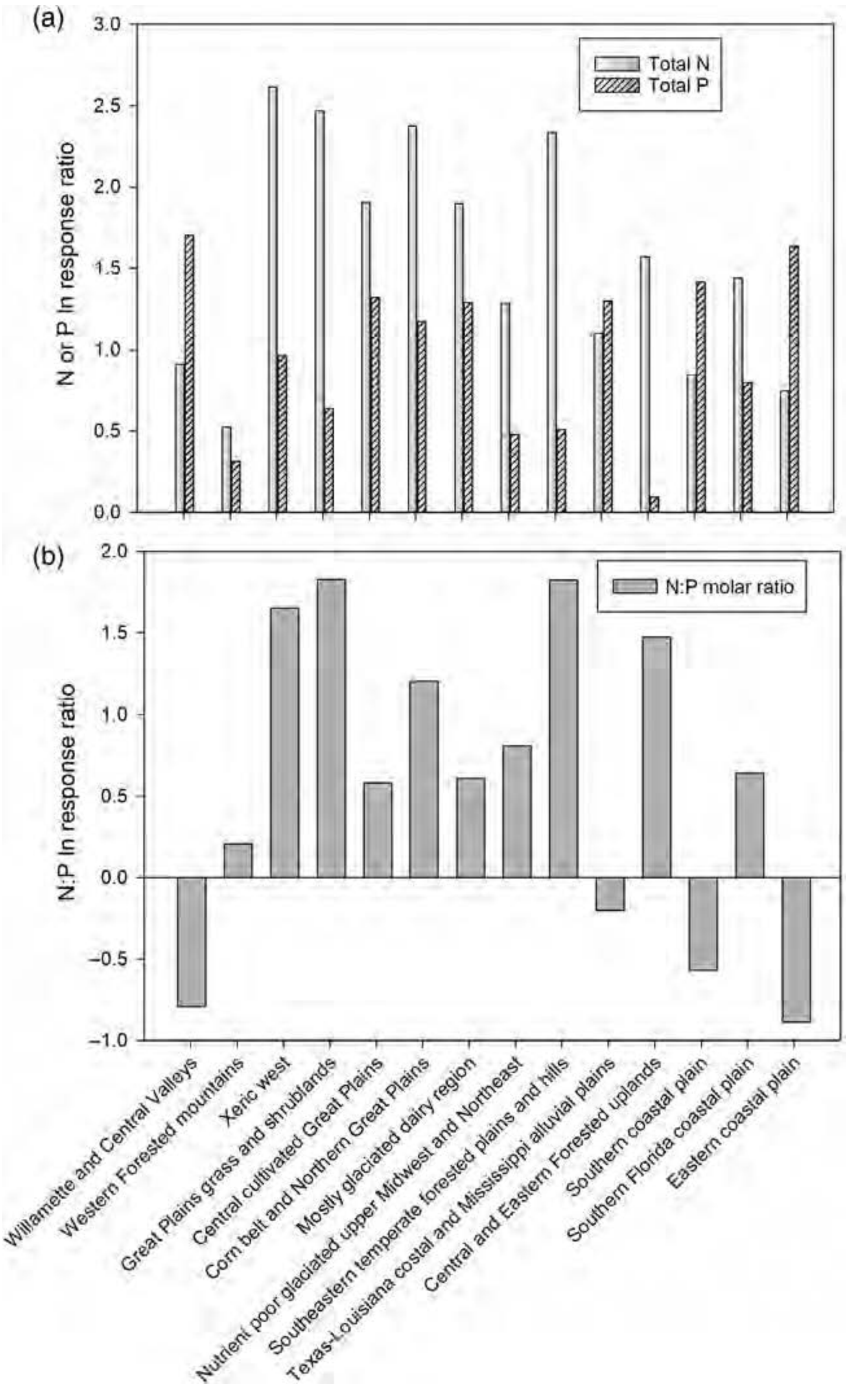


FIGURE 7 (a) Relative increases of total nitrogen and total phosphorus in rivers in 14 Ecoregions of the U.S. values are plotted as response ratios [= ln (current concentration/historic concentration)]. (b) Relative changes in the N:P response ratios in the 14 ecoregions. Derived from Dodds and Smith (2016). For reference, a response ratio of 1.6 indicates a fivefold increase in N or P or the N:P ratio

Eutrophication patterns in lakes and reservoirs across the U.S. (Figure 6b) were similar to the nutrient concentration patterns in rivers (Figure 6a), an expected finding since most lakes receive the majority of their nutrients from tributaries. Survey data from 1,264 lakes in the United States (EPA, 2009a) indicate concentrations of chlorophyll *a*, an indicator of phytoplankton biomass, were very high in many of the lakes sampled (Figure 6b). Over 50% of the surveyed lakes were eutrophic or hypereutrophic and 30% were mesotrophic. Oligotrophic lakes with $<2.5\ \mu g\ L^{-1}$ of chlorophyll *a* represented only 19% of the survey group. However, the distribution of lakes of different trophic status was uneven. Lakes and reservoirs on the US east coast, Florida, the southwest and particularly the agricultural areas of the Midwest were primarily eutrophic and hypereutrophic. In contrast, western lakes and those in the northeast including the Appalachian Mountains were less productive, reflecting less nutrient runoff from agriculture, and lower population densities with consequent decreased urban nutrient pollution.

5 | HARMFUL ALGAL BLOOMS

HABs are increasingly being recognized as one of the most negative aspects of cultural eutrophication (Paerl & Otten, 2013b). Excessive nutrient loading increases blooms of toxin-producing algae such as cyanobacteria and dinoflagellates in lakes and reservoirs, estuarine and coastal waters, and stimulates the growth of undesirable attached algae in streams and rivers (Dodds, Carney, & Angelo, 2006; Lewis et al., 2011). HAB toxins can make waters unsafe and unusable for drinking, irrigation, and recreational purposes (Koreiviene, Anne, Kasperoviciene, & Burskyte, 2014), rendering them dysfunctional from ecologic, economic, and esthetic perspectives (Figure 4).

Some algal toxins (Table 1) are more toxic per unit mass than cobra venom (Cheung, Liang, & Lee, 2013), are generally nonspecific, and can harm humans and many other animal taxa. Marine algal toxins commonly cause poisonings through the consumption of bioaccumulated toxins in fish and shellfish (Grattan, Holobaugh, & Morris Jr., 2016). Although acute poisoning of humans by fresh- and brackish-water cyanotoxins is rare, livestock and are commonly killed because they drink bloom-impacted waters (Henriksen, Carmichael, An, & Moestrup, 1997; Stewart, Seawright, & Shaw, 2008a). Mass mortalities of birds due to cyanotoxins are also common (Alonso-Andicoberry, Garcia-Villada, Lopez-Rodas, & Costas, 2002; Stewart, Seawright, & Shaw, 2008b). Bioaccumulation of freshwater cyanotoxins in fish and their subsequent consumption by humans and other animals can be a significant and sometimes dominant route of exposure (Carmichael & Boyer, 2016; Grattan et al., 2016; Poste, Hecky, & Guildford, 2011). Liver toxins (hepatotoxins) occur in a variety of cyanobacterial species, most notably the wide-spread nuisance taxa, *Microcystis* spp. which in 2007 overwhelmed drinking water plants at eutrophic Lake Taihu, China, cutting drinking water supplies off for ~10 million consumers in the Taihu Basin (Paerl et al., 2011); in 2014 blooms also led to the closure of the drinking water supply of metropolitan Toledo, Ohio, a city of 650,000 located on Lake Erie (Ho & Michalak, 2015). Cyanobacterial toxins from a reservoir killed 76 people in Brazil in 1996 when the filtration system at a dialysis clinic failed. A recent study also detected a significant correlation between HABs and liver-related disease deaths in the U.S. (Zhang, Lee, Liang, & Shum, 2015).

