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Aluminum treatments to control internal phosphorus loading in lakes on Cape Cod, Massachusetts

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ABSTRACT

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Ten lakes on Cape Cod, Massachusetts have been treated with aluminum since 1995, with 2 of those lakes receiving a second treatment. Elimination of cyanobacterial blooms was the goal in each case. Cape Cod lakes are mostly kettleholes with long detention times. Precipitation and groundwater dominated hydrologic inputs, but internal load contributed a major portion of the total phosphorus (TP) load to treated lakes. Aluminum sulfate and sodium aluminate were used in treatments with the intent to maintain a near-neutral pH in low alkalinity waters. Aluminum doses ranged from 10 to 100 g/m^2, depending on mobile P concentrations in the upper 10 cm of sediment. Results varied, although all treatments lowered TP, provided relief from blooms, increased water clarity, and reduced oxygen demand for multiple years. Important lessons learned over 2 decades include how to apply aluminum to prevent fish mortality, options for dose calculation, and expectations for duration of benefits. To control internal loading and minimize cyanobacterial blooms at the lowest cost, treatment of Cape Cod lakes should apply aluminum at a concentration at least 10 times the mobile P concentration in the upper 10 cm of sediment exposed to anoxia and at concentrations that leave no more than 100 mg/kg of iron-bound P in laboratory inactivation assays. Cost of treatment in 2016 dollars averaged \$150/g/m^2 of applied aluminum per ha treated. Aluminum treatment has been demonstrated as a valued lake management tool to improve water quality and reduce nuisance algal blooms in Cape Cod lakes.

KEYWORDS
Algae management; aluminum; P inactivation; water clarity

Aluminum (Al) has been used in water treatment as a coagulant for more than 200 years but has been used to inactivate phosphorus (P) in lakes for only about the last 45 years (Cooke et al. 2005). It has become more common as a means to reverse eutrophication in recent years but has been subject to technical constraints relating to dose determination, management challenges in external load control, and regulatory limitations related to potential Al toxicity. Many advances that maximize performance have been made in the last few decades (e.g., Welch and Cooke 1999, Huser 2012, James and Bischoff 2015, Huser et al. 2016b). Yet concern has lingered about conducting successful treatments with minimal negative effects on fish and invertebrates in low alkalinity lakes, where applied Al compounds can cause pH shifts that create toxic conditions.

Beginning in the 1990s, evaluation of kettlehole lakes on Cape Cod in Massachusetts revealed high internal loading as the dominant P source supporting cyanobacterial blooms (e.g., BEC 1993, ENSR 2001, 2008). Watershed management alone could not rehabilitate these lakes, and in-lake management options were sought. Treatment with Al was perceived as having high potential, but potential toxicity in lakes with alkalinity typically $<$10 mg/L as calcium carbonate ($CaCO_3$) equivalents and pH normally $<$6.5 standard units in the absence of blooms necessitated caution in applications. The experience with Al treatments in Cape Cod lakes over the last 20 years is reported here. We summarize physical features, hydrology, and P loading to targeted lakes, dose determination for Al treatment, both positive and negative treatment effects, duration of benefits, and costs.

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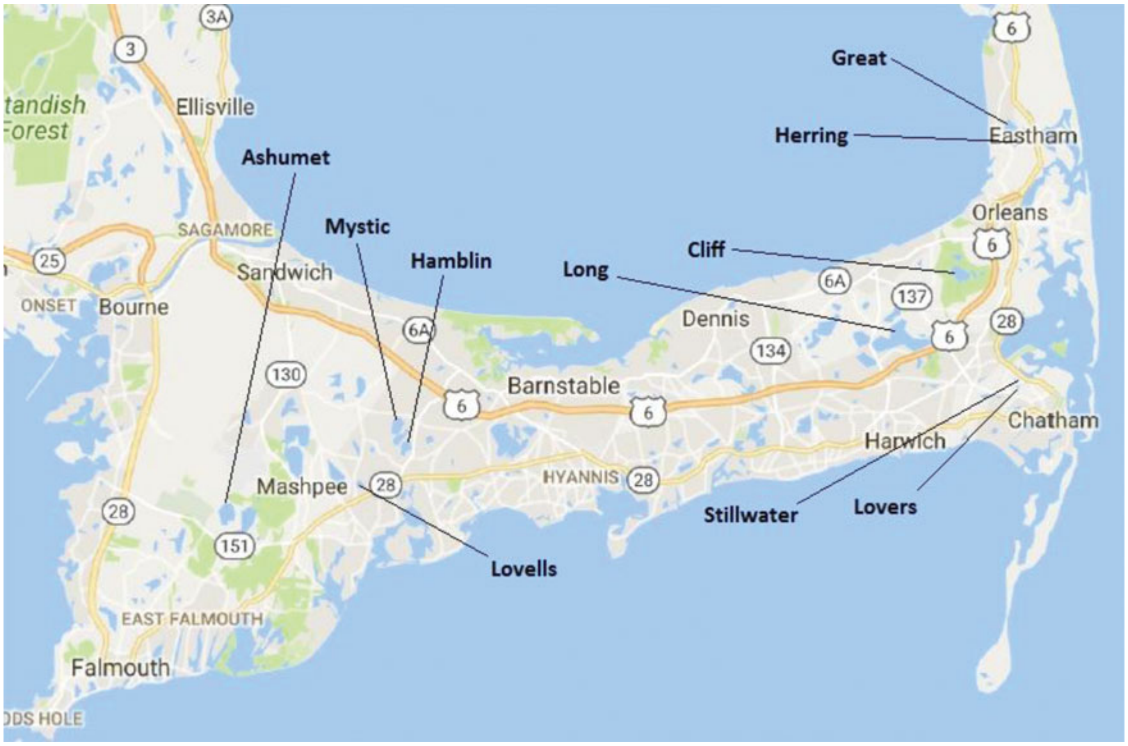

Figure 1. Locations of treatment lakes on Cape Cod, MA. Base map from Acme Mapper 2.1.

Material and methods

Treatment lakes

The 10 kettlehole lakes treated through 2016 (Fig. 1) ranged in size from 7.5 to 296 ha with maximum depths ranging from 10 to 26.7 m (Table 1). Each lake is deep enough to stratify in summer and all have alkalinity <10 mg/L as $CaCO_3$ equivalents. All but 2 are called ponds as part of their names, and although there is no universally accepted threshold for separating ponds and lakes, most lake managers would consider these waterbodies to be lakes.

Each lake was studied prior to treatment under programs sponsored by various organizations, including the Commonwealth of Massachusetts, the Cape Cod Commission, the School for Marine Science and Technology at University of Massachusetts-Dartmouth, the Air Force Civil Engineer Center, and the towns in which the lakes lie. These diverse studies (BEC 1987, 1991, 1993, AFCEE 2001, 2015, ENSR 2001, 2008, Loon Environmental 2012, Moran 2014a, 2014b, WRS 2011, 2014a, 2014b) led to a common recommendation for P inactivation to reduce internal loading, with iron-bound P (Fe-P) as the dominant form of P released from sediment under anoxia. Few of the lakes have any surface tributaries, and direct precipitation and groundwater comprise at least 94% of hydrologic inputs for all but Stillwater Pond in Chatham, which gets

Table 1. Features of lakes treated with aluminum between 1995 and 2015.

