

Mitigating the Expansion of Harmful Algal Blooms Across the Freshwater-to-Marine Continuum

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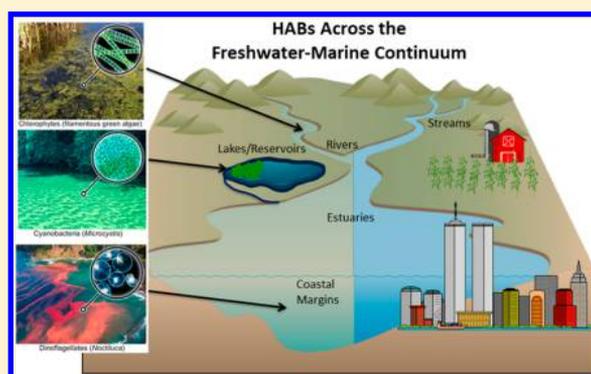
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ABSTRACT: Anthropogenic nutrient overenrichment, coupled with rising temperatures, and an increasing frequency of extreme hydrologic events (storms and droughts) are accelerating eutrophication and promoting the expansion of harmful algal blooms (HABs) across the freshwater-to-marine continuum. All HABs—with a focus here on cyanobacterial blooms—pose serious consequences for water supplies, fisheries, recreational uses, tourism, and property values. As nutrient loads grow in watersheds, they begin to compound the effects of legacy stores. This has led to a paradigm shift in our understanding of how nutrients control eutrophication and blooms. Phosphorus (P) reductions have been traditionally prescribed exclusively for freshwater systems, while nitrogen (N) reductions were mainly stressed for brackish and coastal waters.

However, because most systems are hydrologically interconnected, single nutrient (e.g., P only) reductions upstream may not necessarily reduce HAB impacts downstream. Reducing both N and P inputs is the only viable nutrient management solution for long-term control of HABs along the continuum. This article highlights where paired physical, chemical, or biological controls may improve beneficial uses in the short term, and offers management strategies that should be enacted across watershed scales to combat the global expansion of HABs across geographically broad freshwater-to-marine continua.



INTRODUCTION

Phytoplankton are key primary producers (photoautotrophs) that support biogeochemical cycling, food web structure, and the sustainability of aquatic ecosystems spanning the continuum from upstream headwaters to the coastal ocean. Human population growth, along with its associated agricultural, urban and industrial development, have led to nutrient inputs exceeding levels needed to sustain adequate primary productivity along this continuum. The resulting nutrient overenrichment and excessive production of organic matter at the base of the food web, termed “cultural eutrophication”, has led to a suite of undesirable biogeochemical and ecological consequences—the most obvious and troublesome being the overgrowth of noxious phytoplankton (i.e., “blooms”) (Figures 1 and 2).

The connection between excess nutrient inputs and HABs has been broadly recognized in European and North American waters impacted by large-scale agriculture, industrialization, and urbanization. This troubling trend is now rapidly expanding in developing regions of Asia, Central and South America, Africa, Australia–New Zealand, and the Pacific Basin.^{1–4} Impacts vary depending on the relationship of drainage basin and airshed areas to the size and volume of receiving waters.⁵ Most obvious are relatively small water bodies in urban, industrial, and

agricultural watersheds. However, large lakes, estuaries, and coastal waters are also impacted by accelerating eutrophication, expanding HABs and their toxins. Human hydrologic modifications, including water withdrawals, diversions and dams, have additionally altered the flow paths of water and nutrients entering aquatic ecosystems.

Climate change poses an additional challenge in predicting changes in HAB frequency, intensity, and proliferation.^{6–9} Global warming, altered precipitation patterns, and sea level rise, accompanied by changes in ocean and lake circulation, stratification, and upwelling, wind speed, and cyclone frequency and intensity, play increasingly important roles in modulating HAB dynamics.¹⁰ Hydrologic modifications and climate change enable HABs to reach larger magnitudes and persist longer when accompanied by excessive nutrient loading.^{3,11}

Contemporary HAB problems must be addressed on land–water–air interactive scales that link watersheds to the coastal ocean, since it is increasingly recognized that in order to combat these threats, we need to treat the freshwater-to-marine continuum as one interconnected system rather than a series of individual water bodies and management districts.

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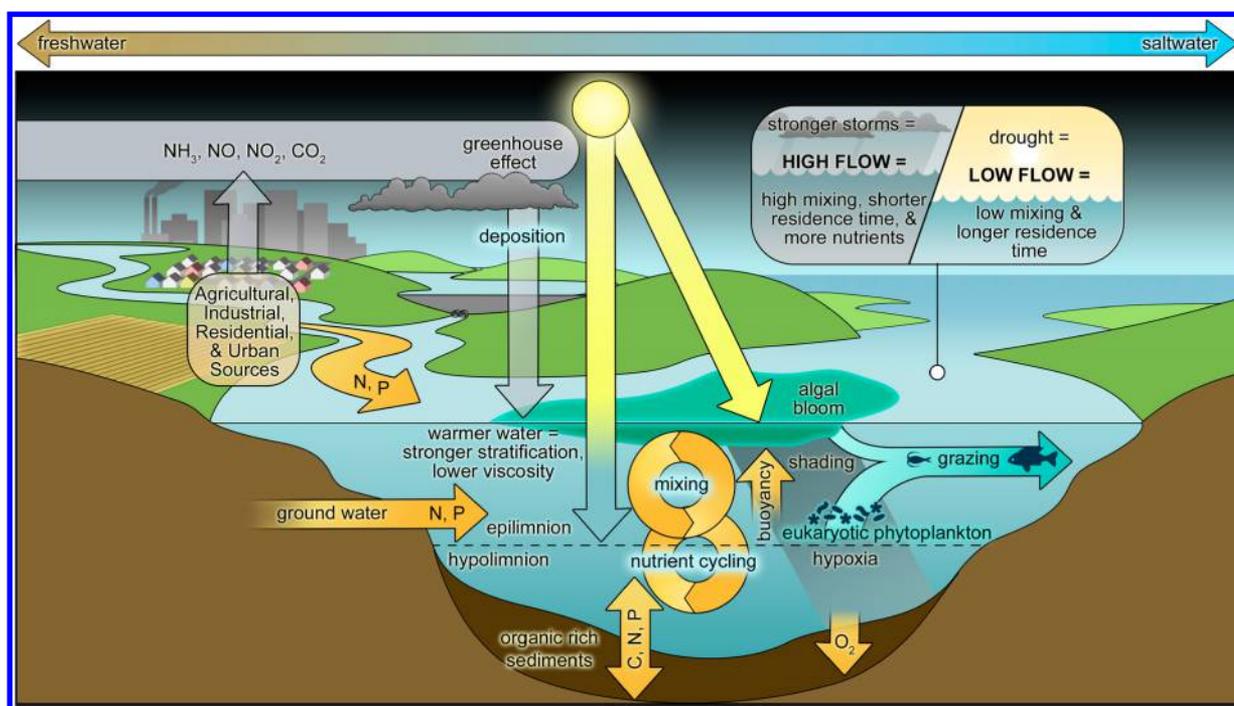


Figure 1. Conceptual diagram of interactive physical, chemical and biological controls on harmful algal bloom formation and proliferation along the freshwater-to-marine continuum.

Here, we discuss the evolving knowledge of factors controlling HAB development and persistence, and offer insights into management strategies that can be implemented across the freshwater-to-marine continuum in geographically diverse regions.

■ THE FRESHWATER–MARINE CONTINUUM

Two key takeaways of this article are first, to reinforce how management principles can and should be applied across watershed gradients, and second, to recognize that seemingly simple problems (blooms) and solutions (local control of N or P) are modulated by sometimes complex interactions with the environment. It is common knowledge that aquatic ecosystems are interconnected; they span from inland mountain streams, down to rivers, lakes and reservoirs, estuaries, and coastal margins. However, most management responses to eutrophication issues involve interventions aimed at treating individual segments of the freshwater-marine continuum and frequently focus on the problem (blooms) rather than the underlying causes that often take place upstream of the bloom itself.