Scientists currently cannot predict when an algal bloom will be harmful because not all species in blooms produce toxins under all conditions, and the types and amounts of toxins vary (Table 1). Light intensity, macronutrients (N and P), micronutrients (iron and other trace metals), and temperature can influence toxin production, but definitive understanding of environmental regulators is still an ongoing field of investigation (e.g., Glibert, 2017; Otten & Paerl, 2015). In general, high temperature and nutrients coupled with low turbulence tend to encourage cyanobacterial blooms. In coastal areas, toxic algal blooms may be categorized as "high-biomass" blooms that are more clearly controlled by macronutrient loading, and "low-biomass" blooms of dinoflagellates, diatoms and other taxa that are not so clearly controlled by nutrient loading, but rather by factors such as turbulence, light availability and water residence times (Davidson et al., 2014; Richardson, 1997). Organic nutrients and mixotrophy are also frequently associated with toxic blooms in coastal waters (Glibert, 2017).

Nitrogen and phosphorus loading, and their relative amounts can influence what types of harmful algae are stimulated. Reduced N forms such as ammonium and urea can favor dinoflagellates and non-nitrogen-fixing cyanobacteria (e.g., *Microcystis* spp.; *Planktothrix agardhii*), whereas the dominant oxidized form, nitrate, can favor diatoms, green algae and some other taxa (Blomqvist, Pettersson, & Hyenstrand, 1994; Glibert et al., 2016; Newell et al., 2019). High nutrient loads of any form can promote production of organic matter, and the subsequent decomposition and release of regenerated ammonium from the sediments can support subsequent summer cyanobacterial HABs like *Microcystis* spp. that do not fix N (Newell et al., 2019). Similarly, lakes and estuaries impacted by wastewater treatment effluents or industrial sources high in ammonium may favor cyanobacteria over more beneficial types of phytoplankton, especially in warm summer months

TABLE 1 Toxins and irritants produced by harmful algal blooms

Toxin	Toxin producers	Structure	Mode of action/human health effect	Lethal dose$_{50}$ (μg/kg)	Bioaccumulation?
Dermal irritants	*Lyngbya, Dolichospermum (Anabaena), Aphanizomenon, Nodularia, Oscillatoria, Gloeotrichia*	Varied	Allergic reaction through dermal contact in sensitive individuals; swimmers itch	Not lethal	—
Microcystin-LR	*Microcystis* spp.	Moncyclic heptapeptides	Inhibition of protein phosphatases; liver hemorrhaging & tumor promotion	50	—
Nodularin	*Nodularia* spp.	Cyclic petapeptide	Inhibition of protein phosphatases; liver tumors	50	In shellfish & other marine organisms
Ciguatoxin	Dinoflagellate, *Gambierdiscus toxicus*	Polycyclic ether	Acute neurologic disease manifested with: Gastrointestinal, neurologic and cardiovascular	0.25	In predatory marine fishes
Domoic acid	Pseudo-nitzschia spp.	Domoic acid	Amnesiac shellfish poisoning; memory loss, brain damage, and death in severe cases	3,600$_{\text{mice}}$	In shellfish, anchovies, and other fish
Saxitoxins	Various dinoflagelates & cyanobacteria	Trialkyl tetrahydropurine	Neurotoxin. Sodium & calcium channel blocking in neurons; respiratory muscle paralysis	10–200	Via marine shellfish
Cylindrospermopsin	*Cylindrospermopsis, Aphanizomenon* spp., *Dolichospermum, Lygnbya* sp.	Polyketide-derived alkaloid	General cytoxic, hepatoxic and neurotoxic effects. Liver, kidney, thymus & heart damage.	200	Production under low nitrogen conditions
Anatoxin-a	*Dolichospermum, Oscillatoria, Aphanizomenon, Cylindrospermum*	Alkaloid	Blocks postsynaptic depolarization	250	—
Anatoxin-a(s)	*Dolichospermuma, Aphanizomenon*	Unique organophosphate	Irreversible inhibitor of acetylcholinesterase	40	—

Note: Derived from World Health Organization (2003), Pearson, Mihali, Moffitt, Kellmann, and Neilan (2010) and TOXNET (2013). Note that there are many strains of cyanobacteria that produce isoforms that vary in their toxicity. The lethal dose killing 50% of exposed mice (LD$_{50}$) of the more common forms are listed here. For comparison, the lethal dose of king cobra venom is near 185 μg/kg.

(Paerl & Otten, 2013b). More research is needed to assess how different forms of nutrients influence algal metabolism, growth, toxin production and eventually the community structure that influences water quality.

Nitrogen additions can also promote toxin production in freshwater cyanobacteria (e.g., Anderson, Glibert, & Burkholder, 2002; Davis, Bullerjahn, Tuttle, McKay, & Watson, 2015) since most cyanotoxins contain substantial amounts of N. However, cyanotoxin production has also been found in lakes with both low and high levels inorganic N (Gobler et al., 2016), and toxin levels can correlate with P concentrations (Poste, Hecky, & Guildford, 2013). Experimental work has shown that both P-limitation and N-limitation induced the production of ichtyotoxins in some marine flagellates (Johansson & Granéli, 1999). The relationships between nutrients, algal growth and toxin production is not simple, because as stated by Glibert (2017), *"[Nutrient] ratios alter biodiversity by virtue of the fact that different taxonomic groups … have distinct eco-physiological characteristics with respect to nutrient requirements*." Each water body has hundreds of different taxa in the phytoplankton and periphyton communities, each with specific needs and functions, so differential responses to N, P and other nutrients are to be expected.

6 | RELATIVE IMPORTANCE OF NITROGEN AND PHOSPHORUS FOR CONTROLLING EUTROPHICATION

The need to control eutrophication in lakes, rivers, estuaries, and coastal oceans is broadly recognized. However, there is considerable debate as to when and where P, N, or both should be managed and/or regulated. Some argue that only P should be controlled based on cost and because N-fixing cyanobacteria may be able to overcome N deficiencies (e.g., Higgins et al., 2018; Schindler et al., 2008; Schindler et al., 2016). Others, however, have questioned whether P control alone is sufficient (e.g., Glibert, 2017; Lewis & Wurtsbaugh, 2008; Paerl et al., 2016; Shatwell & Köhler, 2018). Moss et al. (2013) evaluated situations where both P and N can control algal growth. They argued that most pristine aquatic environments were largely in balance relative to the N and P needed for algal growth, and that pollution with N or P has forced communities toward limitation by one or the other nutrient. For example, fertilized cropland and atmospheric N deposition may have lessened N limitation in portions of North American and Eurasia, causing P to frequently be the limiting nutrient for those environments (Elser et al., 2009; Stoddard, 1994). Conversely, releases of sewage into coastal estuaries with large cities may have caused proportionately high P loads, leading to more N-limited conditions (Moss et al., 2013).