		Depth				Hydrologic load		Phosphorus load		
Lake	Area ha	Mean m	Maximum m	Detention yr	Tributaries	Precipitation %	Groundwater %	Internal %	Surface Flow %	Groundwater %
Hamblin	46.0	8.3	18.8	1.0	No	11	87	67	9	11
Ashumet	82.0	7.0	20.0	1.9	No	17	72	47	4	45
Long	296.0	8.8	21.2	3.5	No	51	44	64	17	9
Mystic	59.0	4.6	14.3	1.0	No	18	80	46	15	21
Lovers	15.0	4.6	10.0	1.2	No	39	55	43	27	12
Stillwater	7.5	6.8	13.9	1.2	From Lovers	13	25	55	32	5
Herring	17.7	6.2	10.9	2.8	No	42	55	40	1	46
Great	44.7	3.6	11.0	0.4	2 small ones	14	83	26	8	45
Lovell's	22.0	5.7	11.4	2.1	1 diverted	43	53	62	4	16
Cliff	83.0	8.6	26.7	5.3	No	71	23	67	5	6

most of its water from upstream Lovers Lake, another treatment lake. Detention time for all but Great Pond in Eastham is >1 year. Internal load of P was estimated at 40–67% of the total load, except for Great Pond estimated at 26%. Most of the lakes were impacted by some watershed activity in the past that contributed to the accumulation of P in the sediments, but influences other than residential development were largely absent by the time of treatment. The environmental setting for each lake (see the online supplemental materials) illustrates the commonalities and idiosyncrasies of these cases.

Dose determination

Chemical doses for nutrient inactivation were calculated using various methods over the years, initially based on the amount of P accumulated in the hypolimnion that required inactivation and later by both direct testing of Fe-P in surficial sediment and lab assays with sediment and Al (BEC 1993, WRS 2014b). The fractionation method of Psenner et al. (1988) allowed assessment of key P fractions for dose planning, but later work (Rydin and Welch 1999, Rydin et al. 2000, Reitzel et al. 2005, James and Bischoff 2015) helped advance dose determination.

The process used for lake treatments on Cape Cod after 2001 involved initial testing for total P (TP), loosely sorbed P, and Fe-P in the upper 10 cm of sediment exposed to anoxia. Loosely sorbed P was low and nearly always below the detectable threshold; Fe-P is the dominant form of mobile P in these waterbodies. Based on percent solids and sediment gravity, P was calculated as grams per square meter in the top 10 cm, and the Al dose was set at a minimum of 10 times Fe-P concentration. The active zone of interaction is possibly >10 cm, but 10 cm has been a traditionally applied threshold for Al-P formation (Welch et al. 2017). Where there is considerable spatial variation of sediment P concentrations, area-specific doses have been established for treatment zones.

An alternative approach applied to some of the lakes was based on modification of the method of Rydin and Welch (1999), following the general approach of Pilgrim et al. (2007). Al as aluminum sulfate and sodium aluminate at a 2:1 volumetric ratio was added to 5 g of sediment suspended in 50 mL of distilled water at amounts corresponding to treatment in grams per square meter and allowed to interact under agitation for at least 12 hours. A sediment pellet was reconstituted by centrifugation and subjected to extraction of Fe-P, which likely includes porewater and loosely sorbed P. By repeating this process with different Al concentrations, a dose response curve (Fig. 2) can be generated, and the tradeoff among dose, degree of P inactivation, and cost can be considered. The range of total doses for Cape Cod lakes has been 10 to 100 g/m^2 (Table 2), although application on any day was limited to 25 g/m^2 to minimize potential Al toxicity.

Aluminum application

All but the original Hamblin Pond treatment (conducted by Sweetwater Technology from Minnesota) were performed by Aquatic Control Technology (now part of SOLitude Lake Management) of Massachusetts. Treated areas typically included the sediment area exposed to anoxia, although reduction in treated area occurred if sediment testing revealed low Fe-P concentrations or low organic sediment content. The treatment footprint most often reflected the area overlain by soft organic sediment; where organic substrate dominates, oxygen depletion often occurs, whereas Fe-P and oxygen demand tend to be low in sandy areas. The first Ashumet Pond treatment was an exception; a smaller area was treated to temporarily improve conditions while the geochemical barrier was installed, and the second treatment in 2010 covered twice as much area at a similar dose.

Al compounds were added from a self-propelled barge with 2 tanks, 1 for aluminum sulfate and 1 for sodium aluminate. Each Al compound was injected separately by metered pumping through ports on a manifold lowered to a depth of about 2.5 m below the lake surface. A deeper (10 m) depth of injection, involving a specialized boom, was mandated by permit in the first Ashumet treatment in 2001 to minimize possible impacts to biota in the epilimnion. Although this slowed treatment, added cost, and limited the opportunity to strip P from the entire water column, it was necessary to ensure permit approvals after the Hamblin Pond treatment in 1995 caused fish mortality. The second treatment in 2010 was approved for injection in the epilimnion at a depth of 3 m. GPS guidance was used on the treatment barge to ensure an accurate and even application.

The default ratio for chemical addition was 2 parts aluminum sulfate to 1 part sodium aluminate by volume, although this ratio can be altered to maintain the targeted pH range of 6.0 to 8.0 standard units. Based

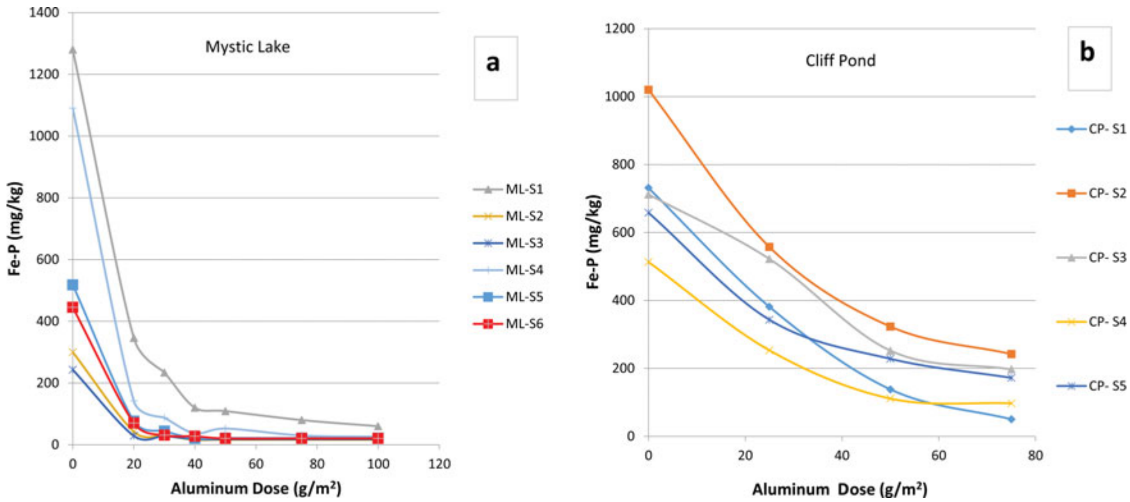

Figure 2. Dose response curve for Fe-P in (a) 6 sediment samples from Mystic Lake and (b) 5 sediment samples from Cliff Pond treated with aluminum in the laboratory.

on experience gained in Cape Cod lakes and elsewhere in New England, but particularly applicable to low alkalinity lakes, Al was applied at rates that resulted in no more than 5 mg/L in the mixed zone behind the barge, which was typically 5 m deep. This Al concentration has been found to minimize toxicity to fish and invertebrates, even if the pH exceeds the target range (unpublished lab assay data); therefore, any dose >25 g/m^2 was applied in at least 2 periods separated by at least 1 day. Additionally, the target treatment area was broken into zones (Fig. 3), and contiguous zones were not treated consecutively if possible. This 3-level protection system (pH control, dose control, treatment area control) has prevented significant fish mortality in all treatments after 2000 in New England, defined as no more than 50 dead fish observed per day during the treatment period in permit documents. Impacts to fish eggs or fry are possible but were not observed.