Unfortunately, it is the hydrologic interconnectedness that makes management outcomes not a black-and-white situation. For example, in Upper Klamath Lake (OR, USA) dense blooms of the N_2 -fixing cyanobacterium *Aphanizomenon flos-aquae* proliferate for much of the year and are even harvested and sold as nutritional supplements. However, a sizable portion of these cells gets exported down the Klamath River where they are lysed during transit through a series of hydroelectric dams. These organically enriched waters flow into two large, stratifying reservoirs (Copco and Iron Gate) located approximately 120 km downstream from the lake, where large blooms of toxin-producing *Microcystis* sp. (a non- N_2 -fixer) persist.¹² The *Microcystis* sp. blooms and its toxins (microcystins) are subsequently exported another 300 km down to the Pacific Ocean, impairing fisheries and water quality all along the way. What is the solution? Some may argue that the *A. flos-aquae*

blooms—that ultimately sustain the downstream *Microcystis* sp. blooms—are a natural part of the system due to their ability to fix atmospheric nitrogen and the high concentration of naturally occurring geogenic phosphorus. However, investigations of akinetes from lake water and sediment samples clearly show that there has been a substantial increase in *A. flos-aquae* since the late 1800s, corresponding to when the basin was settled by pioneers.¹³

This observation provides a strong indication that human activities have enabled these blooms to become more prolific. The lessons are that (1) cultural eutrophication via habitat alteration (e.g., wetland removal and deforestation) and increased nutrient loads (both N & P) entering into Upper Klamath Lake have turned a naturally eutrophic system into a hypertrophic one; (2) it makes little sense to attempt nutrient control in the downstream reservoirs without first addressing the predominant source coming from the headwaters; and (3) watersheds are connected and should be managed across local-to-regional scales. Presently, the situation along the Klamath River remains unresolved; however, a broad coalition is now actively trying to solve it by reducing nutrient inputs, restoring wetlands and by moving forward on a series of dam removals that will be the largest such project in U.S. history. Unfortunately, this situation is often repeated elsewhere since most watersheds are experiencing accelerating anthropogenic impacts and most of these impaired rivers around the world have been dammed, thus the situation along the Klamath River is far from unique.

In the following sections we will envision following a parcel of water originating from a headwater source as it moves down through higher order streams and rivers, through and out a lake or reservoir, before moving down an estuary and to the coast. Throughout the freshwater-to-marine continuum the parcel of water will traverse a variety of physicochemical gradients that strongly influence biogeochemical cycles and phytoplankton growth potentials. Our primary focus as we move through the



Figure 2. Harmful algal bloom species representative of major taxonomic algal groups along the freshwater-to-marine continuum.

watershed will be to emphasize the key conditions that most strongly stimulate the overgrowth of harmful algal bloom taxa. Within each habitat we will highlight some of the management and mitigation techniques that have been successfully applied to reduce HAB impacts and to improve water quality. Our intent is not to provide an exhaustive review of all management strategies possible for a given ecosystem, but instead to challenge the reader to view eutrophication problems as watershed problems that are driven by a variety of interconnected factors that need to be thoughtfully addressed at both local and regional spatial scales. One commonality stressed throughout will be the importance of dual N and P reduction strategies. While this is well described in the literature,^{14–17} it is still often ignored, as documented in the following sections. Working at larger scales will require coalition building and creative thinking in order to develop mitigation strategies that function across management boundaries and jurisdictions.

Because most of the geographic distance that the hypothetical parcel of water will travel takes place in freshwater, where cyanobacteria are the dominant bloom formers (CyanoHABs), this group will be prominently discussed in relation to its environmental drivers and controls. This is not to

say that overgrowths of other phytoplankton groups cannot impose water quality impairment issues (e.g., golden algae and dinoflagellates that may cause fish kills), but broadly speaking cyanobacteria are a globally widespread problem in many watersheds. We also emphasize that common mitigation strategies are called out for specific regions along the water parcel's transit, but that most mitigation strategies can be applied to some degree across the entire freshwater-marine continuum.

Since they are so widespread and problematic, an overview of key characteristics pertaining to the phylum Cyanobacteria will first be introduced to the reader. Cyanobacteria are true bacteria, exhibiting prokaryotic features and behaviors. They exist primarily in two morphologically distinct groups broadly characterized as coccoidal or filamentous (Figure 3). Coccoid

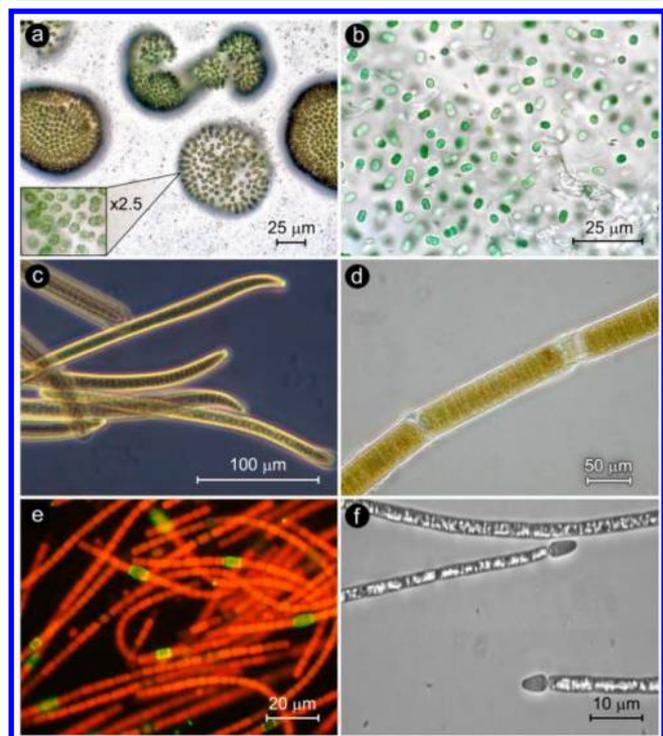


Figure 3. Photomicrographs of representative harmful cyanobacterial (CyanoHAB) genera found along the freshwater to marine continuum. a and b show colonial and solitary coccoid forms of *Microcystis* and *Synechococcus*, respectively; c and d show the filamentous, non-diazotrophic genera, *Oscillatoria* and *Lyngbya*, respectively; e and f show the filamentous diazotrophic (heterocystous) genera, *Dolichospermum* and *Cylindrospermopsis*, respectively.

cyanobacteria often aggregate into colonies that can accumulate at the water's surface as highly visible scums (e.g., *Microcystis*), but they may also occur as single-cells (e.g., *Cyanobium* or *Synechococcus*) that are dispersed throughout the photic zone of the water column. Most types cannot utilize atmospheric nitrogen (N_2), and hence are dependent on organic and inorganic nitrogen supplied to or recycled within the water column. Free-living picoplanktonic ($<2 \mu\text{m}$ diameter) coccoid genera make up an important, ubiquitous, and at times dominant ($>50\%$) fraction of phytoplankton biomass along the continuum.^{18–21} Filamentous cyanobacteria are comprised of either undifferentiated, mostly non- N_2 fixing genera (e.g., *Lyngbya*, *Oscillatoria*, *Planktothrix*, and *Phormidium*) or N_2 -fixing genera (diazotrophs) which possess biochemically

Table 1. Common Harmful Bloom-Forming Phytoplankton Genera Observed Across the Freshwater-Marine Continuum and Their Known Toxins^a

genus	group	potential toxin(s)	characteristics	salinity range		
				low (0–4)	mod. (4–16)	high (16+)
<i>Anabaena</i>	cyanobacteria	ATX, CYN, MC, STX	B,D,F	×	×	
<i>Anabaenopsis</i>	cyanobacteria	MC	P,D,F	×	×	×
<i>Aphanizomenon</i>	cyanobacteria	ATX, CYN, STX	P,D,F	×	×	
<i>Cylindrospermopsis</i>	cyanobacteria	ATX, CYN, STX	P,D,F	×		
<i>Cylindrospermum</i>	cyanobacteria	ATX, MC	B,D,F	×		
<i>Dolichospermum</i>	cyanobacteria	ATX, CYN, MC, STX	P,D,F	×	×	
<i>Fischerella</i>	cyanobacteria	MC	B,D,F	×	×	×
<i>Haplosiphon</i>	cyanobacteria	MC	B,D,F	×		
<i>Lyngbya</i>	cyanobacteria	CYN, LYN, STX	B,F	×	×	×
<i>Microcystis</i>	cyanobacteria	MC	P,C	×		
<i>Nodularia</i>	cyanobacteria	NOD	B/P,D,F	×	×	×
<i>Nostoc</i>	cyanobacteria	ATX, MC	B,D,F	×	×	
<i>Oscillatoria</i>	cyanobacteria	ATX, CYN, MC, STX	B/P,D,F	×	×	×
<i>Phormidium</i>	cyanobacteria	ATX, MC	B,F	×	×	×
<i>Planktothrix</i>	cyanobacteria	ATX, MC	P,F	×	×	
<i>Raphidiopsis</i>	cyanobacteria	ATX, CYN, MC	P,F	×	×	
<i>Scytonema</i>	cyanobacteria	MC, STX	B,D,F	×	×	×
<i>Umezakia</i>	cyanobacteria	CYN, MC	P,D,F	×		
<i>Pseudonitzschia</i>	diatom	DA	P		×	×
<i>Alexandrium</i>	dinoflagellate	STX	P		×	×
<i>Gymnodinium</i>	dinoflagellate	STX	P			×
<i>Karenia</i>	dinoflagellate	BRV	P			×
<i>Karlodinium</i>	dinoflagellate	ICX	P	×	×	×
<i>Pyrodinium</i>	dinoflagellate	STX	P		×	×
<i>Prymnesium</i>	haptophyte	ICX	P	×	×	×

^aToxin abbreviations: ATX = anatoxin-a; BRV = brevetoxin; CYN = cylindrospermopsin; DA = domoic acid; ICX = ichthyotoxins; LYN = lyngbyatoxin; MC = microcystin; NOD = nodularin; STX = saxitoxin. Characteristics abbreviations: B = benthic; C = coccoid; D = diazotrophic; F = filamentous; P = planktonic.

specialized cells called heterocysts where N₂ fixation is localized and vegetative cyst-like cells called akinetes. Diazotrophic cyanobacteria comprise many commonly observed bloom-forming genera, including: *Aphanizomenon*, *Cylindrospermopsis*, *Dolichospermum*, and *Nodularia*. Filamentous cyanobacteria may exhibit planktonic or benthic lifestyles and are problematic from a risk characterization standpoint, because they contain species capable of producing multiple classes of toxins^{22–24} (Table 1).