The natural and anthropogenic factors discussed previously that influence nutrient loading into streams, lakes, and coastal oceans influences can have important consequences influencing which nutrient(s) may be controlling algal growth in a given area at a specific time. Nutrient loads from primary and secondary-treated urban wastewater are rich in P and typically have low N:P ratios (ca. 4:1 by weight), but this ratio can be even lower in regions where phosphate detergents are still allowed (Carey & Migliaccio, 2009), as is the case in many developing countries. Many densely populated urbanized areas deliver high P loads to estuaries and coastal areas relative to N. In contrast, nonpoint source pollution from conventional agriculture is very high in bioavailable nitrate relative to P (Figure 5c; EPA, 2009b). Phosphorus binds to soil particles and enters stream waters primarily as particulate P, and only about 30% of this phosphorus is bioavailable (Uusitalo et al., 2000). Consequently, median N:P ratios of *bioavailable* nutrients from conventional tillage agriculture are near 21:1, much higher than the 7:1 (by weight) needed by most algae and cyanobacteria. We expect algae in streams, lakes, and estuaries influenced primarily by agriculture pollution to be P-limited until algal growth is so high that light or some other factor limits algal growth.

Both nutrient loading and the complex processing and losses of N and P within a water body determines what is actually available for algal growth (Figure 8). In streams dominated by benthic periphyton in complex biofilms, nutrients are removed from the water column into the biotic community, processed by various trophic levels, and then are released into the water column, moving downstream in the current, in a process called nutrient spiraling (Ensign & Doyle, 2006). Water residence times of both P and N are usually measured in days or less at baseflow (Hall Jr., Baker, Arp, & Koch, 2009). The algal production and any other metabolic activities that are locally important are most heavily influenced by average nutrient concentrations delivered by the water column during baseflow. Downstream nutrient transport, however, is often dominated by high flows with scouring events and those that move larger particles (Banner, Stahl, & Dodds, 2009; Meyer & Likens, 1979). Nitrogen fixation in the biofilms can help offset N deficiencies (Scott & Marcarelli, 2012). Streams can either be depositional or erosional, retaining or transporting nutrients. Lowland rivers and streams with low elevation loss are more likely to be depositional. In these systems nutrients are moved into riparian wetlands and lakes and captured by the biota (Hamilton, 2010).

In estuaries, and more importantly in lakes, longer residence times allow greater processing and nutrient accumulation (Figure 8; Richardson et al., 2018). Incoming P either sediments to the bottom or is incorporated into the periphyton or phytoplankton. Particulate P sinks, and anoxia ultimately may allow release of highly bioavailable phosphate into the water column

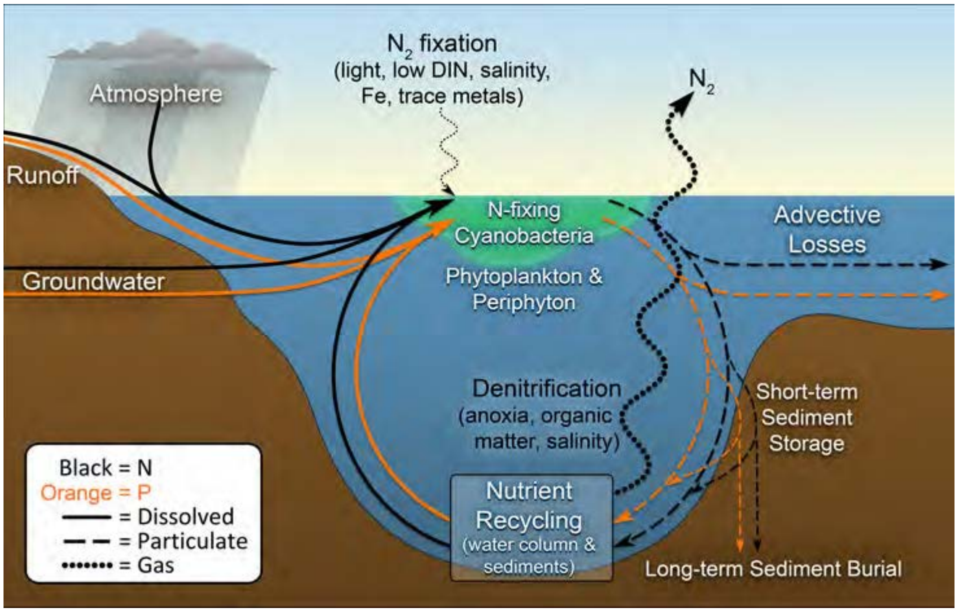


FIGURE 8 Simplified view of how external loading and cycling of nitrogen and phosphorus interact with loss factors for these nutrients to control the size of algal populations and cyanobacteria in inland and coastal waters. Some environmental factors influencing N_2 fixation and denitrification are shown. Adapted from (Paerl et al., 2016). Phosphorus loss from the system is due to advective loss and burial. High sediment concentrations of phosphorous provide "legacy" nutrient loading back into the water column. Although this can also be a problem for nitrogen, denitrification can remove much of the sedimented nitrogen from the system

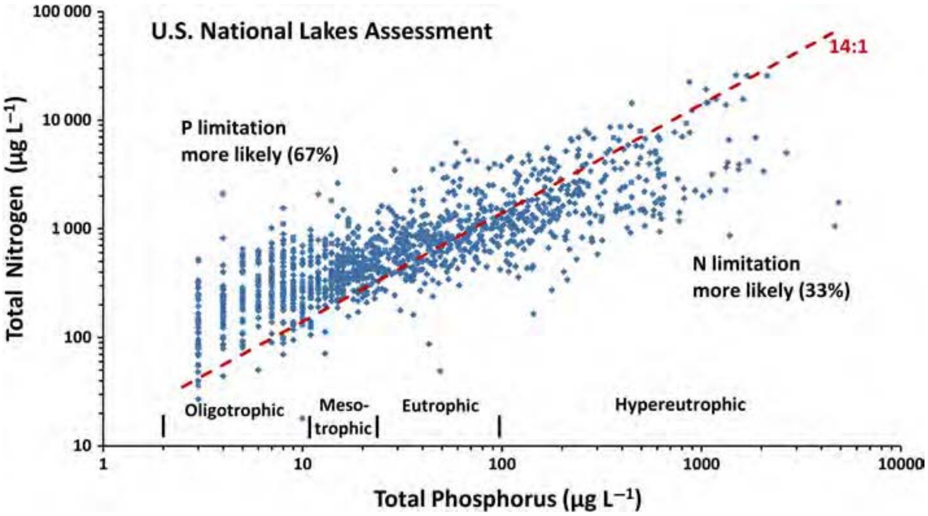


FIGURE 9 Relationship between nitrogen and phosphorus concentrations in 1268 lakes and reservoirs sampled by the EPA (2009a, 2009b) during the National Lakes Assessment (NLA). The red dashed line shows where there is a 14:1 ratio (by weight) between N and P (Downing & McCauley, 1992). Classification of lake trophic state is from Carlson (1977)

(Hupfer & Lewandowski, 2008). Nitrogen processing is more complex with the dominant factors being N fixation by benthic and planktonic cyanobacteria which brings new N into the ecosystem, and denitrification which removes N from the bioavailable pool for most organisms. The balance of N fixation and denitrification is a primary determinant of whether a water body can compensate for a deficiency in this nutrient (Paerl et al., 2016; Scott, McCarthy, & Paerl, 2019).