Treatment in spring was preferred because algae concentrations were lower (limited interference with Al reactions), little P had yet been released from targeted sediment (improved treatment efficiency), and thermal gradients were minimal (allowed a larger vertical mixing zone and limited toxicity). Yet the timing of treatment was subject to regulation by permit, with concern expressed by state agencies over treatment in lakes with herring runs during April into June. Treatments of lakes with herring runs were therefore performed in September and early October while the lakes were still stratified and P and algal concentrations were elevated.

Monitoring program and analytical methods

Al treatments had no standard monitoring protocol, and measured water quality features and frequency of sampling varied based on permit conditions

Table 2. Information for 12 treatments of 10 lakes on Cape Cod.

			Aluminum application			
			Treatment area		Treatment dose	
Lake	Year Treated	Season Treated	ha	% of lake	g/m^2	max. mg/L
Hamblin	1995	Spring	32.3	69.6	45	9.0
Hamblin	2015	Spring	27.0	58.7	45–71	4.5
Ashumet	2001	Summer	11.3	13.8	43	4.3
Ashumet	2010	Fall	22.8	27.8	40	4.0
Long	2007	Fall	148.0	50.0	10–30	3.0
Mystic	2010	Fall	23.2	39.3	30–50	3.3
Lovers	2010	Fall	10.0	66.7	100	5.0
Stillwater	2010	Fall	5.0	66.7	100	5.0
Herring	2012	Fall	8.0	45.2	75	5.0
Great	2013	Fall	11.2	25.1	25	5.0
Lovell's	2014	Spring	14.0	63.6	50	2.5
Cliff	2016	Spring	31.0	37.3	75	3.8

Figure 3. Treatment zones and dates of treatment (month/day in 2007) for Long Pond.

and funding. All the treatment lakes were studied in advance of the treatment (supplemental material), so pre-treatment conditions were generally well known. For this analysis, we used data from up to 3 years prior to treatment to establish pre-treatment water quality. Monitoring during treatment was focused on maintaining pH within the acceptable range, conducting treatment during favorable weather conditions (i.e., avoiding high wind and waves), and visual surveys for any fish and mussel mortality or behavioral anomalies.

For most lakes, post-treatment monthly data from May through September were collected for at least 2 years. For some lakes data were available for the April through October period, and in some cases monitoring has continued in all years since treatment. In a few cases, only a single assessment was made in late summer each year as part of the Pond and Lake Steward (PALS) program for Cape Cod, administered by the School for Marine Science and Technology at the University of Massachusetts, Dartmouth. Up to 3 years of data were used to establish post-treatment conditions for comparison to pre-treatment water quality. Where a longer (>3 years) record was available, long-term trends after treatment were examined to assess longevity of benefits, but additional data were not factored into the basic pre- and post-treatment comparisons. The extent of available data is addressed in the supplemental material.

Monitoring included surface (upper 1 m) and bottom (lower 1 m) TP, summer Secchi disk transparency, summer chlorophyll *a* (Chl-*a*) and temperature–dissolved oxygen profiles for all lakes. TP was assessed by multiple analytical laboratories, usually by Standard Method SM 4500 PE (APHA 2005). Secchi transparency was measured using a view tube in almost all monitoring events. Chl-*a* was assessed by Standard Method 10200 H for discrete samples but was sometimes supplemented with field fluorescence measurements using probes from Turner Designs. Temperature and dissolved oxygen were assessed with calibrated field probes on YSI or Hach multi-probe instruments.

Additional monitoring conducted for some lakes included phytoplankton and zooplankton identification and enumeration by Standard Methods 10200 F and G, respectively, dissolved and/or total Al by Standard Method 3500-Al B, and forms of nitrogen (N; nitrate, ammonium, total Kjeldahl N, and total N), also by standard methods. Conductivity, turbidity, and pH were usually assessed with calibrated field probes on YSI or Hach instruments. Alkalinity was measured by titration in the field or laboratory (SM 2320 B).

Table 3. Summary of pre- and post-treatment monitoring results for treated Cape Cod lakes.

	Surface total P		Bottom total P		Summer Secchi depth		Summer Chl-*a*		Oxygen demand	
Lake	Pre-trtmt µg/L	Post-trtmt µg/L	Pre-trtmt µg/L	Post-trtmt µg/L	Pre-trtmt m	Post-trtmt m	Pre-trtmt µg/L	Post-trtmt µg/L	Pre-trtmt mg/m^2/d	Post-trtmt mg/m^2/d
Hamblin 1995	42	10	454	46	1.8	5.6	21.3	2.0	1720	219
Hamblin 2015	20	7	310	13	1.9	7.8	37.9	1.2	980	515
Ashumet 2001	26	17	290	100	2.8	3.5	6.4	4.1	1034	No data
Ashumet 2010	16	10	300	60	2.9	3.8	5.7	3.2	No data	No data
Long	30	16	163	62	2.8	5.4	12.6	5.5	1823	935
Mystic	35	15	555	65	1.2	3.9	19.7	3.5	780	213
Lovers	32	12	116	24	1.0	3.0	32.2	2.4	1500	555
Stillwater	28	11	290	38	1.3	3.3	21.6	1.8	1500	311
Herring	21	11	357	21	0.5	4.4	19.0	2.9	866	334
Great	22	14	57	32	2.3	2.5	8.4	6.5	1625	300
Lovell's	61	12	167	35	2.0	4.2	14.3	2.3	1310	530
Cliff	23	5	87	12	2.0	5.7	12.6	1.8	2465	1680

Oxygen demand was calculated as the mass difference over time in oxygen in the water column below the depth at which the thermocline normally forms (in mg/m^2/d). Changes in uptake kinetics dictate that oxygen demand is not fully expressed when values decline to <2 mg/L, so only profiles with values >2 mg/L at all assessed depths were used; these occurred mainly in spring. As the water warmed over this period and held less oxygen, demand was adjusted for losses due to temperature change.

Comparisons among lakes were mainly graphical, and changes were apparent without need for statistical analysis. However, the relationships among water clarity, Chl-*a*, and P were explored with least squares linear regression in Microsoft Excel, as were relationships between Al dose versus P concentration and oxygen consumption versus temperature.

Results

Phosphorus

Comparison of pre-treatment and post-treatment summer P concentrations between surface or bottom sampling locations indicate a decrease in all cases (Table 3). P near the bottom during stratification was elevated prior to treatment and much lower after treatment, consistent with Al binding of surficial sediment P and decreased release from that sediment. All post-treatment average P concentrations for bottom samples were <100 µg/L, with 8 of 12 averages <46 µg/L. The difference between pre- and post-treatment bottom P was smallest for Great Pond, which was treated preemptively rather than to remediate existing algal blooms. Pre-treatment summer P concentrations in surface waters were not as high as near the bottom, so differences between pre- and post-treatment surface P were not as striking. Yet the decrease was substantial in terms of nutrient supplies for algal blooms, with the surface P concentration declining to <17 µg/L in all cases and <12 µg/L in 8 of 12 treatments (Table 3).

There was no strong relationship between surface and bottom P concentration before treatment or the decrease in bottom or surface P concentration with Al dose (all R^2 values <0.01), but there was a relationship between surface and bottom P concentrations after treatment ($R^2 = 0.57$). The mechanisms by which P released from sediments accumulates in the hypolimnion or reaches the photic zone are more complicated than can be explained by such simple comparisons, but reducing hypolimnetic P does limit how much P can be transported into the epilimnion. The 4 spring treatments yielded surface P reductions that ranged from 65% to 80% compared to 35% to 63% for the 8 summer or fall treatments, suggesting that spring treatments may be more effective in suppressing P recycling.