Cyanobacterial blooms pose a direct health threat via food, water, and recreational exposures and cause ancillary economic impacts to tourism and real estate. From an ecological perspective, CyanoHABs can lead to undesirable biogeochemical changes, including large accumulations of biomass that create hypoxic zones as they decay, often resulting in fish kills and the release of toxic gases such as hydrogen sulfide, ammonia and methane from the sediments.⁵ Their poor nutritional value, large colony sizes, and prolific production of toxic secondary metabolites all negatively impact food webs, inducing cascading effects on biodiversity, fisheries production, and habitat.^{25–27} Blooms may also produce a variety of earthy and/or musty taste-and-odor compounds (e.g., geosmin and 2-methylisoborneol) that render waters unpalatable for drinking. Lastly, numerous CyanoHAB species produce potent toxins that impact the liver, digestive, and nervous systems of animals and humans that ingest contaminated waters^{22–24} (Table 1).

■ HEADWATER STREAMS

Streams originating from groundwater springs or alpine snowmelt tend to be narrow, shallow, and relatively oligotrophic. First-to-third order streams derive the majority of their matter and energy via the microbial degradation of allochthonous matter. These streams are more often limited by nitrogen than phosphorus bioavailability, due to relatively high naturally occurring (geogenic) phosphorus present in many source waters.²⁸ Because of the shallow depth and flow, most of the algal community is comprised of benthic attached diatoms and chlorophytes, with cyanobacteria generally comprising a low percentage of total biomass.²⁹ Some of the potentially toxic cyanobacterial genera that may be found in these streams include the N₂-fixers *Nostoc* and *Anabaena* and the non-diazotrophs *Phormidium* and *Oscillatoria*.

Because headwater streams are often remote from major anthropogenic activities, the impacts of cultural eutrophication, aside from atmospheric nitrogen deposition, are generally minimal. As such, with no nutrient point-sources to be controlled, there are limited management options available to improve water quality in these streams. These systems are often fast flowing (i.e., high flushing rates and short water residence times), which will minimize the opportunity for cyanobacteria to develop planktonic blooms. Instead, if adequate light is available, attached benthic algal communities tend to be the dominant primary producers.

A survey of over 1200 wadeable stream segments in California revealed the widespread occurrence of potentially toxic benthic cyanobacteria, with *Leptolyngbya*, *Anabaena*,

Nostoc, and *Phormidium* being especially prevalent genera,²⁴ and with cyanotoxins found in greater than one-third of sites. Benthic *Anabaena* from the Eel River (California) may produce microcystin or anatoxin-a.³⁰ In New Zealand, benthic *Phormidium* mats often exceed 20% coverage of the river bed,³¹ and contain exceptionally high anatoxin-a concentrations—up to 712 mg/kg dry weight,³² vastly exceeding the acute oral lethal dose (LD₅₀) of anatoxin-a in mice of ~16 mg/kg.³² In brackish lakes, rivers and coastal lagoons benthic *Nodularia* mats are often observed,³³ producing high concentrations of the hepatotoxin nodularin, a structural analog of microcystin.

Because upland streams are frequently N-limited,³⁴ N₂-fixing cyanobacterial genera (e.g., *Anabaena*, *Nostoc*, *Scytonema*) can be a significant component and source of biologically available N in the benthic microalgal community. These species can also produce secondary metabolites that may be toxic to animal consumers, including humans.²²

■ RIVERINE SYSTEMS

Headwater streams feed into progressively larger streams that eventually become rivers. This hydrologic transition is often accompanied by a human-nutrient footprint in the form of increasing agricultural and urban development. As river size increases, so does the likelihood of anthropogenic impacts and utilization, including increased nutrient loads and altered hydrology, such as water withdrawal and diversions. Furthermore, increases in river size are generally accompanied by slower flows and hence longer water residence times. Combined, these conditions are more favorable for algal biomass accumulation and bloom formation. The typical progression is for blooms to form in backwaters, pools and rivers during slow-moving, relatively high temperature summer months, fueled by high nutrient laden “freshets” from spring rain runoff^{25,26} (<http://www.dispatch.com/article/20151003/NEWS/310039755>; <https://www.cincinnati.com/story/news/2015/09/18/know-mile-algal-bloom-ohio-river/72401378/>). Increasingly, nutrient-impacted river systems in agricultural and urban catchments (e.g. Klamath River, CA; Ohio and Maumee Rivers, OH; St. Johns River, FL; Sacramento and San Joaquin Rivers, CA; Yangtze River, China; Nile River, Egypt) are exhibiting protracted summer and fall algal blooms, often dominated by toxic cyanobacteria, and attributable to rising nutrient loads as well as rising temperatures, droughts, construction of dams, and diversions which tend to increase water residence times.³ Nutrient addition bioassays and stoichiometric analyses conducted on these systems reveal a complex picture that indicates there are times where P or N may individually stimulate blooms, but more frequently it is the combined effect of both N and P enrichment that sustain the largest and most persistent blooms.^{34,35}

Agriculture is recognized as a major source of riverine nutrient inputs. There are relatively simple, logical steps that can be taken to minimize these inputs. The 4R Nutrient Stewardship Strategy (right rate, right time, right place, and right source) has been advanced as a way to engage and teach farmers to apply fertilizers in a sustainable manner and is being used in the U.S. midwest in an effort to reduce CyanoHABs in Lake Erie.³⁶ Applying fertilizers at agronomic rates, formulated on amounts needed to optimize crop yield without saturating the soil is a first step, with obvious benefits for most ecosystems regardless of the location within the watershed. Given that we are facing an increased likelihood of more frequent extreme weather events (e.g., flooding),³⁷ appropriate timing of fertilizer

application is increasingly important. Nutrient runoff during the wet winter and spring months has been shown to be significantly constrained by planting cover crops (e.g., winter barley), and by planting riparian buffers that help to keep soil and nutrients in place.³⁸ Riparian buffers around agricultural lands ranging in width from several to hundreds of meters, have been shown to reduce edge of field N and P losses by as much as 50%.³⁹ Modifications to timing and cover crops is attractive economically in the face of increasing costs of N and P fertilizers. Lastly, recycling wastes from animal operations colocated with row crop, ornamentals, and silviculture operations creates a “closed loop” of fertilizer generation and use in modern agriculture that makes sense economically and ecologically.

Another beneficial, low-tech option is to install flashboard risers in drainage ditches that increase water residence time and enhance N processing (i.e., denitrification), thereby reducing N export to downstream waters. As with riparian buffers, constructed wetlands can serve to retain and process nutrients. There are technologies available to convert unused agricultural, rural, and urban lands to wetlands that can serve as treatment facilities for agricultural wastes and stormwater runoff.^{38,39}

■ LAKES AND RESERVOIRS

As receptacles for riverine systems, lakes and reservoirs are often the sites of the most intense and persistent HABs. In addition to receiving and sequestering upstream anthropogenic nutrient and sediment inputs, lakes and reservoirs have comparatively long water residence times and may exhibit periodic water column stratification. Both of these factors are most pronounced during summer periods, when elevated temperatures and maximum irradiance coincide, resulting in optimal conditions for CyanoHAB development.³ Persistent stratification promotes strong vertical biogeochemical gradients, and under highly productive HAB conditions, hypolimnetic bottom waters will exhibit excessive oxygen depletion, which will lead to nutrient release from sediments, promoting internal nutrient cycling and perpetuating bloom conditions. Most planktonic CyanoHAB genera are adept at taking advantage of these conditions because they are able to control their vertical position in the water column by adjusting their buoyancy through intracellular gas vesicle formation and collapse.²⁶ This enables cyanobacteria to access nutrients at depth at night, before moving back to the surface to resume photosynthesis.⁴⁰ The ability to form thick scums at the water's surface provides cyanobacteria with a competitive advantage by enabling them to shade out nonbuoyant algae and submerged aquatic vegetation.⁴¹ Some flagellated algae (e.g., dinoflagellates, cryptophytes) are also capable of rapid vertical movement in the water column,⁴⁰ but in general, in freshwater systems these taxa tend to be nontoxic and therefore pose less of risk than surface scum-forming CyanoHABs.