6.1 | Monitoring and small-scale experimental evidence for N and P control of eutrophication

One approach used to determine whether N or P is the nutrient limiting algal growth is to compare the ratios of these in the water, to the needs of the organism. Although most algae have a ratio near 7 N:1P by weight (16:1 M—the Redfield ratio; Redfield, 1958), experiments have shown that when N:P is >14 (by weight), N is present in excess and P most likely limits algal growth. Conversely, when N:P < 14, N may be limiting (Downing & McCauley, 1992; Scott et al., 2019). A random sampling of nutrient concentrations in over 1,200 United States lakes in the National Lakes Assessment (EPA, 2009a) indicates: (1) the tremendous range of N and P concentrations in lakes; (2) that approximately 67% are more likely to be limited by P, and 33% by N, and; (3) that P is more commonly limiting in oligotrophic and mesotrophic lakes, whereas N is more likely limiting in nutrient-rich eutrophic and hypereutrophic systems (Figure 9). Similar conclusions were reached by Downing and McCauley (1992), using a smaller data set. This nutrient ratio approach has limitations, however, because not all N or P in the water is bioavailable to algae, and many lakes have ratios that fall close to 14:1, making predictions difficult. While

TN:TP ratios can be misleading (e.g., when there are high levels of unavailable inorganic particulate P or recalcitrant dissolved organic N), in most cases they are the best indicator of nutrient availability in freshwaters (Dodds, 2003) and N:P agrees with numerous other physiological and growth-based tests for nutrient limitation (Dodds & Priscu, 1990).

Empirical analyses of algal biomass (measured as chlorophyll) relative to P and N levels in lakes and streams suggests that these two nutrients act synergistically to promote eutrophication (Figure 10). The EPA National Lake Survey (EPA, 2009a) discussed above was used to determine how total N and total P were related to chlorophyll concentrations. Both TN and TP were equally well correlated ($p < .001$) with chlorophyll concentrations (each with $r^2 = 0.53$), and a multiple linear regression with both nutrients improved the relationship ($r^2 = 0.58$). Previously, Smith (1982) did a similar analysis of a more restricted number of lakes and also found that N and P acted together to increase phytoplankton biomass in lakes. However, as pointed out by Lewis and Wurtsbaugh (2008), attributing causation to these correlations is problematic because phytoplankton are made up of both N and P and thus some degree of autocorrelation is implicit. Analyses of streams have also shown a positive relationship between benthic algal biomass and water column nutrients, with models using both N and P explaining more of the variance in benthic chlorophyll than either alone (Dodds, Smith, & Lohman, 2002). Other stream and river systems appear to be limited primarily by P (e.g., Hilton, O'Hare, Bowes, & Jones, 2006; Moisander, Steppe, Hall, Kuparinen, & Paerl, 2003).

Short-term bioassays provide a more rigorous test of what nutrient(s) might be limiting algal growth at a particular location and at a specific time than do broad-scale correlations. In a global survey of over 1,600 small-scale bioassays Elser et al. (2007) found that phytoplankton and stream periphyton were about equally limited by P and N. In contrast, periphyton on the bottom of lakes was more frequently limited by P than by N. This meta-analysis also demonstrated that adding N simultaneously with P frequently increased algal growth over single nutrient additions. A detailed experimental analysis of nutrient-limited streams of eight ecoregions of the U.S (Johnson, Tank, & Dodds, 2009) also showed that in less impacted reference streams, periphyton increased most when N or when N and P were added jointly. However, in impacted agricultural regions where N in runoff is high (Figure 5), P limitation was more prevalent. Their study also found that heterotrophic activity (decomposition of organic matter) was more frequently colimited by both N and P than was autotrophic activity. Given the importance of exogenous organic matter for both lakes and streams, future research should address nutrient controls on heterotrophic activity.

The Elser et al. (2007) meta-analysis of 485 bioassays from coastal waters reported that both phytoplankton and periphyton were more limited by N than by P, but like the freshwater studies, additions of N + P usually acted synergistically. Another meta-analysis by (Downing, Osenberg, & Sarnelle, 1999) also found that phytoplankton in estuaries, harbors, bays and landlocked seas were more frequently stimulated by N than by P. The more efficient cycling of P from sediments in sulfate-rich marine water described by Blomqvist et al. (2004), coupled with denitrification is consistent with the fact that these systems

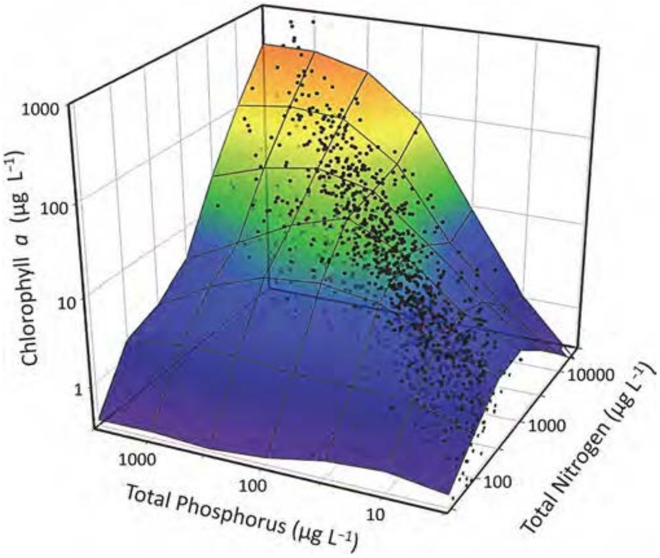
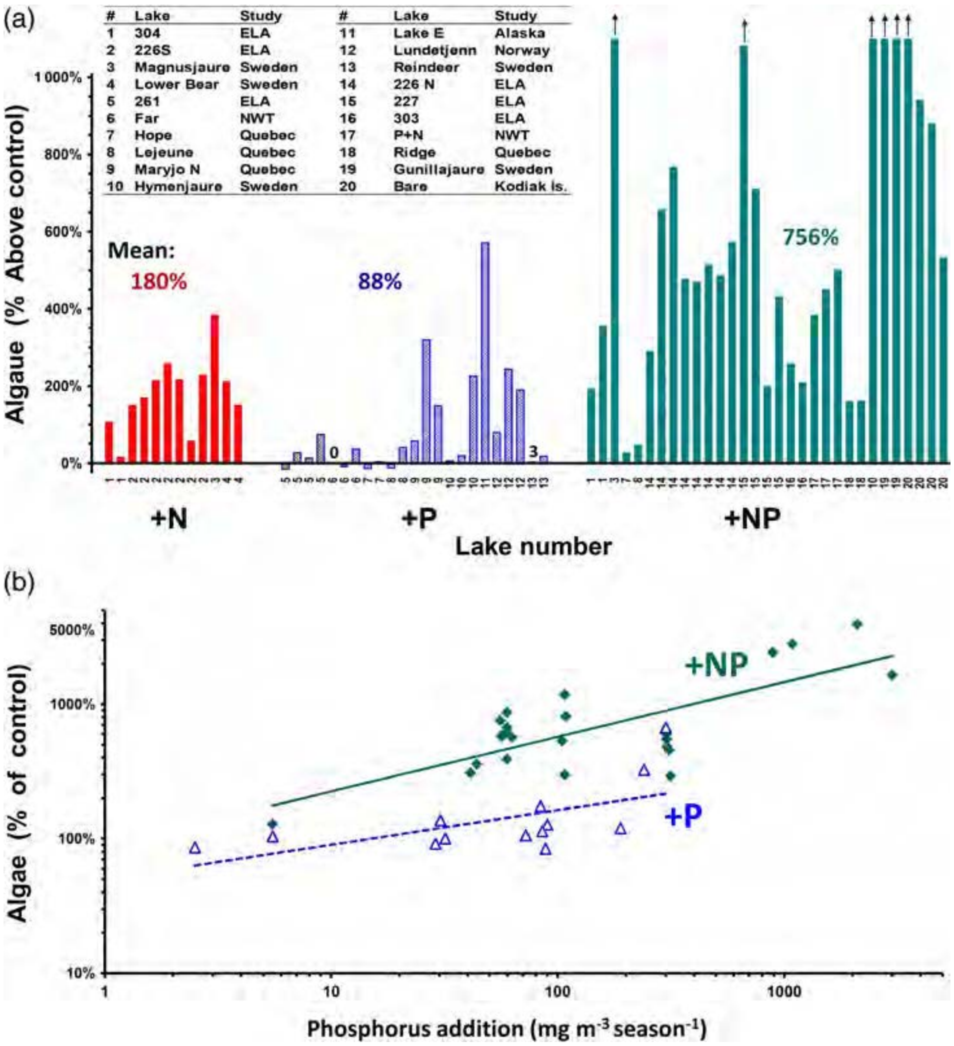