Water clarity

Post-treatment Secchi transparency values increased from pre-treatment values (Table 3) in all cases, but the relative changes were highly variable. Pre-treatment Secchi transparency during summer ranged from 0.5 to 2.9 m with an average of 1.9 m, whereas post-treatment summer values ranged from 2.5 to 7.8 m with an average of 4.4 m. Great Pond experienced only a 0.2 m increase in Secchi transparency, whereas transparency increased in nearby Herring Pond by almost 4 m and in Hamblin Pond by 5.9 m through 2 years after the second (2015) treatment.

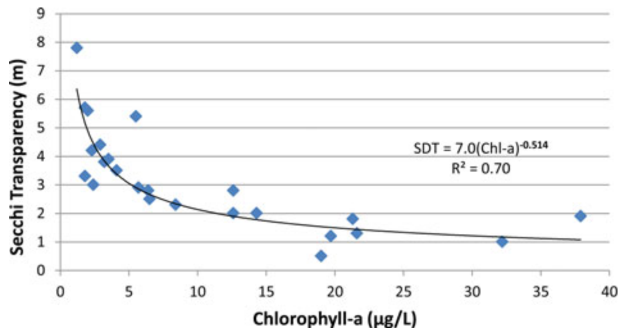

Figure 4. Average summer chlorophyll *a* vs. Secchi transparency for study lakes.

Water clarity in all treated lakes is mainly a function of algal abundance (Fig. 4); inflows are low even during storms, and wind-induced turbulence rarely extends to depths where soft sediments are abundant, limiting other sources of turbidity. Algal abundance is clearly linked to surface P (Fig. 5), but the relationship is not as strong as for Chl-*a* and transparency, so additional factors beyond P concentration are important to algae abundance.

Algae

The quantity of algae in each lake after treatment was reduced, based on the change in Chl-*a* concentration (Table 3). Equally important was meeting the important restoration objective to decrease dominance of the phytoplankton community by cyanobacteria. This algal reduction was not evident from Chl-*a* values but has been reported from all treated lakes in which cyanobacterial blooms significantly impaired lake uses. Long Pond provides the best algae record and experienced a major decline (from 69% pre-treatment to 9% post-treatment on average) in the portion of algal biomass represented by cyanobacteria for April to October samples since treatment (Fig. 6). Hamblin Pond was free of cyanobacterial blooms for almost 19 years after its first treatment, and after a 1-year period of resumed blooms by the same genus of cyanobacterium (*Dolichospermum*), the second treatment again eliminated blooms. However, although algal biomass was reduced in Lovell's Pond and cyanobacteria were limited for most of 2 summers, cyanobacteria resurged in late 2015 (Fig. 7) and continued to increase in 2016 with a concurrent decrease in clarity.

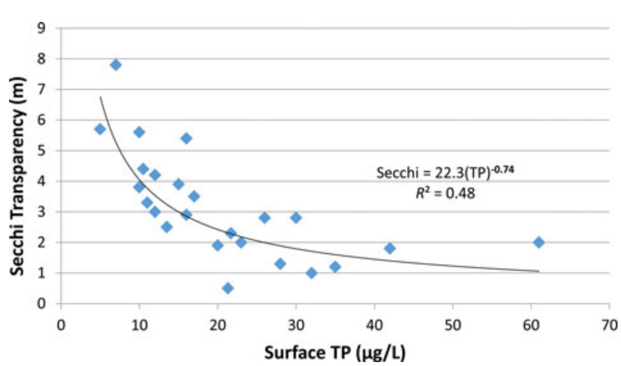

Figure 5. Average surface total phosphorus (TP) vs. Secchi transparency for study lakes. SDT = Secchi disk transparency.

Oxygen demand

Reduction of algal biomass that eventually sinks and decays is expected to cause a decrease in oxygen demand in response to Al treatment. Decreased oxygen demand (Table 3) was observed in all treated lakes except for Ashumet, where the monitoring program did not include spring sampling and precluded assessment of oxygen demand as calculated here. The range of estimated pre-treatment hypolimnetic oxygen demand (HOD) ranged from 866 to 2465 mg/m^2/d with an average of 1418 mg/m^2/d, whereas post-treatment HOD has ranged from 213 to 1680 mg/m^2/d with an average of 559 mg/m^2/d. Individual lake HOD decreases ranged from 32% to 87% with an average decline of 63%. The highest values for both pre- and post-treatment HOD of 2465 and 1680 mg/m^2/d, respectively, occurred in Cliff Pond, treated in 2016. Only one value was available for each period from Cliff Pond, and the post-treatment value represents a period when algae settled from the water column by treatment would be decaying. The next highest HOD values for the dataset were 1823 mg/m^2/d (pre-treatment) and 935 mg/m^2/d (post-treatment) from Long Pond, where more data were available.

Despite reduced oxygen demand, treated lakes still experienced anoxia in deep water, but complete hypolimnetic anoxia was shifted to later in summer and did not always occur after treatment. Monitoring at Long Pond provided HOD assessments over multiple years (Fig. 8), indicating the variability in HOD among post-treatment years and among the 2 basins of the lake, both treated, but at different doses (30 g/m^2 east, 15 g/m^2 west). There was no clear trend over time, although HOD has declined in recent years. Weather seemed to play a major role in development of anoxia, and there was a positive linear correlation between hypolimnetic temperature and oxygen demand in Long Pond ($R^2 = 0.62$ and 0.70, respectively).

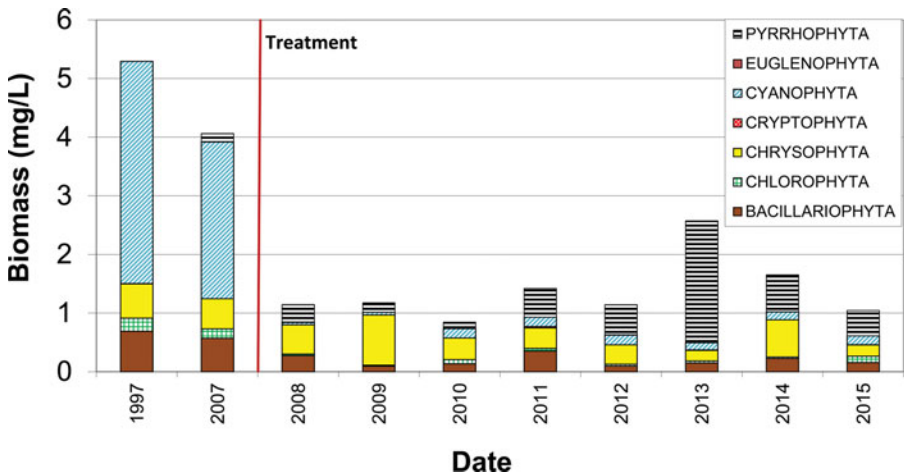

Figure 6. Average Jun–Sep biomass of major algal divisions in Long Pond before and after treatment with aluminum.