Once CyanoHABs become established in eutrophic lakes and reservoirs, they tend to exhibit annual, recurring successional patterns. Eventually, the cyanobacterial bloom will collapse due to either nutrient depletion or other unfavorable environmental conditions (e.g., sudden cold spells and mixing events following storms, low light conditions, bacterial or viral lysis), causing its stored nutrients to be released. These autochthonous nutrients can then be reutilized to promote subsequent CyanoHAB events (e.g., N₂-fixing *Aphanizomenon* blooms are often followed, either within a lake/reservoir or downstream, by *Microcystis* blooms).^{25,40} This is quite common during the

spring to fall period, when relatively long residence times and elevated temperatures favor CyanoHAB dominance and persistence.^{41,42} Usually this replacement cycle is broken during the fall to winter transition, when cooler temperatures and water column turnover negate the ecological advantage that CyanoHABs possess during warmer months.^{3,26}

Excessive P loading has traditionally been linked to lake and reservoir eutrophication^{44,45} and CyanoHAB formation.^{14,46} In certain cases, P reductions can be effective without parallel N removal because it may reduce total P availability enough to reduce the overgrowth of bloom taxa. Examples include Lake Washington, WA, where reduction of sewage-based P inputs led to the reversal of eutrophication,⁴⁷ and Himmerfjärden, Sweden, where reduction of wastewater, agricultural and industrial P discharges led to a decline in CyanoHABs.^{14,48} Similar P reductions have curtailed CyanoHAB bloom activities in large European and Asian lakes (e.g., Lakes Constance and Lucerne, Germany-Switzerland, Lago Maggiore, Italy, Lake Biwa Japan).¹⁴ Phosphorus input reductions, largely from improved wastewater treatment and a detergent ban, helped reduce eutrophication and CyanoHAB impacts in Lake Erie (U.S.) during the 1970s.^{44,45} However, the more recent resurgence of mostly non-N₂ fixing CyanoHABs in Lake Erie is thought to be the result of increasing nonpoint nutrient loads, legacy nutrients stored in its sediments and the widespread colonization of invasive dreissenid mussels that selectively graze other groups of phytoplankton over cyanobacteria.⁴⁹ Algal bioassay studies conducted in Western Lake Erie indicate that N-loading may specifically stimulate the growth of nondiazotrophic toxin-producers such as *Microcystis* sp. and *Planktothrix* sp.,¹⁷ and highlight the need to reduce both N and P inputs as a logical and tractable management step for mitigating blooms.¹⁴ Examples of nutrient colimitation in numerous other large lakes with a lengthy history of eutrophication, include Okeechobee (U.S.), Taihu (China), Kasumagaura (Japan), Balaton (Hungary), and Peipsi (Estonia).¹⁴

The historic primary focus on P control of freshwater eutrophication is based on the theory that N-loading reductions will not control blooms because N₂-fixing cyanobacteria will circumvent N-limitation by augmenting a water body's internal nitrogen pool.⁵⁰ However, recent studies have shown that even under highly eutrophic conditions, within-system N₂ fixation rates often fall far short of meeting ecosystem N requirements.^{14,51,52} This is because N₂ fixation is controlled by multiple environmental factors in addition to P availability, including: adequate sunlight to support this energy-demanding process in turbid eutrophic waters, limitation by iron and other essential cofactors, and supersaturated dissolved oxygen concentrations in blooms which are inhibitory to the N₂ fixation process.⁵³ At the ecosystem-scale, annual rates of denitrification often exceed rates of N₂ fixation,^{14,51} indicating that lakes tend to lose more N than they gain by diazotrophy. From a nutrient management perspective, this within-system N shortfall means that external N inputs play a critical role in supporting eutrophication and sustaining HABs.¹⁴

In conjunction with nutrient input reductions, there are several additional methods available to expedite the recovery of eutrophic lakes and reservoirs. In shallow water systems impacted by HABs, increasing the aerial cover of macrophytes can be an effective means of sequestering and processing nutrients. Technologies are now available for establishing macrophytes either as planted fields in littoral zones or on

floating rafts.⁵⁴ Removal of nutrients stored in the sediments by dredging has been utilized to reduce internal loading, but it needs to be combined with source control. A successful example is Lake Trummen, Sweden, a small (~1 km², mean depth 1.6 m) lake that experienced CyanoHAB water quality degradation in response to domestic sewage and industrial nutrient inputs during the mid-1900s that was successfully remediated using suction dredging of the upper half meter of sediments during a two year period.^{55,56} The Lake Trummen success can be attributed to its small, easily manipulated size, and the ability to simultaneously target reductions of external nutrient loads from its small (13 km²) watershed. In other sediment dredging efforts on sections of large lakes, results have been less successful when not paired with external loading controls.⁵⁷

As noted above, hydrologic conditions also play a key role in modulating and controlling HABs, especially CyanoHABs. Horizontal flushing, by increasing the water flow through water bodies, reduces water residence time, thereby providing less time for the development of relatively slow-growing cyanobacterial blooms.^{58,59} However, from a mitigation perspective, hydrologic modifications can be quite expensive, and as such, are generally restricted to relatively small water bodies. Additionally, many rivers that may be diverted to flush lakes can carry high nutrient loads that ultimately exacerbate the eutrophication problem in the lake. Nevertheless, it is important to recognize that factors such as depth and flushing can exert a major influence on the intensity of CyanoHABs. If the future portends prolonged droughts and short intense rain events, these factors could promote CyanoHAB events.^{9,11} Lastly, ultrasonic cell disruption techniques have been used in small impoundments to temporarily mitigate CyanoHABs,³ but this is a temporary measure which does not address the "nutrient issue" and can lead to release of toxins into the bulk water phase.

Alternative approaches may also include chemical treatments that precipitate P and help it to remain adsorbed in the sediments. A commercial treatment called "Phoslock" uses a bentonite clay infused with lanthanum that strongly binds to phosphate anions in the waterbody.⁶⁰ The bound phosphate and clay then settles out of the water column and the thin layer (~1 mm) of Phoslock on the sediment surface forms a barrier to phosphate diffusing out of the sediments. Phoslock has been applied in small lakes and reservoirs, where it can lead to P-limited conditions that can control CyanoHABs in some,⁶⁰ but not all situations.⁶¹ This approach is most likely to be effective in relatively small, deep lakes. Long-term success in controlling CyanoHABs is best assured by maintaining parallel reductions in external nutrient inputs. Another widely used method for removing algae from the euphotic zone and retarding growth is the application of flocculating agents such as clays^{62,63} or treatment with barley straw.⁶⁴ The basic principle in flocculation is that charge differences between the flocculating agent and the algal cell membranes causes adsorption via ionic bonding, allowing the cells to be settled out of the water column. While clay flocculation is best known for dispersal of marine dinoflagellates, the method works equally well in freshwater systems.⁶³ Chemical treatments may also include the application of barley straw and algacides, including formulations containing copper. While effective, copper is toxic to a wide variety of plant and animal species and its residue in the sediments is problematic as a legacy pollutant. Hydrogen peroxide has also been shown to be an effective algacide for

controlling CyanoHABs,⁶⁵ and it poses no serious long-term pollution problem since it dissociates into oxygen and water. Due to cost and application challenges, algacide treatments are generally restricted to small impoundments.