FIGURE 10 Relationship between phytoplankton chlorophyll levels (*z*-axis) and total phosphorus and total nitrogen levels in 1264 lakes studied in the Environmental Protection Agency's National Lakes Survey (EPA, 2009a). Only lakes from the EPA database where TP was ≥3 μg L^{-1} were used in the analysis. The surface curve was fit with local regression smoothing (LOESS; SigmaPlot) and a sampling proportion of 0.4. Dodds and Smith (2016) recently reported a very similar relationship between N, P and chlorophyll in benthic periphyton of streams

are frequently N-limited. Downing et al. (1999) found that in oceanic zones with limited upwelling of deep nutrients, both iron and N may be limiting, a finding consistent with the recent analysis of Moore et al. (2013).

7 | WHOLE-SYSTEM ANALYSES OF NUTRIENT LIMITATION

Although short-term bioassays provide a "snapshot" of nutrient limitation, they do not necessarily indicate how a community will respond to long-term nutrient enrichment (Schindler et al., 2008). For example, in lakes with long residence times, N fixed by cyanobacteria may be able to build up and alleviate N deficiencies (Higgins et al., 2018; Schindler et al., 2008), or



#	Lake	Study	#	Lake	Study
1	304	ELA	11	Lake E	Alaska
2	226S	ELA	12	Lundetjenn	Norway
3	Magnusjaure	Sweden	13	Reindeer	Sweden
4	Lower Bear	Sweden	14	226 N	ELA
5	261	ELA	15	227	ELA
6	Far	NWT	16	303	ELA
7	Hope	Quebec	17	P+N	NWT
8	Lejeune	Quebec	18	Ridge	Quebec
9	Maryjo N	Quebec	19	Gunillajaure	Sweden
10	Hymenjaure	Sweden	20	Bare	Kodiak Is.

FIGURE 11 Whole-lake experiments. (a) Response of phytoplankton to nitrogen (+N), phosphorus (+P), or N + P additions in 20 whole-lake experiments relative to controls (either pretreatment sampling or nearby control lakes). Each lake is indicated by a different number. Some lakes were treated for multiple years (shown sequentially). Although there are many lake-years of analysis, relatively few lakes have been studied to test N-alone (4) or P-alone (9) nutrient responses. Measured, or estimated chlorophyll levels on control lakes ranged from 0.3 to 4.8 μg L^{-1}, with a mean of 1.8 μg L^{-1}, indicating that all had naturally low productivity. Studies were done in the Experimental Lakes area in Ontario, Canada (ELA), northern Sweden, Northwest Territories of Canada (NWT), Norway, Quebec, Alaska, and Kodiak Island (AK). The values over each set of bars are the mean annual responses for each nutrient treatment. Details and references for these studies are given in Appendix S1, Supporting Information. (b) Relationship between phosphorus fertilization rates in the whole-lake experiments with ($n = 8$) and without ($n = 5$) concomitant nitrogen additions. Only a subset of lakes shown in the top frame (a) had sufficient data to calculate addition rates. Some lakes were fertilized for multiple years. Phytoplankton responses were measured as either chlorophyll levels, algal biovolume, or primary production. Both phosphorus level and the presence/absence of nitrogen were significant ($p < .000$) factors influencing responses. Regression equations: +P, Log Response = −0.301 + 0.257 Log [P]; N + P, Log Response = −13.46 + 10.52 Log [P]. Detailed data are given in Appendix S1

conversely, increased eutrophication may stimulate bottom-water anoxia, and denitrification, resulting in the loss of N (Álvarez Cobelas & Piña, 2006; Grantz, Haggard, & Scott, 2014; Seitzinger & Nixon, 1985).

A moderate number of lakes have been fertilized for entire growing seasons or multiple years to better assess long-term responses to N and P (Figure 11; See also Elser, Marzolf, & Goldman, 1990). These whole-lake data should, however, be extrapolated with care because: (a) all of them were oligotrophic lakes located in northern regions; (b) nearly all of them were small but thermally stratified, whereas many lakes with severe eutrophication are often shallow and well-mixed and; (c) most of the studies utilized joint N + P fertilization, with relatively few lakes tested for their response to N or P alone.

Whole-lake experiments with only N additions have been done in only four lakes, but all showed an increase in chlorophyll levels or phytoplankton biomass 100% or more above background or control levels (Figure 11). For example, Lake Magnusjaure in northern Sweden was fertilized for just 1 year with N alone, and the phytoplankton biomass increased 380% above background, indicating strong N limitation (Holmgren, 1984).

Mean whole-lake lake responses to P-only fertilization were lower than for N-only treatments, but with considerable variability among the seven lakes tested (Figure 11a). Phytoplankton in Lake 261 in the Experimental Lakes Area (ELA) of Canada responded only marginally to P-only additions, even after 4 years of P fertilization. In contrast, Lake E in Alaska showed a strong response (570%) to P additions after 1 year of fertilization. Some lakes receiving P for multiple years have shown progressive increases in phytoplankton. For example, phytoplankton in Lake Hymenjaure in northern Sweden showed a progressive increase over 3 years to 230% above controls, and in this experiment, increased N fixation by benthic cyanobacteria helped offset N-deficiencies (Holmgren, 1984). Schindler (2012) also mentions that N-fixing benthic cyanobacteria in Lake 261 of the ELA increased after P fertilization, thus emphasizing the need for nutrient limitation studies that address both benthic and pelagic processes.