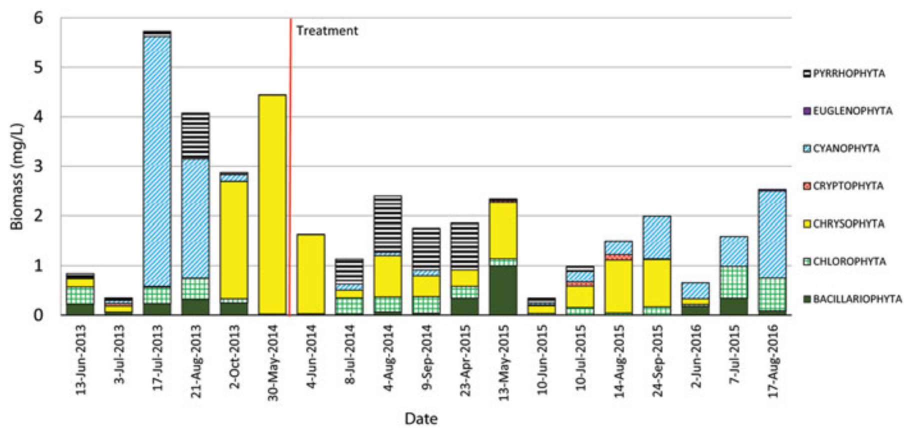

Figure 7. Biomass of major algal divisions in samples from Lovell's Pond.

The reduction in HOD translated into greater volumes of "trout water," the water layer defined by an upper boundary where the temperature exceeds 21 C and a lower boundary where oxygen is <5 mg/L during stratification. Many Cape Cod lakes are stocked with trout, but survival over summer is limited where there is no suitable habitat. Hamblin and Cliff Ponds have long been considered premier trout fishing lakes on Cape Cod and are stocked every year, but neither had any trout water for multiple summers prior to Al treatment. Cliff Pond was treated in 2016 and the record was too limited to draw any conclusion. The depth of water available to trout in Hamblin Pond increased by about 5 m after the first treatment (Fig. 9a), and most of that benefit lasted until the second treatment. The second treatment increased trout water by 2 m (Fig. 9b), returning the volume of water available to trout to that available shortly after the first treatment.

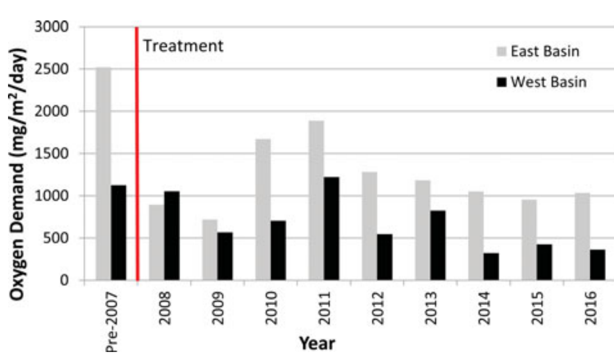

Figure 8. Hypolimnetic oxygen demand in 2 basins of Long Pond over time.

Nontarget impacts

Fish mortality beyond that allowed by permit for treatments of Cape Cod lakes occurred only during the first Hamblin Pond treatment, where a low ratio of aluminum sulfate to sodium aluminate (1.66:1) caused the pH to rise to >9 standard units from about 6.3 standard units over the 2 days of treatment covering ~70% of the lake. Average Al concentration was estimated at 9 mg/L. Assays and jar tests conducted prior

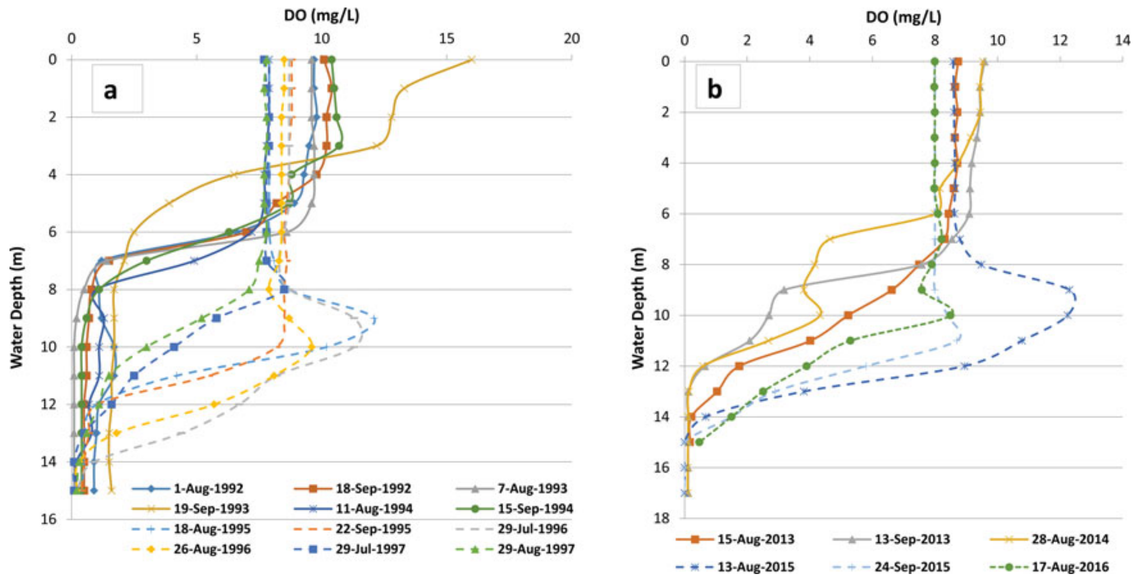

Figure 9. Dissolved oxygen (DO) profiles for Hamblin Pond before (solid lines) and after (dashed lines) (a) 1995 and (b) 2015 aluminum treatments.

to treatment did not properly guide treatment, and the Massachusetts Department of Environmental Protection estimated that ∼16,000 fish (perch, bass, and trout) were killed (unpublished data). This and a similar incident in Connecticut in 2000 resulted in more detailed but unpublished laboratory work that shaped the treatment limits and procedures after 2000. By maintaining the pH between 6 and 8 standard units and Al concentrations <5 mg/L, along with treating noncontiguous zones in successive treatments, observable fish mortality has been avoided in all other treatments on Cape Cod.

Permits issued to date for lakes that support herring runs have required timing of treatment to avoid the spawning period in late April to early June; instead, such lakes were treated in September and October. Although treatments are generally applied in water deeper than that used by fish for spawning, slight drift of floc into shallower water is common during treatment if there is any wind. There is no evidence of egg or fry mortality from Al toxicity, however, and monitoring during late summer treatments has detected many young-of-the-year herring swimming in the Al floc with no evident mortality.

Impacts to invertebrates have been poorly documented, but concern over potential damage to endangered mussel populations has influenced some treatments. A permit for treatment of Mystic Lake was initially denied due to concerns over possible Al toxicity to mussels, but after massive mussel mortality occurred following a cyanobacterial bloom, control of cyanobacteria was viewed more favorably and treatment was approved. The dose was limited to ≤50 g/m^2, and a pilot test in an area with surviving mussels was required to study direct impacts. No mortality or behavioral impairment for mussels in a zone treated at 50 g/m^2 was found (Nedeau 2011). No mussel mortality has been observed via visual surveys associated with treatments of Long, Lovell's, or Cliff ponds. Hamblin Pond contained no mussels at the time of the first treatment in 1995; physical habitat is quite suitable and nearby lakes have many mussels, so they may have been eliminated by algal blooms or other pre-treatment conditions. The invasive Asian clam (*Corbicula fluminea*) has been detected in the town swimming area of Hamblin Pond as of 2015, but no other mussels have been found.

The response of soft-bodied benthic invertebrates has not been quantitatively assessed in any of the treated Cape Cod lakes. Fishermen reported reduced insect hatches from Hamblin Pond following the 2015 treatment, suggesting a potential impact for at least 2 summers. Because all treated sediment of Cape Cod lakes experience anoxia, however, losses are likely limited to chironomids and other invertebrates tolerant of extremely low oxygen.