■ THE TRANSIT TO ESTUARINE AND COASTAL WATERS

As the hypothetical water parcel transits from lakes/reservoirs into estuarine and coastal waters, it continues to accumulate N and P from point and nonpoint sources that have been recycled and transformed into primarily oxidized inorganic forms of N (NO_2/NO_3) and P (PO_4) and organic N and P. Even with these elevated nutrient concentrations, CyanoHAB abundances tend to rapidly attenuate in riverine systems due to the short hydraulic residence times and their relatively slow growth rates. As in headwater streams, benthic/epiphytic eukaryotic algae and cyanobacteria (e.g., *Nostoc*, *Phormidium*) become more dominant than planktonic genera in flowing waters. However, planktonic blooms that originate in lakes and reservoirs may still be exported downstream and represent transient risks as they make their way through the watershed. Brackish water segments of estuaries that have experienced human nutrient enrichment can also sustain CyanoHABs. These include several of the U.S.'s largest estuarine ecosystems, for example, the Potomac River and James River tributaries of Chesapeake Bay, which have supported recurring blooms of both N_2 -fixing and non- N_2 -fixing CyanoHAB.⁶⁶ North Carolina's Albemarle-Pamlico Sound system has also been impacted by such blooms, and they are increasing in the brackish transition where the Chowan and Roanoke Rivers empty into the Albemarle Sound.⁶⁷ Florida's largest riverine-estuarine system, the St. John's River, the Indian River Lagoon (on the Atlantic side) and Sanibel Bay (Gulf of Mexico side) are regularly plagued by toxic CyanoHABs, including: *Microcystis*, *Dolichospermum*, *Lyngbya*, and *Cylindrospermopsis*. Brackish waters of the largest estuarine system on the U.S. west coast, the San Francisco Bay Delta, are now experiencing transient *Microcystis* blooms that originate in upland waters but are transported down through the lower estuary, with evidence suggesting that these blooms are exacerbated by both excessive nutrient loads and persistent droughts.^{68,69} Even where marine systems are not directly affected by actively growing algae, CyanoHAB contaminated freshwater discharge can lead to adverse ecological and health effects. Examples include sea otter poisoning events linked to microcystins originating in an inland lake that were exported and bioaccumulated in shellfish in Monterey Bay, CA.⁷⁰ In San Francisco Bay, freshwater and marine toxins are routinely found in shellfish, highlighting the importance of diffuse transport pathways.⁷¹

Large rivers almost always have significant human development in close proximity to river margins, resulting in both point (e.g., wastewater treatment discharges) and nonpoint (e.g., agricultural runoff) nutrient sources. As we move further from lakes and into lower rivers and estuaries, the paradigm that P regulation is the key tool for management and bloom mitigation has also been challenged.⁷² As with lakes, algal biomass is often decoupled from P loads due to mediating physical and biological factors. In marine waters, P is rarely the limiting nutrient, and a consensus exists for N being the dominant limiting nutrient in estuaries and the coastal ocean.^{73,74}

Major nutrient sources into estuarine and coastal waters include partially or entirely untreated urban and industrial

waste, septic systems, lawn and golf course fertilizers and wetland removal for urban development. Most wastewater treatment plants now effectively reduce P in their effluents; however, many lag with regard to N treatment—a critical need for N-sensitive estuarine and coastal waters. Treatment of inorganic nitrogen waste using biological nitrogen reduction (BNR) via microbially mediated nitrification-denitrification steps has been shown to be effective at reducing N from wastewater effluent. However, BNR costs are significant, therefore, societal economic priorities need to be established.

■ COASTAL MARGINS

As the hypothetical water parcel crosses the boundary from the terrestrial to marine environment, it is convenient but incorrect to assume that “dilution is the solution to pollution”. For example, upwelling dominated systems have generally been perceived to be less affected by anthropogenic nutrients due to the sheer magnitude of natural (upwelled) nutrients as well as the highly dynamic conditions making these systems potentially more resilient. However, a growing number of studies have suggested that our perception of the resilience of these systems may be flawed (c.f., ref 75). Within the Southern California Bight (CA), where point sources dominate nutrient inputs, 92% of total terrestrial N loading to coastal waters is from wastewater effluent discharged directly into coastal waters via outfall pipes.⁷⁶ The total load is approximately equivalent to N inputs from upwelling at spatial scales of tens of km, effectively doubling N loading to the shelf, and increasing the N:P ratio as relatively little P is discharged in wastewater effluent.⁷⁷ While Southern California is unusual in that it has low riverine discharge and a large anthropogenic footprint (wastewater from outfall pipes), this is not an isolated problem. N loading is projected to increase through climate-induced precipitation alone by 19% for the major river systems in the U.S., and precipitation changes are expected to exacerbate N loading even more in India, China, and Southeast Asia.⁷⁸ A changing climate will also alter atmospheric deposition of reactive N; for aquatic systems, the impacts are poorly resolved,⁷⁹ but numerous examples of HABs related to N-deposition have been documented.⁸⁰ Controlling atmospheric emissions, especially of N, should go hand-in-hand with watershed based nutrient reduction strategies along the continuum, as it has been shown that deposition of biologically reactive N compounds compose a significant fraction of “new” N inputs to waterbodies.^{80–82}

The form of N is also important. Modulation of toxicity in marine algae occurs in response to nutrient ratios, absolute concentrations and nutrient speciation. It has been demonstrated, for example, that ammonium leads to a greater enhancement of toxin content than nitrate in the marine dinoflagellate *Alexandrium*, a paralytic shellfish poison (PSP) producer.^{83–86} In *A. tamarense*, urea stimulates PSP toxin production relative to growth on nitrate, but with lower cell quotas than for cells grown on ammonium.⁸⁶ Growth on urea also enhances cellular domoic acid production compared to growth on nitrate or ammonium under comparable conditions in the marine diatom *Pseudonitzschia*. Thus, “anthropogenic” nutrients such as ammonium and urea may play disproportionate roles in driving toxicity of estuarine and marine HABs.

While coastal receiving waters are primarily limited by N, there is also evidence, similar to the terrestrial side of the continuum, that P cannot be ignored. For example, in the Pacific Northwest, coastal waters could easily be pushed into P

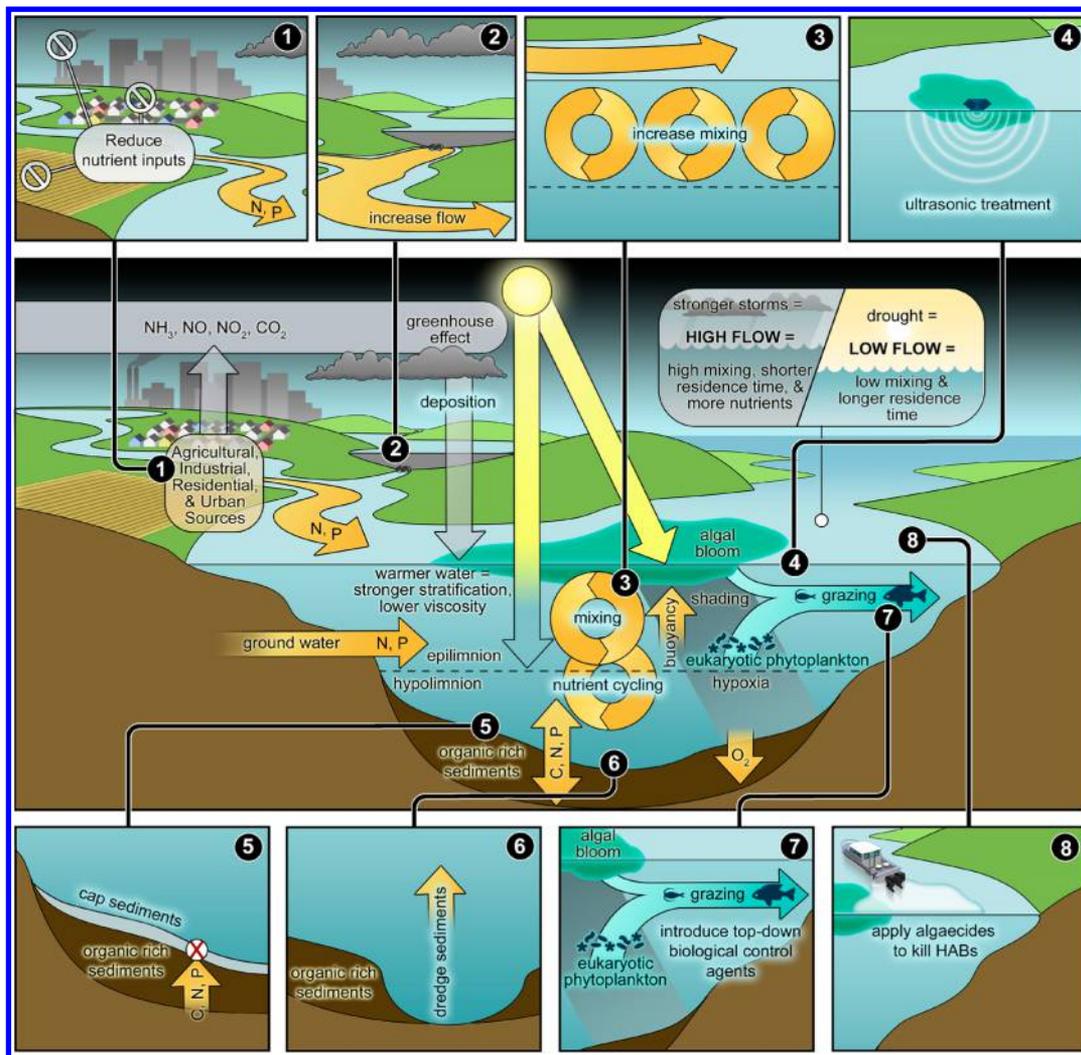


Figure 4. Summary of major approaches for mitigating HABs along the freshwater-to-marine continuum.