In contrast to moderate increases from N- or P-alone additions, phytoplankton in most whole-lake studies that were fertilized with both N and P usually had responses several hundred percent above background (Figure 11a), an expected finding when both nutrients are initially balanced with respect to demand by the phytoplankton. The fertilization of Lake Hymenjaure in Sweden is particularly instructive. As mentioned previously, three successive years of P-alone fertilization increased algal biomass to 230% above background, but when N was also added during the fourth year, phytoplankton levels increased to 1,550% above background (Figure 11). Analysis of a subset of the experimentally fertilized lakes where P loading levels were clearly stated, shows that increments of fertilization with P alone significantly increased phytoplankton abundance ($p = .030$), but concurrent additions of N and P increased phytoplankton abundance or productivity approximately fourfold more than with P-fertilization alone ($p < .001$; Figure 11b).

It is also instructive to review the classic results from ELA Lake 226 N, where N and P were added for 8 years with an average increase of 530% in phytoplankton biomass above background. This response has frequently been interpreted as due to P-alone (Schindler, 2012). However, when contrasted to the true P-alone experiment in the ELA, Lake 261, a more logical interpretation is that the increased algal response in Lake 226 N was due to the combined N + P addition (Figure 11a). The P-alone interpretation of this response was extremely useful for promulgation of P reduction standards and has been instrumental in guiding eutrophication control efforts in North America and Europe. Although the P-alone explanation for Lake 226 N was addressed long ago (Elser et al., 1990; Fee, 1979), this misinterpretation has persisted and impeded efforts to understand when and where N and P may act jointly to influence algal communities.

8 | THE INFLUENCE OF NITROGEN FIXATION, DENITRIFICATION AND PHOSPHORUS RECYCLING ON EUTROPHICATION

In some aquatic ecosystems, N fixation by cyanobacteria can contribute substantially to the N budget (Howarth, Marino, Lane, & Cole, 1988) and thus contribute to eutrophication (Figure 8). This is particularly evident in some eutrophic lakes, streams, and wetlands with dense populations of N-fixing cyanobacteria in the plankton or periphyton (e.g., Schindler et al., 2008; Scott & Marcarelli, 2012). However, in many freshwater and saline systems, N fixation rates are often low relative to external N inputs (Marcarelli, Wurtsbaugh, & Griset, 2006; Scott & Marcarelli, 2012; Vitousek et al., 2002) and contribute little toward ecosystem-scale productivity, despite the fact that many of them exhibit N limitation or N + P colimitation (Box 1).

A critical question in the eutrophication debate is whether N fixation by cyanobacteria can completely satisfy algal N requirement, leaving P as the sole important nutrient (Schindler, 2012). Several lines of evidence indicate that N can remain limiting, even after active N-fixation by cyanobacteria (Baker, Wilson, & Scott, 2018; Paerl, 2018; Paerl et al., 2016; Paerl et al., 2018; van Gerven et al., 2018), and thus, external N inputs can exacerbate eutrophication. First, N fixed in systems may be lost and not accumulate. Denitrification can partially or wholly offset N fixation gains (Figure 8), reducing N accumulation

BOX 1 LAKE ERIE AND THE P VERSUS DUAL-CONTROL DEBATE

The recent surge in toxic *Microcystis* blooms (Figure 12) in Lake Erie (US/Canada) (NOAA, 2014) highlights the controversy over whether controls on P, or dual control of P and nitrogen are needed. Reductions in P-loading to Erie from 1970 to 2000 substantially reduced algal blooms (International Joint Commission, 2014) but since 2000 blooms of toxic *Microcystis* have increased (Ho, Stumpf, Bridgeman, & Michalak, 2017). The increased bloom frequency from 2000 to 2013 is correlated with increased loading of total P (+25%) and soluble P (+50%). Nitrate and TN loading decreased 18% and 9%, respectively, but organic nitrogen loading increased 25% (Stow, Cha, Johnson, Confesor, & Richards, 2015). Management agencies and some scientists argue that "phosphorus is the key element limiting the amount of…algae in Lake Erie," and have pushed for stricter controls on P-loading (International Joint Commission, 2014; Michalak et al., 2013; Ohio EPA, 2013; Verhamme et al., 2016). In contrast, proponents of dual P and N control point to experimental evidence that *Microcystis* is N-limited (Chaffin, Davis, Smith, Baer, & Dick, 2018; Gobler et al., 2016) and that populations may shift from nontoxic to toxic strains with increasing NO_3 concentrations (Berry et al. 2017). The inability of these cyanobacterial genera to fix N_2, and their affinity for NH_4^+ and urea, suggests that N delivered through agricultural runoff and internal release of NH_4^+ may play critical roles controlling these blooms. Dual-control proponents also argue that "legacy loading" of P from the sediments makes it difficult to reduce P sufficiently to induce strict P-limitation (McCrackin, Jones, Jones, & Moreno-Mateos, 2017; Shatwell & Köhler, 2018). Others have stressed the importance of long residence times in Western Lake Erie and wind-mixing of sediments as additional factors driving the increased blooms (Sayers, Fahnenstiel, Shuchman, & Whitley, 2016). The complexity of driving factors and the entrenched positions of some of the scientists involved could delay resolution of this issue for some time.

(Hellström, 1996; Paerl & Scott, 2010; Seitzinger, 1988). The relationship between P and N is U.S. lakes (Figure 9), suggests that denitrification may outweigh N in eutrophic and hypereutrophic lakes where bottom water anoxia is expected, thus promoting denitrification. Second, factors other than P, such as iron, light, salinity, sulfate, turbulence and oxygen supersaturation may limit N fixation (Lewis & Wurtsbaugh, 2008; Marcarelli et al., 2006; Paerl, 1990; Stal, Staal, & Villbrandt, 1999; Wurtsbaugh & Horne, 1983; Wurtsbaugh, Vincent, Tapia, Vincent, & Richerson, 1985). Long-term results from Lake 227 in the ELA, suggest that in some situations N fixation may compensate sufficiently to balance P inputs and maintain a lake in a eutrophic state (Higgins et al., 2018; Schindler, 2012). However, the results of this ongoing experiment are controversial (Paerl & Scott, 2010; Scott & McCarthy, 2010) because the lake was originally fertilized with high levels of both N and P, and even though N fertilization ceased 25 years ago, water column N continues to decline significantly (Higgins et al., 2018). Additionally, Lake 227 is unusual in that it is currently fishless, but when piscivorous fish were added for 3 years, algal levels dropped nearly 50% and nitrogen-fixing cyanobacteria disappeared (Elser et al., 2000; Paterson, Schindler, Hecky, Findlay, & Rondeau, 2011). Nevertheless, this ambitious long-term experiment demonstrates that N fixation can contribute substantially to eutrophication. Given the relatively modest number of whole-lake experiments over limited geographic areas, more research is needed in a variety of more representative lakes on factors limiting N fixation, and on the balance between fixation and denitrification (Scott et al., 2019).