Zooplankton were monitored in some of the treated Cape Cod lakes and included a suite of cladocerans, copepods, and rotifers at highly variable abundance. Some of the treated lakes support herring runs but others do not. Where herring fry grow through the summer, zooplankton biomass peaked in winter and was minimal in summer. Winter zooplankton biomass in Mystic Lake, which has a herring run, was between 50 and 100 μg/L, whereas summer biomass was <20 μg/L (WRS 2011). Long Pond, which also has a herring run, had summer zooplankton biomass <10 μg/L (AECOM 2009). Lakes without herring runs exhibited variability of zooplankton biomass in relation to fish community structure and water quality, but data were insufficient to adequately examine and explain the variability.

Based on zooplankton samples from treated Cape Cod lakes (not all data are shown), the Al treatment initially depressed zooplankton abundance; the coagulation process may remove zooplankton as well as algae and other particulates. The second treatment of Hamblin Pond in 2015 severely depressed zooplankton biomass immediately following treatment, but biomass recovered substantially by June 2016 (Fig. 10). Biomass was also depressed following treatment of Lovell's Pond in 2014 (Fig. 11), but in 2015 and 2016 biomass recovered to concentrations higher than average for the 2013 pre-treatment study period.

Plants have generally been unaffected by Al treatment in Cape Cod lakes, based on our observations. Treatments target areas deeper than plants grow in these lakes, and many of the lakes have limited plant communities because of light limitation by algal blooms and coarse sandy sediment in the littoral zone. Few studies have been conducted, but quantification of the plant community in Long Pond before and after treatment revealed no appreciable change (AECOM 2009). Some colonization by macrophytes of shallow areas in Lovers Lake has been reported by residents, but quantitative data are lacking. The improved clarity in most lakes after treatment has not resulted in substantial expansion of plant growth. This lack of growth was most evident in Hamblin Pond, which was clear for more than 18 years after the first treatment and has a limited plant community.

Duration of treatment benefits

Only 2 of 10 treated lakes have been retreated: Hamblin after 20 years and Ashumet after 9 years. Hamblin Pond provides the longest and most complete record for Al treatment on Cape Cod. The first treatment was conducted in May 1995 over 70% of the lake area at a dose of 45 g/m^2 and provided rapid and lasting improvement in water clarity (Fig. 12) with elimination of cyanobacterial blooms. A slight decline in clarity may be perceived over 18 years following the first treatment, but clarity decreased abruptly in late August 2013 with a cyanobacterial bloom and remained low throughout the 2014 summer season. A second treatment was planned with an advanced approach that included extensive sediment testing to accurately identify areas in need of treatment and to set appropriate doses. Relative to the first treatment, a smaller portion (59%) of the lake was treated the second time, in mid-June 2015, with the target area divided into 3 zones receiving differential doses of 45, 58, and 71 g/m^2, which eliminated algal blooms and restored high water clarity. Watershed land use has changed little since the 1995 treatment.

The first Ashumet Pond treatment was limited in scope and area and was only part of the overall P control project. Only the deepest area was treated, with an expectation of improved conditions for up to 5 years, enough time to implement other P controls, especially an Fe-based geochemical barrier for inactivation of high P in a groundwater plume (AFCEC 2015). Monitoring into 2008 indicated increasing P concentration in deep water and high Chl-*a* in surface water, and a second treatment over twice the original area at a similar dose was applied in 2010. That second treatment has provided satisfactory results for about 6 years to date.

The next oldest treatment was at Long Pond in 2007, now 9 years old. The average monthly Secchi transparency depths for June through September were 60% to 100% greater than pre-treatment depths (Fig. 13). There was variation among months, but summer averages did not decline through 2015, 8 years post-treatment. Watershed land use has changed little since the 2007 treatment.

All other treatments are no older than 7 years (Table 2). Mystic Lake experienced increased clarity more gradually than the other treated lakes, likely related to slow flushing and the processing of a large organic P load from dead mussels prior to treatment that was not efficiently stripped from the water column during treatment (WRS 2011); summer P concentrations continued to decline for several years after treatment. Great Pond has shown little improvement, but the internal P load represented the smallest fraction of the TP load for all treated lakes (Table 1). Lovell's Pond has experienced declining clarity since the year after treatment for unknown reasons; Secchi transparencies were much higher than before treatment but were not

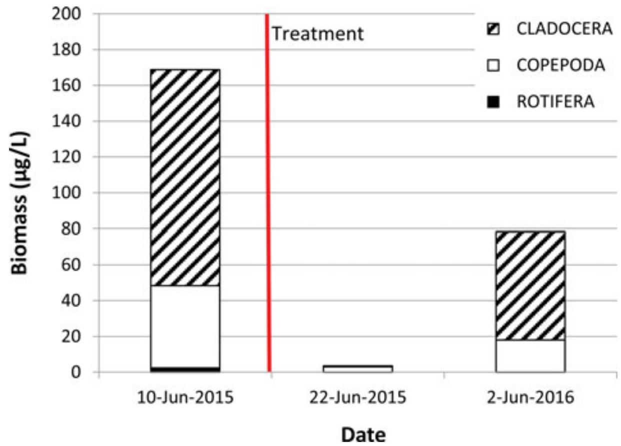

Figure 10. Zooplankton biomass in Hamblin Pond from June samples before and after treatment.

as stable as for other treated lakes. External inputs do not seem to have increased, so the dose may have been low or more area may have required treatment for sustained results. Biogenic P sources were possibly more important in Lovell's Pond than previously determined, and increased oxygen in deeper water after treatment may have promoted the release of organically bound P. The other 5 treated lakes experienced rapid and sustained clarity since treatment.

Costs

The cost of treatments varied mostly by area treated and dose but was also influenced by inflation, market swings, and changes in technology. The cost of Al chemicals was only moderately stable and represented about half the cost of most treatments. On a unit cost basis, considering the cost per gram of Al applied to a square meter of lake area and the total area of application, average cost for the chemicals and labor to conduct a treatment was about \$150/g/m^2 per ha in 2016 dollars. For example, treatment of 25 ha at a dose of 50 g/m^2 would cost about \$187,500. The cost of permitting and pre- and post-treatment studies was additional and has varied substantially but was much lower than the application cost.

Discussion

Projects on Cape Cod have furthered our knowledge of dosing to inactivate P in surficial sediment in stratified waterbodies with low alkalinity. As glacially created kettlehole lakes, the treated Cape Cod lakes have much in common, notably relatively long detention time, an infertile sandy bottom overlain in deeper water by organic material that can cause oxygen depletion, and high Fe concentrations that bind P under oxic conditions but release it under anoxia. These characteristics makes them ideal candidates for Al treatment, but concern over low alkalinity and the potential for Al toxicity if the pH moves outside the range of 6 to 8 standard units produced a cautious regulatory stance to this technique. Documentation of minimal or temporary adverse impacts to biota since 2001 has facilitated and expedited permitting by regulatory agencies.