limitation⁸⁷ by changing the relative proportion of upwelled and riverine waters on the shelf. Similarly in the Gulf of Mexico, excessive N-loading from the Mississippi River may shift coastal waters to P-limitation, leading to an expanding zone of eutrophication as the excess N is transported further offshore.⁸⁸ Other examples include a shift from diatoms to dinoflagellates in Tolo Harbor, Hong Kong, in response to human-driven increases in P runoff, and increases in the toxic dinoflagellate *Karenia* off the Florida shelf attributed to low N:P ratios.⁷³

Management options for coastal receiving waters should focus primarily on reducing N and P in riverine and wastewater discharge. In regions of intense, localized blooms, clay flocculation can be effective in transporting bloom biomass to the sediments,⁶² but the only realistic, long-term solution to deal with the growing nutrient load from point and nonpoint sources is to reduce loads before they reach the coast.^{14,73,74}

CONCLUDING REMARKS

A summary of the various HAB mitigation strategies that have been applied along the continuum is presented in Figure 4. Since most freshwater systems drain to estuarine and coastal waters, there are multiple reasons for considering dual nutrient (N and P) input constraints for long-term control of HABs along this continuum.^{14–16,89} This is because even if upstream

primary production is controllable with P input reductions alone, the excess N that was unable to be utilized in the watershed will be transferred to downstream N-sensitive estuarine and coastal waters, allowing eutrophication to proceed.^{86,87} Therefore, upstream dual nutrient reduction is likely to have a positive controlling influence on HAB events and their cascading effects such as hypoxia in estuarine and coastal systems.^{14–16}

Changing threshold relationships between environmental drivers and metabolic growth responses of phytoplankton communities will strongly modulate HAB potential and persistence in response to climate change. For CyanoHABs, faster growth rates can be expected under warmer and climatically extreme conditions (i.e., both droughts and heavy rainfall events)^{6,10,90} and will be accompanied by increased nutrient demand, increased respiration and mineralization rates, which will lead to increased biological oxygen demand and hypoxia/anoxia potentials. Under this future climate scenario, stronger vertical stratification is likely to occur, which will further increase hypoxia/anoxia. In concert, these changing conditions will promote mobilization of P and N as well as most micronutrients from sediments, which will change the kinetic relationships between nutrient concentrations, supply rates and cyanobacterial growth rates, leading to greater growth and bloom potentials. This constitutes a positive feedback loop

between rising temperatures, increased CyanoHAB potentials, and the nutrients to sustain them.

In marine ecosystems, we expect a similarly complex response to climate change, but unlike freshwater systems, specific predictions are more difficult to make, in part because of the lack of studies focusing on multistressor responses.^{8,11} Despite this uncertainty, it is clear that the global increase in marine HABs is directly related to increasing nutrient loads.⁹¹ As with CyanoHABs, a first-order focus on reducing nutrient loads is critical.

Failure to act decisively now will exacerbate the already daunting challenges we face from global climate change scenarios. In an overwhelming number of cases, short-term mitigation approaches must be accompanied by permanent N and P input reductions as part of a comprehensive nutrient management strategy if we are to achieve a long-term, sustainable reduction of HABs on regional and global scales. It is important to recognize the linkages within watersheds and to understand how upstream activities have direct impacts on downstream water quality. As such, nutrient management strategies should target basin-wide water quality objectives and not treat each water body as a closed system.⁴³

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REFERENCES

- Halpern, B. S.; Walbridge, S.; Selkoe, K. A.; Kappel, C. V.; Micheli, F.; D'agrosa, C.; Bruno, J. F.; Casey, K. S.; Ebert, C.; Fox, H. E.; et al. A global map of human impact on marine ecosystems. *Science* **2008**, *319*, 948–952.
- Diaz, R. J.; Rosenberg, R. Spreading Dead Zones and Consequences for Marine Ecosystems. *Science* **2008**, *321*, 926–929.
- Paerl, H. W.; Gardner, W. S.; Havens, K. E.; Joyner, A. R.; McCarthy, M. J.; Newell, S. R.; Quin, B.; Scott, J. T. Mitigating cyanobacterial harmful algal blooms in aquatic ecosystems impacted by climate change and anthropogenic nutrients. *Harmful Algae* **2016**, *54*, 213–222.
- Duprey, N. N.; Yasuhara, M.; Baker, D. M. Reefs of tomorrow: eutrophication reduces coral biodiversity in an urbanized seascape. *Global Change Biol.* **2016**, *22*, 3550–3565.
- Wetzel, R. G. *Limnology. Lake and River Ecosystems*; Academic Press: San Diego, 1991.
- Paerl, H. W.; Huisman, J. Blooms like it hot. *Science* **2008**, *320*, 57–58.
- Paerl, H. W.; Huisman, J. Climate Change: A Catalyst for Global Expansion of Harmful Cyanobacterial Blooms. *Environ. Microbiol. Rep.* **2009**, *1* (1), 27–37.
- Wells, M. L.; Trainer, V. L.; Smayda, T. J.; Karlson, B. S.; Trick, C. G.; Kudela, R. M.; Ishikawa, A.; Bernard, S.; Wulff, A.; et al. Harmful algal blooms and climate change: Learning from the past and present to forecast the future. *Harmful Algae* **2015**, *49*, 68–93.
- Havens, K. E.; Paerl, H. W. Climate change at a crossroad for control of harmful algal blooms. *Environ. Sci. Technol.* **2015**, *49*, 12605–12606.
- Paerl, H. W.; Paul, V. Climate Change: Links to Global Expansion of Harmful Cyanobacteria. *Water Res.* **2012**, *46*, 1349–1363.
- Paerl, H. W. Controlling harmful cyanobacterial blooms in a climatically more extreme world: Management options and research needs. *J. Plankton Res.* **2017**, *39*, 763.
- Otten, T. G.; Crosswell, J. R.; Mackey, S.; Dreher, T. W. Application of molecular tools for microbial source tracking and public health risk assessment of a *Microcystis* bloom traversing 300 km of the Klamath River. *Harmful Algae* **2015**, *46*, 71–81.
- Eilers, J. M.; Kann, J.; Cornett, J.; Moser, K.; Amand, A. S. Paleolimnological evidence of change in a shallow, hypereutrophic lake: Upper Klamath Lake, Oregon, USA. *Hydrobiologia* **2004**, *520*, 7–18.
- Paerl, H. W.; Scott, J. T.; McCarthy, M. J.; Newell, S. E.; Gardner, W. S.; Havens, K. E.; Hoffman, D. K.; Wilhelm, S. W.; Wurtsbaugh, W. A. It takes two to tango: When and where dual nutrient (N & P) reductions are needed to protect lakes and downstream ecosystems. *Environ. Sci. Technol.* **2016**, *50*, 10805–10813.
- Conley, D. J.; Paerl, H. W.; Howarth, R. W.; Boesch, D. F.; Seitzinger, S. P.; Havens, K. E.; Lancelot, C.; Likens, G. E. (Controlling eutrophication: Nitrogen and phosphorus. *Science* **2009**, *323*, 1014–15.
- Paerl, H. W. Controlling Eutrophication along the freshwater–Marine Continuum: Dual Nutrient (N and P) Reductions are Essential. *Estuaries Coasts* **2009**, *32*, 593–601.
- Chaffin, J. D.; Davis, T. W.; Smith, D. J.; Baer, M. M.; Dick, G. J. Interactions between nitrogen form, loading rate, and light intensity on *Microcystis* and *Planktothrix* growth and microcystin production. *Harmful Algae* **2018**, *73*, 84–97.
- Kuosa, H. Picoplanktonic algae in the northern Baltic Sea seasonal dynamics and flagellate grazing. *Mar. Ecol.: Prog. Ser.* **1991**, *73*, 269–276.
- Sánchez-Baracaldo, P.; Handley, B. A.; Hayes, K. Picocyanobacterial community structure of freshwater lakes and the Baltic Sea revealed by phylogenetic analyses and clade-specific quantitative PCR. *Microbiology* **2008**, *11*, 3347–3357.
- Gaulke, A. K.; Wetz, M. S.; Paerl, H. W. Picophytoplankton: A major contributor to planktonic biomass and primary production in a eutrophic, river-dominated estuary. *Estuarine, Coastal Shelf Sci.* **2010**, *90*, 45–54.
- Jakubowska, N.; Szlag-Wasielewska, S. E. Toxic Picoplanktonic Cyanobacteria—Review. *Mar. Drugs* **2015**, *13*, 1497–1518.
- Carmichael, W. W. The cyanotoxins. *Adv. Bot. Res.* **1997**, *27*, 211–256.
- Toxic Cyanobacteria in Water*. Chorus, I.; Bartram, J.; Eds.; E&F Spon: London, 1991.
- Fetscher, A. E.; Howard, M. D. A.; Stancheva, R.; Kudela, R. M.; Stein, E. D.; Sutula, M. A.; Busse, L. B.; Sheath, R. G. Wadeable streams as widespread sources of benthic cyanotoxins in California, USA. *Harmful Algae* **2015**, *49*, 105–116.
- Paerl, H. W.; Fulton, R. S.; Moisaner, P. H.; Dyble, J. Harmful Freshwater Algal Blooms, With an Emphasis on Cyanobacteria. *Sci. World J.* **2001**, *1*, 76–113.
- Harmful Cyanobacteria. Springer Aquatic Ecology Series 3*; Huisman, J. M.; Matthijs, H. C. P.; Visser, P. M.; Eds.; Springer: Dordrecht, the Netherlands, 2005.
- Jeppesen, E.; Meerhoff, M.; Jacobsen, B. A.; Hansen, R. S.; Sondergaard, M.; Jensen, J. P.; Lauridsen, T. L.; Mazzeo, N.; Branco, C. W. C. Restoration of shallow lakes by nutrient control and

biomanipulation-the successful strategy varies with lake size and climate. *Hydrobiologia* **2007**, *581*, 269–285.