FIGURE 12 MODIS satellite image of a harmful algal bloom in Lake Erie on September 29, 2014. Data from NASA's Aqua satellite. https://www.noaa.gov/stories2015/images/aqua.2014272.0929.1845C.L3.LE3.v670.truecolor_logos.png

Nitrate and ammonia additions can be used to suppress N-fixing cyanobacteria (Elmgren & Larsson, 2001; Leonardson & Ripl, 1980; Wurtsbaugh & Horne, 1983), but these additions can also induce shifts to the toxic, nonfixing cyanobacteria *Microcystis* (Barica, Kling, & Gibson, 1980) and can and further exacerbate eutrophication. Additionally, non-N-fixing cyanobacteria, and in particular toxic *Microcystis* and *Planktothrix*, frequently proliferate in lakes when N levels are high (e.g., Paerl et al., 2011). Many lakes (e.g., Lakes Balaton, Zurich, Ontario, Taihu) are now severely impacted by toxic, non-N-fixing cyanobacteria blooms, even though P loading has declined (EPA Science Advisory Board, 2015; Schindler, 2012).

8.1 | Managing nutrients in complex and linked stream-lake-marine systems

Results from eutrophication remediation efforts indicate that recovery of lakes and coastal oceans is slow, and that full recovery is difficult to obtain. A meta-analysis of 89 recovery efforts indicated that after nutrient inputs from polluting sources ceased, investigators found that after a mean of 13 years, response variables had recovered, on average, only 34% of baseline conditions (McCrackin et al., 2017). Control efforts have focused on dual management of N and P (64%), P alone (33%), and only 3% on N alone (McCrackin et al., 2017). The review found that water-column N concentrations responded more quickly to nutrient reductions than did P because denitrification removes N, whereas internal recycling of legacy P from the sediments can slow recovery, despite external reductions (Sondergaard, Bjerring, & Jeppesen, 2013; Vahtera et al., 2007).

Historically, reducing P loading has been a cost-efficient means of controlling lake eutrophication, and this approach has been widely implemented and beneficial (e.g., Schindler, 2012; Schindler et al., 2016). However, in other systems, such as estuaries, marine coastal zones, and lakes afflicted with non-N-fixing cyanobacteria blooms, controlling both N and P inputs could be warranted (e.g., Boesch, 2019; Conley et al., 2009; Glibert & Burford, 2017; Paerl, 2009; Wurch et al., 2019). In their review of 89 control efforts McCrackin et al. (2017) did not find any significant differences in success from efforts to control P, N, or N + P, but variability was high, and few management programs have attempted to control N alone. Nevertheless, dual N and P control is beginning to be implemented with some success, either in experimental situations, or in entire systems (Baker et al., 2018; Elmgren & Larsson, 2001; McCrackin et al., 2017; Shatwell & Köhler, 2018). Agencies in the U.S. and Europe are drafting regulations to enforce dual nutrient control (EPA, 2015b; Ferreira et al., 2010). In China, recent work has also called for the control of excessive N discharges to freshwaters (Yu et al., 2019).

Some systems oscillate between P and N limitation of algal growth and present a more complicated management problem. For example, in hypereutrophic Lake Taihu in China, winter and spring algal populations are limited primarily by P, but the combination of high N uptake rates by cyanobacteria, denitrification, and sediment P releases during the summer and fall period lead to N limitation (Paerl et al., 2011). The dominant cyanobacterial bloom-former in this system, *Microcystis,* cannot fix N, and hence relies on combined N sources (NH_4^+, NO_3^-, urea), the availability of which controls its production. Similarly, nuisance algae in streams (*Cladophora* spp.) can be P limited early in the year but shift to N limitation in fall and late summer as internal stores of P decrease (Lohman & Priscu, 1992). In such situations, dual N and P input control will lead to most rapid bloom control and water quality improvement (Paerl et al., 2011).

Managing linked systems across the freshwater to marine continuum is complicated, because different parts of the continuum may have different nutrient limitation characteristics (Figure 13). For example, the estuaries of the Baltic Sea and Great Salt Lake are primarily P limited, but in open areas, where salinity is higher, they are N limited (Elmgren & Larsson, 2001; Wurtsbaugh, 1988). Even within an estuary such as the Chesapeake Bay, P may be limiting in the upper reaches, whereas N is limiting in the lower areas, and this pattern can vary seasonally with the ebb and flow of runoff (Kemp et al., 2005). Similarly, while many freshwater lakes are P limited, rivers, downstream estuaries and coastal areas often are N limited. Consequently, P control may be effective in many lakes, but still allow N export to downstream ecosystems. Controlling P can exacerbate N export to N-limited downstream systems because it reduces the uptake, sedimentation, denitrification and burial of N in upstream lakes and rivers (Elmgren & Larsson, 2001; Finlay, Small, & Sterner, 2013; Paerl, 2009; Ratmaya et al., 2019). Graneli, Wallstrom, Larsson, Graneli, and Elmgren (1990) reached a similar conclusion regarding the movements of N and P through bays and into the open area of the Baltic Sea. Consequently, when dealing with linked systems with different limiting nutrients, dual control has been recommended (Cole & Fulweiler, 2013).

Despite claims that P removal is less expensive than N removal (e.g., Schindler et al., 2008), there are very few data to verify these claims. The United States Environmental Protection Agency (EPA, 2015a) has recently estimated costs associated with eutrophication control. While banning phosphate-containing detergents is relatively inexpensive, much P enters wastewater streams from urban areas even when these detergents are banned. Thus, chemical removal is required. The operating costs for N removal from waste water treatment plants ranged from \$0.47 to \$8.40 m^{-3} to reduce levels to 0.60–1.4 mg N L^{-1}, and for P removal costs ranged from \$0.10 to \$27.70 m^{-3} to reduce levels to <1 mg P L^{-1}. Further, upgrading or construction