An additional concern has been the addition of sulfur with aluminum sulfate because sulfur tends to bind with Fe and make it unavailable as a P binder, thus reducing the natural capacity to inactivate P in lakes. Although sulfur is added in these treatments, there is little indication it will lead to severe Fe shortages. Herring Pond does have some saltwater influence (average conductivity of 625 µS) and may well have an Fe shortage as a result of sulfur influx with seawater, but the amount of sulfur added through treatment would be minor in comparison. The other ponds are not influenced by saltwater (average conductivity <150 µS) and are continually supplied with considerable Fe through

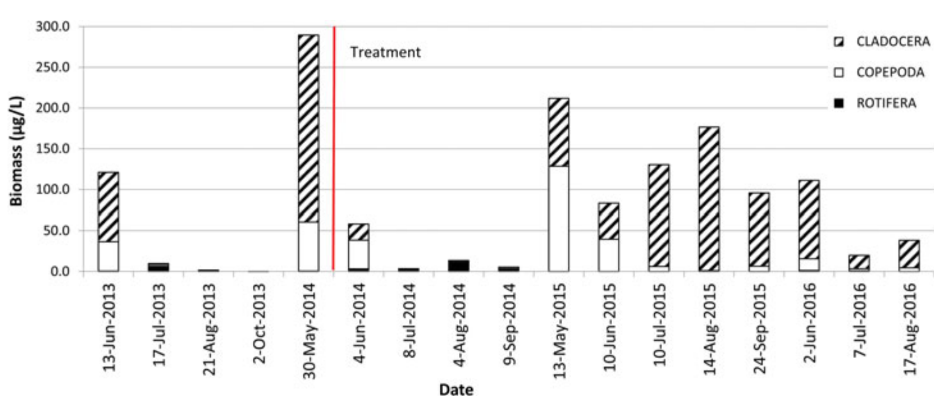

Figure 11. Zooplankton biomass in Lovell's Pond before and after treatment.

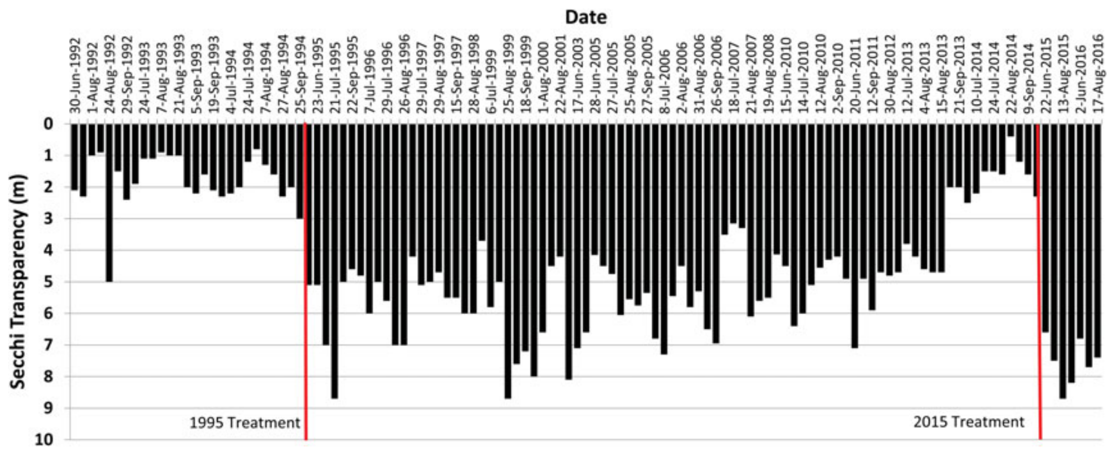

Figure 12. Summer Secchi transparency depths from Hamblin Pond between 1992 and 2016 through 2 aluminum treatments.

groundwater inflow, as evidenced from sampling in many of the same studies that established P loads and the importance of internal loading (e.g., BEC 1993, ENSR 2001, WRS 2014b). Hypolimnetic dissolved Fe levels frequently exceed 1 mg/L during late summer. Additionally, with the use of sodium aluminate to balance the pH from aluminum sulfate additions, about 54% less sulfur is added than if only aluminum sulfate was applied.

Consideration of the serious habitat and use impairment occurring in the absence of lake management has also convinced many towns and agencies on and off Cape Cod to consider Al treatment when internal recycling is scientifically established as a major P source. For example, Mattson (2015) reported a successful Al treatment to follow up cranberry bog management to achieve water quality standards in a lake

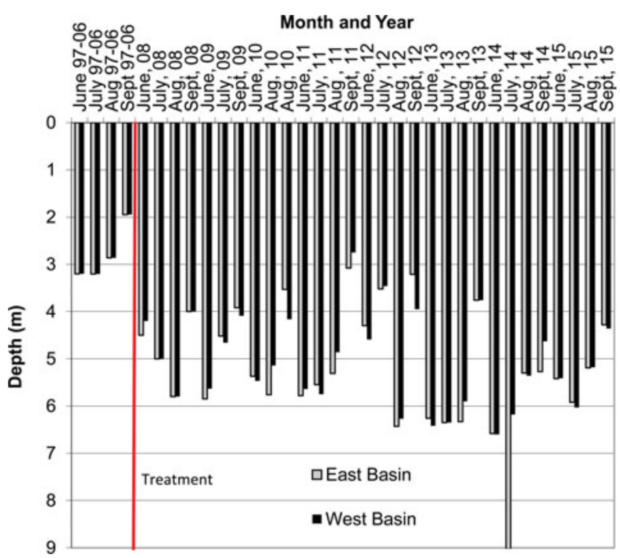

Figure 13. Summer Secchi transparency in 2 treated basins of Long Pond between 1997 and 2015.

between Boston and Cape Cod that was sponsored by the Massachusetts Department of Environmental Protection with Section 319 funds, a first for that agency. Both watershed and in-lake management were necessary to meet water quality goals, and work on Cape Cod helped shape that treatment. Certainly ecological risks are associated with Al treatment, but the potential benefits are substantial. This treatment is not a substitute for watershed management but a complement to it, much as bailing water out of a boat complements patching a leak; in many cases both may be necessary to achieve the desired result.

One major benefit of Al treatment is the differential reduction in available P versus N. Al treatment reduces the internal load of P but seems to have little effect on N concentrations in Cape Cod lakes (data not shown). Although reduction of both P and N may seem desirable, especially in a coastal environment like Cape Cod, greater reduction of P in a lake will result in higher N:P ratios and favor algae other than cyanobacteria (Smith 1983, Harris et al. 2014). Except for Great Pond, all the treated lakes experienced severe cyanobacterial blooms prior to treatment, and all showed not just a reduction in overall algal biomass but also in the portion of the community represented by cyanobacteria.

P concentrations were reduced by Al treatment to <17 µg/L but rarely <8 µg/L, the ecoregion threshold for preventing algal blooms in this area (USEPA 2014). The shift in nutrient ratios seems to favor a different algal community, one that is more easily assimilated into the pelagic food web and results in a lower standing crop of algae. A thorough investigation of complete food web effects is lacking, but any decrease in actual algal productivity may be countered by an increase in edible and nutritious algae rapidly assimilated into the food web and thus not measured as algal biomass. Low zooplankton biomass beyond initial reductions during treatment may be related to food limitation or predation. Budget cuts have minimized valuable fish surveys by the Division of Fisheries and Wildlife in Massachusetts since the 1980s, leaving most fishery effects to speculation.

An indirect benefit of Al treatment is reduced oxygen demand. Reduced algal biomass translates into less organic matter decomposing in the water column or on the bottom, and HOD has been substantially lowered by Cape Cod treatments. Prior to treatment, most of these lakes had no oxygen below the thermocline by sometime in July. Where the decrease in HOD is large enough and the depth is great enough, a zone just below the thermocline can remain oxygenated through the summer and support trout, which are stocked annually in many of these lakes. The ability to maintain holdover fish increases size and angling enjoyment.

The oxic zone below the thermocline may also be important to minimizing algal blooms by creating a zone in which P released from anoxic sediment recombines with Fe to precipitate and settle before it reaches the photic zone to fuel algal growth. Several of the treated lakes were known to have intense metalimnetic cyanobacterial blooms (especially *Planktothrix* in Mystic and Cliff) that mixed into upper waters during windy periods and were associated with toxic events. Others have experienced synchronous rises of cyanobacteria (*Dolichospermum lemmermannii* in Hamblin and *Microcystis aeruginosa* in Lovell's) from sediment in shallower portions of the hypolimnion exposed to anoxia. Lowering the anoxic interface within the hypolimnion seems to minimize development of such blooms.