(28) Jarvie, H. P.; Smith, D. R.; Norton, L. R.; Edwards, F. K.; Bowes, M. J.; King, S. M.; Scarlett, P.; Davies, S.; Dils, R. M.; Bachiller-Jareno, N. Phosphorus and nitrogen limitation and impairment of headwater streams relative to rivers in Great Britain: A national perspective on eutrophication. *Sci. Total Environ.* **2017**, *621*, 849–862.

(29) Sharma, R. C.; Singh, N.; Chauhan, A. The influence of physico-chemical parameters on phytoplankton distribution in a head water stream of Garhwal Himalayas: A case study. *Egypt. J. Aquat. Res.* **2016**, *42*, 11–21.

(30) Bouma-Gregson, K.; Power, M. E.; Bormans, M. Rise and fall of toxic benthic freshwater cyanobacteria (*Anabaena* sp.) in the Eel River: Buoyancy and dispersal. *Harmful Algae* **2017**, *66*, 79–87.

(31) McAllister, T. G.; Wood, S. A.; Hawes, I. The rise of toxic benthic *Phormidium* proliferations: A review of their taxonomy, distribution, toxin content and factors regulating prevalence and increased severity. *Harmful Algae* **2016**, *55*, 282–294.

(32) Stevens, D. K.; Krieger, R. I. Effect of route of exposure and repeated doses on the acute toxicity in mice of the cyanobacterial nicotinic alkaloid anatoxin-a. *Toxicol.* **1991**, *29*, 134–138.

(33) McGregor, G. B.; Stewart, I.; Sendall, B. C.; Sadler, R.; Reardon, K.; Carter, S.; Wruck, D.; Wickramasinghe, W. First Report toxic *Nodulariaspumigena* (Nostocales/Cyanobacteria) Bloom in Sub-Tropical Australia. I. Phycological and Public Health Investigations. *Int. J. Environ. Res. Public Health* **2012**, *9*, 2396–2411.

(34) Dodds, W. K.; Smith, V. H. Nitrogen, phosphorus, and eutrophication in streams. *Inland Waters* **2016**, *6*, 155–164.

(35) Keck, F.; Lepori, F. Can we predict nutrient limitation in streams and rivers? *Freshwater Biol.* **2012**, *57*, 1410–1421.

(36) Vollmer-Sanders, C.; Allman, A.; Busdeker, D.; Moody, L. B.; Stanley, W. G. Building partnerships to scale up conservation: 4R Nutrient Stewardship Certification Program in the Lake Erie watershed. *J. Great Lakes Res.* **2016**, *42*, 1395–1402.

(37) Trenberth, K.E.. The impact of climate change and variability on heavy precipitation, floods, and droughts. In *Encyclopedia of Hydrological Sciences*; Anderson, M. G., Ed.; John Wiley and Sons, Ltd., 2005; pp 1–11.

(38) Bosch, N. S.; Allan, J. D.; Selegean, J. P.; Scavia, D. Scenario-testing of agricultural best management practices in Lake Erie watersheds. *J. Great Lakes Res.* **2013**, *39*, 429–436.

(39) King, S. E.; Osmond, D. L.; Smith, L.; Dukes, M.; Evans, R. O.; Knies, S.; Kunickis, S.; Burchell, M.; Gilliam, J. W. Effects of riparian buffer vegetation and width: A 12-year retrospective study. *J. Environ. Qual.* **2015**, *45*, 1243–1251.

(40) Paerl, H. W.; Otten, T. G. Duelling 'CyanoHABs': unravelling the environmental drivers controlling dominance and succession among diazotrophic and non-N₂-fixing harmful cyanobacteria. *Environ. Microbiol.* **2016**, *18*, 316–324.

(41) Carey, C. C.; Ibelings, B. W.; Hoffmann, E. P.; Hamilton, D. P.; Brookes, J. D. Eco-physiological adaptations that favour freshwater cyanobacteria in a changing climate. *Water Res.* **2012**, *46*, 1394–1407.

(42) Paerl, H. W.; Hall, N. S.; Calandrino, E. S. Controlling harmful cyanobacterial blooms in a world experiencing anthropogenic and climatic-induced change. *Sci. Total Environ.* **2011**, *409*, 1739–1745.

(43) Paerl, H. W.; Otten, T. G. Harmful Cyanobacterial Blooms: Causes, Consequences and Controls. *Microb. Ecol.* **2013**, *65*, 995–1010.

(44) Likens, G. E., Ed.; *Nutrients and Eutrophication*. *Am. Soc. Limnol. Oceanogr. Special Symp.* **1**, 1972.

(45) Schindler, D. W.; Vallentine, J. R. *The Algal Bowl: Over-fertilization of the World's Freshwaters and Estuaries*; Univ. of Alberta Press, 2008.

(46) Smith, V. H. Nitrogen, phosphorus, and nitrogen fixation in lacustrine and estuarine ecosystems. *Limnol. Oceanogr.* **1990**, *35*, 1852–1859.

(47) Edmondson, W. T.; Lehman, J. T. The effect of changes in the nutrient income and conditions of Lake Washington. *Limnol. Oceanogr.* **1981**, *26*, 1–29.

(48) Elmgren, R.; Larsson, U. Nitrogen and the Baltic Sea: Managing nitrogen in relation to phosphorus. *Sci. World J.* **2001**, *1*, 371–377.

(49) Vanderploeg, H. A.; Liebig, J. R.; Carmichael, W. W.; Agy, M. A.; Johengen, T. H.; Fahnenstiel, G. L.; Nalepa, T. F. Zebra mussel (*Dreissena polymorpha*) selective filtration promoted toxic *Microcystis* blooms in Saginaw Bay (Lake Huron) and Lake Erie. *Can. J. Fish. Aquat. Sci.* **2001**, *58*, 1208–1221.

(50) Schindler, D. W.; Hecky, R. E.; Findlay, D. L.; Stainton, M. P.; Parker, B. R.; Paterson, M.; 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*, 11254–58.

(51) 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*, 1265–70.

(52) Lewis, W. M.; Wurtsbaugh, W. A.; Paerl, H. W. Rationale for control of anthropogenic nitrogen and phosphorus in inland waters. *Environ. Sci. Technol.* **2011**, *45*, 10030–10035.

(53) Paerl, H. W. Physiological ecology and regulation of N₂ fixation in natural waters. *Adv. Microb. Ecol.* **1990**, *11*, 305–344.

(54) Scheffer, M. *Ecology of Shallow Lakes*; Chapman and Hall: London, 1982.

(55) Petersen, S. A. Lake restoration by sediment removal. *J. Am. Water Resour. Assoc.* **1982**, *18*, 423–35.

(56) Cronberg, G. Changes in the phytoplankton of Lake Trummen induced by restoration. *Hydrobiologia* **1982**, *86*, 185–93.

(57) Qin, B.; Zhu, G.; Gao, G.; Zhang, Y.; Li, W.; Paerl, H. W.; Carmichael, W. A drinking water crisis in Lake Taihu, China: Linkage to climatic variability and lake management. *Environ. Manage.* **2010**, *45*, 105–112.

(58) Mitrovic, S. M.; Oliver, R. L.; Rees, C.; Bowling, L. C.; Buckney, R. T. Critical flow velocities for the growth and dominance of *Anabaena circinalis* in some turbid freshwater rivers. *Freshwater Biol.* **2003**, *48*, 164–174.

(59) Maier, H. R.; Kingston, G. B.; Clark, T.; Frazer, A.; Sanderson, A. Risk-based approach for assessing the effectiveness of flow management in controlling cyanobacterial blooms in rivers. *River Res. and Applic.* **2004**, *20*, 459–471.

(60) Robb, M.; Greenop, B.; Goss, Z.; Douglas, G.; Adeney, J. Application of Phoslock, an innovative phosphorus binding clay, to two Western Australian waterways: preliminary findings. *Hydrobiologia* **2003**, *494*, 237–243.