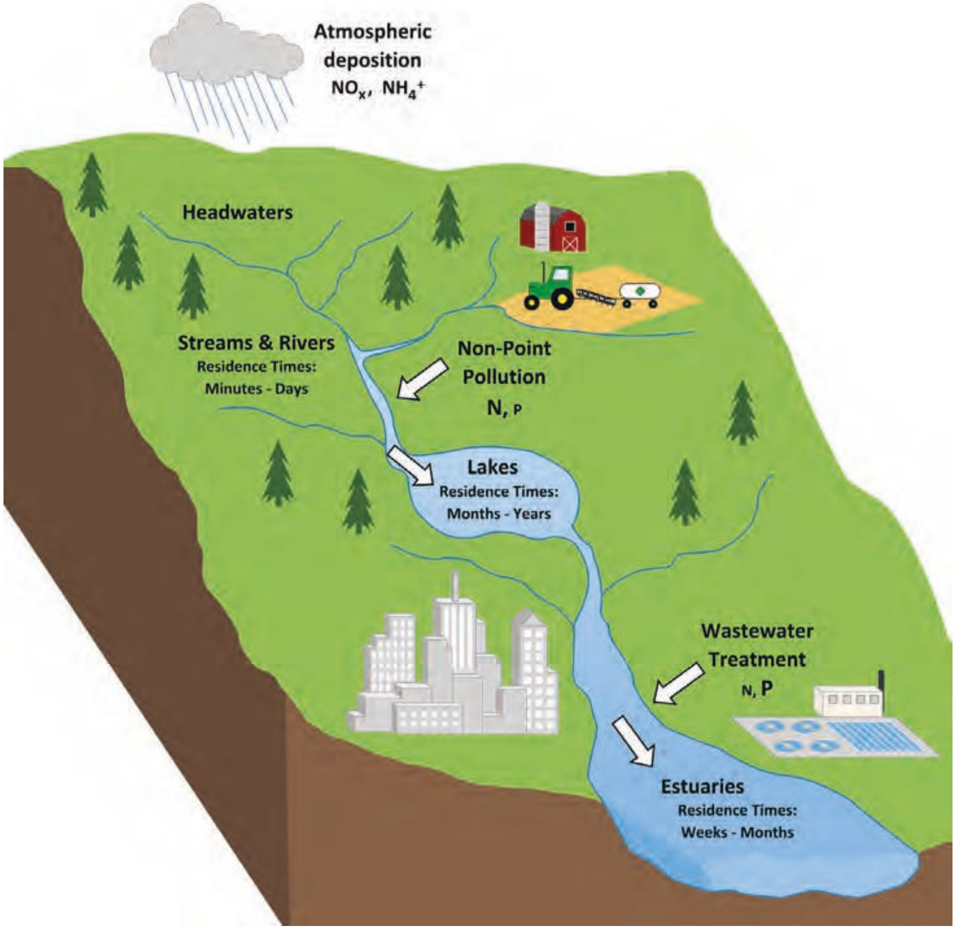


FIGURE 13 A hypothetical watershed showing the linkages among streams, lakes and coastal zones. Pollution sources for nitrogen and phosphorus, and approximate water retention times that influence eutrophication are shown. Nitrogen fixed by cyanobacteria can accumulate in systems with long residence times but will be flushed out in systems with short retention times. Movement of these two nutrients through complex watersheds and into marine systems complicates nutrient control strategies to reduce eutrophication

costs for facilities did not differ substantially for N or P control. Costs of nonpoint source control in urban areas were variable with no clear monetary savings for N versus P control. These data for urban removal also cannot account for the fact that costs for treatment facilities can decrease over time as technology improves and demand increases for the technology. Additionally, both advanced biological nutrient removal (BNR) and wastewater reuse, which are increasingly used, reduce both N and P simultaneously, thus negating the need to choose between one nutrient or the other (Carey & Migliaccio, 2009). We are not aware of analyses of relative costs of N or P control on agricultural lands; however, managers are now encouraging the 4R strategy—right form, right amount, right place, right time for fertilization with both N and P to maximize crop yields and reduce losses of both N and P to streams (e.g., Minnesota Pollution Control Agency, 2014). Maintenance of riparian buffer zones or the use of winter cover crops decreases sediment outputs and helps reduce N and P loading to streams and rivers. In general, for croplands, management strategies for one nutrient tend to also decrease the other.

9 | THE FUTURE

Although we understand many of the factors driving eutrophication, as well as control measures that can mitigate these problems, important scientific questions remain to be solved to improve efficacy of management, including:

- Where can eutrophication be controlled by P-only reductions, and where is dual control of P and N needed? Answering this question will require a better understanding of how the complexities of the N cycle influence the balance of N versus P limitation along the freshwater to marine continuum. For example, can general principles be found to predict under what circumstances the opposing effects of N-fixation and denitrification (Figure 8) lead to N-limited ecosystems (Scott et al., 2019)? Are long residence times in lakes and estuaries necessary for N-fixing cyanobacteria to accumulate sufficient N to balance P availability (Higgins et al., 2018)? Management of P or dual control with N included is also complicated by the linkage of freshwater and marine ecosystems, each of which may be limited by different nutrients (Paerl et al., 2018). Additional complications include the high cost of dual nutrient reductions, the difficulties of controlling both N and P export from agriculture, and the legacy effects of high P loading and concomitant internal loading that may limit the success of P-only control.

- What are the economic aspects of reducing N and P loading to linked ecosystems along the freshwater to marine continuum? Controls of both P and N from wastewater treatment plants have become increasingly efficient and more economical. However, nonpoint source controls, lacking stringent enforcement options, have been less effective and remain the biggest challenge for controlling eutrophication (Carpenter et al., 1998). Fortunately, both N and P are frequently reduced with nonpoint control strategies such as riparian buffers and no-till agriculture.
- How does N and P loading influence the breakdown of organic matter? Most lakes, streams and rivers receive large amounts of exogenous organic matter, either in dissolved form, or from leaves and other detrital matter from riparian area, and these are extremely important for ecosystem function. Despite this recognition, research is just beginning on how nutrients effect the heterotrophic breakdown of these exogenous carbon sources, and how this translates to effects on food webs and biodiversity (Dodds & Cole, 2007; Rosemond et al., 2015b).
- How does eutrophication and the proliferation of phytoplankton in the water column influence the production of primary producers in the benthic communities of rivers, lakes and estuaries? Eutrophication and subsequent shading by phytoplankton blooms reduces benthic algal production in lakes (Vadeboncoeur et al., 2002) and estuaries (Davidson et al., 2014), but we have limited understanding of this process in large rivers.
- What role does periphyton play in eutrophication? The growth of periphyton and phytoplankton in lakes may be limited by different nutrients (Elser et al., 2007), yet periphyton have been largely ignored in studies of eutrophication in lakes, even though they often dominate primary production (Brothers, Vadeboncoeur, & Sibley, 2016). We also need a better understanding of how benthic cyanobacteria and other N fixers influence the eutrophication process (Diehl et al., 2018), since preliminary observations suggest they may contribute to reducing N deficits in N-limited oligotrophic lakes (Holmgren, 1984; Schindler, 2012).
- How do species interactions influence eutrophication? Although there is considerable research on increases in N-fixing cyanobacteria when N becomes limiting (e.g., Paterson et al., 2011), we know considerably less about interactions among other taxa and how they respond to different chemical forms of N and P (e.g., Glibert et al., 2016). Microbiome research methods should facilitate this research direction.

10 | CONCLUSION

As we develop a more holistic view of aquatic ecosystems, it becomes clear that nutrient enrichment can have broad effects. Harmful or excessive algal blooms and dead zones in lakes and coastal areas are among the most conspicuous and well-studied, but the impacts of nutrient additions extend beyond these problems and influence biotic processes ranging from individual organisms to ecosystems. Several effects of eutrophication are just beginning to be understood, and the importance of others has not received adequate attention. For example, the food web implications of the shift between benthic primary production in un-polluted situations to a dominance of phytoplankton production in eutrophicated systems are just beginning to be appreciated.

Additional stressors are likely to increase eutrophication problems. Global climate change will increase temperature and lower pHs, tipping the balance toward toxic marine and freshwater microbes, lending further urgency to nutrient control attempts (Griffith & Gobler, 2019; Paerl, 2016). As the human population grows, more food will be needed to feed the global population and more intensive agriculture will be used to provide that food, leading to more nutrient loading of aquatic ecosystems (Sutton et al., 2013; van Puijenbroek et al., 2018). In arid regions, increasing agricultural demand for water will decrease runoff, but not necessarily nutrients. Streamflow in many populated arid urban areas will be dominated by wastewater. Wastewater and runoff from impervious surfaces will increase nutrient loading and atmospheric deposition will increase with increasing rates of fossil fuel combustion in urbanizing areas. Consequently, more effective environmental regulation will be required to curb nutrient loading from point-, and especially nonpoint sources in both developed and particularly developing nations.

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CONFLICT OF INTEREST

The authors have declared no conflicts of interest for this article.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of this article.

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