(61) Lüring, M.; Oosterhout, F. Case study on the efficacy of a lanthanum-enriched clay (Phoslock®) in controlling eutrophication in Lake Het Groene Eiland (The Netherlands). *Hydrobiologia* **2013**, *710*, 253–263.

(62) Yu, Z.; Song, X.; Cao, X.; Liu, Y. Mitigation of harmful algal blooms using modified clays: Theory, mechanisms, and applications. *Harmful Algae* **2017**, *69*, 48–64.

(63) Li, L.; Pan, G. A universal method for flocculating harmful algal blooms in marine and fresh waters using modified sand. *Environ. Sci. Technol.* **2013**, *47*, 4555–4562.

(64) Lembi, C. A. *Aquatic Plant Management*. In *Barley Straw for Algae Control*; Purdue University Cooperative Extension: West Lafayette, IN, 2002.

(65) Matthijs, H. C. P.; Visser, P. M.; Reeze, B.; Meeuse, J.; Slot, P. C.; Wijn, G.; Talens, R.; Huisman, J. Selective suppression of harmful cyanobacteria in an entire lake with hydrogen peroxide. *Water Res.* **2012**, *46*, 1460–1472.

(66) Krogman, D. W.; Butalla, R.; Sprinkle, J. Blooms of Cyanobacteria on the Potomac River. *Plant Physiol.* **1986**, *80*, 667–671.

(67) Calandrino, E.; Paerl, H. W. Determining the potential for the proliferation of the harmful cyanobacterium *Cylindrospermopsis raciborskii* in Currituck Sound, North Carolina. *Harmful Algae* **2011**, *11*, 1–9.

(68) Lehman, P. W.; Kurobe, T.; Lesmeister, S.; Baxa, D.; Tung, A.; The, S. J. Impacts of the 2014 severe drought on the *Microcystis* bloom in San Francisco Estuary. *Harmful Algae* **2017**, *63*, 94–108.

- (69) Kurobe, T.; Lehman, P. W.; Haque, M. E.; Sedda, T.; Lesmeister, S.; The, S. Evaluation of water quality during successive severe drought years within *Microcystis* blooms using fish embryo toxicity tests for the San Francisco Estuary, California. *Sci. Total Environ.* **2018**, *610*, 1029–37.
- (70) Miller, M. A.; Kudela, R. M.; Mekebri, A.; Crane, D.; Tinker, M. T.; Oates, C. S.; Staedler, M.; Miller, A. W.; Hardern, D.; Ward, K. Evidence for a novel marine harmful algal bloom: cyanotoxin (microcystin) transfer from land to sea otters. *PLoS One* **2010**, *5* (9), e12576.
- (71) Gobble, C. M.; Peacock, M. B.; Kudela, R. M. Evidence of freshwater algal toxins in marine shellfish: Implications for human and aquatic health. *Harmful Algae* **2016**, *59*, 59–66.
- (72) Jarvie, H. P.; Sharples, A. N.; Withers, P. J. A.; Scott, J. T.; Haggard, B. E.; Neal, C. Phosphorous mitigation to control river eutrophication: Murky waters, inconvenient truths, and “postnormal” science. *J. Env. Qual.* **2013**, *42*, 295–304.
- (73) Heisler, J.; Glibert, P.; Burkholder, J.; Anderson, D.; Cochlan, W.; Dennison, W.; Gobler, C.; Dortch, Q.; Heil, C.; Humphries, E.; Lewitus, A.; Magnien, R.; Marshall, H.; Sellner, K.; Stockwell, D.; Stoecker, D.; Suddleson, M. Eutrophication and harmful algal blooms: a scientific consensus. *Harmful Algae* **2008**, *8* (1), 3–13.
- (74) 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.
- (75) Capone, D. G.; Hutchins, D. A. Microbial biogeochemistry of coastal upwelling regimes in a changing ocean. *Nat. Geosci.* **2013**, *6*, 711–717.
- (76) Sengupta, A.; Sutula, M. A.; McLaughlin, K.; Howard, M. D. A.; Tiefenthaler, L.; Bitner, T. V. Terrestrial Nutrient Loads and Fluxes to the Southern California Bight, USA. In *Southern California Coastal Water Research Project 2013 Annual Report*; Schiff, K. C.; Miller, K., Eds.; Southern California Coastal Water Research Project: Costa Mesa, CA, 2013, pp 245e258.
- (77) Howard, M. D. A.; Kudela, R. M.; McLaughlin, K. New insights into impacts of anthropogenic nutrients on urban ecosystem processes on the Southern California coastal shelf: Introduction and synthesis. *Estuarine, Coastal Shelf Sci.* **2017**, *186*, 163–170.
- (78) Sinha, E.; Michalak, A. M.; Balaji, V. Eutrophication will increase during the 21st century as a result of precipitation changes. *Science* **2017**, *357*, 405–408.
- (79) Greaver, T. L.; Clark, C. M.; Compton, J. E.; Vallano, D.; Talhelm, A. F.; Weaver, C. P.; Band, L. E.; Baron, J. S.; Davidson, E. A.; Tague, C. L.; Felker-Quinn, E. Key ecological responses to nitrogen are altered by climate change. *Nat. Clim. Change* **2016**, *6* (9), 836.
- (80) Paerl, H. W. Coastal eutrophication and harmful algal blooms: Importance of atmospheric deposition and groundwater as “new” nitrogen and other nutrient sources. *Limnol. Oceanogr.* **1997**, *42* (Spart2), 1154–1165.
- (81) Glibert, P. M.; Maranger, R.; Sobota, D. J.; Bouwman, L. The Haber Bosch–harmful algal bloom (HB–HAB) link. *Environ. Res. Lett.* **2014**, *9* (10), 105001.
- (82) Zhan, X.; Bo, Y.; Zhou, F.; Liu, X.; Paerl, H. W.; Shen, J.; Wang, R.; Li, F.; Tao, S.; Dong, Y.; Tang, X. Evidence for the Importance of Atmospheric Nitrogen Deposition to Eutrophic Lake Dianchi, China. *Environ. Sci. Technol.* **2017**, *51* (12), 6699–6708.
- (83) Levasseur, M.; Gamache, T.; St-Pierre, I.; Michaud, S. Does the cost of NO₃ reduction affect the production of harmful compounds by *Alexandrium excavatum*. In *Harmful Marine Algal Blooms*; Lassus, P.; Arzul, G.; Erad, E.; Gentien, P.; Marcaillou, C.; Eds.; Lavoisier Science Publishers: Paris, 1995; pp 463–468.
- (84) John, E. H.; Flynn, K. J. Growth dynamics and toxicity of *Alexandrium fundyense* (Dinophyceae): the effect of changing N [ratio] P supply ratios on internal toxin and nutrient levels. *Eur. J. Phycol.* **2000**, *35*, 11–23.
- (85) Hamasaki, K.; Horie, M.; Tokimitsu, S.; Toda, T.; Taguchi, S. Variability in toxicity of the dinoflagellate *Alexandrium tamarense* isolated from Hiroshima Bay, western Japan, as a reflection of changing environmental conditions. *J. Plankt. Res.* **2001**, *23*, 271–278.
- (86) Leong, S. C. Y.; Murata, A.; Nagashima, Y.; Taguchi, S. 2004. Variability in toxicity of the dinoflagellate *Alexandrium tamarense* in response to different nitrogen sources and concentrations. *Toxicol.* **2004**, *43*, 407–415.
- (87) Kudela, R. M.; Peterson, T. D. Influence of a buoyant river plume on phytoplankton nutrient dynamics: What controls standing stocks and productivity? *J. Geophys. Res.* **2009**, *114*, COOB1.
- (88) Howarth, R.; Chan, F.; Conley, D. J.; Garnier, J.; Doney, S. C.; Marino, R.; Billen, G. Coupled biogeochemical cycles: eutrophication and hypoxia in temperate estuaries and coastal marine ecosystems. *Front. in Ecol. and Environ.* **2011**, *9*, 18–26.
- (89) Elser, J. J.; Bracken, M. E. S.; Cleland, E. E.; Gruner, D. S.; Harpole, W. S.; Hillebrand, H.; Bgai, J. T.; Seabloom, E. W.; Shurin, J. B.; Smith, J. E. Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. *Ecol. Lett.* **2007**, *10*, 1124–1134.
- (90) O’Neil, J. M.; Davis, T. W.; Burford, M. A.; Gobler, C. J. The rise of harmful cyanobacteria blooms: potential role of eutrophication and climate change. *Harmful Algae* **2012**, *14*, 313–334.
- (91) Glibert, P. M.; Burford, M. A. Globally changing nutrient loads and Harmful Algal Blooms: Recent advances, new paradigms, and continuing challenges. *Oceanography* **2017**, *30*, 58–69.