Although lower oxygen demand is a benefit experienced by all treated lakes on Cape Cod, 4 of the treated lakes would still be classified as eutrophic based on Hutchinson's (1957) threshold of 550 $mg/m^2/d$ for HOD, and the others would be considered mesotrophic. Oxygen demand is still sufficient to cause some anoxia after treatment, so the overall metabolism of treated lakes was not changed nearly as much as increases in water clarity might suggest. Al treatments address the serious issue of excessive algal biomass and cyanobacterial blooms but are not completely restorative in terms of other eutrophication issues.

Al treatment addresses internal loading to reduce P availability and shift nutrient ratios to favor more desirable algae and improve food web function. Benefits may be compromised if external loading remains elevated (Brattebo et al. 2017), but where the internal load is the dominant P source and external loads are minimized, many years of improved water quality can be expected. Where internal load is the dominant P source, Welch and Cook (1999) suggested a reasonable expectation of 15 years of improvement for treated stratified lakes, and Huser et al. (2016b) documented an average duration of benefits of 15 years for all lakes and 21 years for stratified lakes. While all but 2 Cape Cod treatments occurred <10 years ago, and the first Ashumet treatment was not intended to provide long-term improvement by itself, results for Hamblin Pond are consistent with those estimates.

The reasons for eventual diminution of treatment benefits include ongoing P loading from the watershed, release of P through organic decay, bioturbation of treated sediment, and upward movement of Fe-P from beneath the inactivated zone (Huser et al. 2016b). In stratified lakes with small watersheds, no significant point sources, and anoxic hypolimnia, the upward movement of P through the inactivated zone is likely the primary cause of a return to significant internal P flux from the sediments. The progression at Hamblin Pond seems to exemplify this mechanism, although increased Fe-P possibly represents an accumulation from organic decay followed by precipitation with Fe. Only a gradual decline in water clarity was observed over almost 19 years (Fig. 12), after which conditions deteriorated quickly. Both watershed inputs and organic decomposition would not be expected to cause such rapid deterioration, and there are no carp or major populations of deep water invertebrates to cause bioturbation. Sediment assessment revealed elevated Fe-P concentrations in the upper 10 cm in the treatment zone 19 years after treatment. An assumed effective inactivation in the upper 10 cm suggests an upward movement of mobile P at an average rate of just >0.5 cm/yr.

Welch et al. (2017) report on inactivation of sediment P in Green Lake (Washington) to a depth well beyond 10 cm, but their comparison with other studies suggests 10 cm as the typical depth of inactivation in the absence of extreme bioturbation or wind driven resuspension. Neither of these mechanisms seems to be a major force in the treated portions of the Cape Cod lakes, where anoxia limits bioturbation and strong stratification resists mixing. Huser et al. (2016a) found that common carp greatly increased the depth of mixing and potential sediment P release in shallow lakes, but Bajer and Sorensen (2015) found no significant impact in a stratified lake. Doses for Cape Cod lakes were based on a target sediment depth of 10 cm.

The ratio of Al added to Al-P formed (by weight) varies inversely with P concentration (Huser and Pilgrim 2014, James and Bischoff 2015) and may be higher (less efficient) at high doses and where bottom slope is high (Huser 2012) but seems to average <20:1 in sediments over time (Rydin et al. 2000, Welch et al. 2017). Achieving a ratio of at least 10:1 for added Al to targeted P is a suggested threshold for extended treatment benefits (Jensen et al. 2015, Huser et al. 2016b). Most Cape Cod lakes have been treated at Al doses between 10 and 20 times the Fe-P in the upper 10 cm of sediment. Predicted Al:P binding ratios based on Huser's (2012) equation range from 6.7 to 32.8 for treated Cape Cod lakes, with an average of 16.1. One of the challenges in many treated lakes is steep bottom slope in some areas of application, focusing Al in deeper water sediments. Schütz et al. (2017) demonstrated the use of an injection system to apply Al to sediments, which might prove useful on slopes steep enough to cause focusing.

More research is needed to advance our capability to predict the duration of benefits from Al treatment, but one useful diagnostic approach is to examine slices of core samples over depths of up to 0.5 m to assess the vertical distribution of forms of P and to repeat this process over time following a treatment to evaluate redistribution and especially upward mobility of Fe-P. A chemical gradient is created by treatment, and anoxia within the sediment would be expected to allow dissociated Fe-P to move upward into the zone of lower Fe-P. This monitoring approach has been initiated on multiple Cape Cod lakes in the last few years.

Treatment timing may warrant further investigation. Spring treatments of Cape Cod lakes provided a greater reduction in TP versus pre-treatment values than fall treatments. Although reduced algae and increased clarity were observed the following summer, fall treatments exhibited TP reductions mostly below the threshold of ~60% reduction suggested by Huser et al. (2016b) as indicative of maximum longevity of benefits. All spring treatments exhibited TP reductions in excess of 60%. Inefficient stripping of the water column by fall treatment after a summer of P release from sediment may leave more P for future recycling. However, Mystic Lake was treated in the fall and showed continued improvement in clarity over several years as "leftover" P from a major mussel kill moved through the system. Also, by contrast, Lovell's Pond has experienced lower clarity and mild cyanobacterial blooms 2 and 3 years after a spring treatment. These examples indicate multiple factors are involved, and we have more to learn about the magnitude and timing of treatment before maximum performance can be achieved with Al treatments.

The interface between science (dose) and economics (cost) is an important consideration in most treatments. High doses inactivate more sediment P, but the binding efficiency is lower and the cost per unit of P inactivated will be higher. Certainly the work of Huser (2012) and Welch et al. (2017) and laboratory assays conducted for some of the Cape Cod treatments suggest that lower doses inactivate more P per unit of Al applied, but Jensen et al. (2015) caution against adding less Al than 10 times the amount of mobile P. Because upward migration of P through the inactivated zone may be an important mechanism for diminution of treatment benefits for Cape Cod lakes, the initial benefit from all treatments of adequate dose to inactivate the upper few cm of sediment should be the same, with higher dose treatments providing those benefits for a longer time period than lower doses. Beyond some site-specific minimum required dose that determines attainment of benefits, any additional Al would be expected to determine the duration of benefits, and the shape of the dose-response curve becomes important.

In some cases (e.g., Mystic Lake) the fraction of Fe-P inactivated in laboratory assays is highest at low doses then declines sharply at higher doses (Fig. 2a), asymptotically approaching the Fe-P detection limit. In other cases (e.g., Cliff Pond), the relationship is more linear (Fig. 2b), with only a slight increase in the amount of Al needed for each successive increment of Fe-P inactivation. Where laboratory assays have been used to aid dose determination, a target of between 50 and 100 mg/kg of Fe-P remaining after Al dosing is considered to signal satisfactory inactivation. Calculation of possible P release has indicated acceptable loading at <100 mg/kg by methods put forth by Pilgrim et al. (2007), and the asymptotic approach to the detection limit from values <100 mg/kg suggests that much higher Al doses will be necessary to reduce Fe-P further. Economics have sometimes dictated a slightly higher Fe-P target where the slope of the inactivation curve from laboratory assays declines sharply.

At the empirically derived cost of $150/g/m^2 per ha treated, costs rise proportionally to dose, but there may be an economy of scale based on treated area that is not obtainable in relatively small Cape Cod lakes. Conversion of unpublished data from other treatments (e.g., Grand Lake St. Mary's in Ohio) suggest that for treatments of large areas that yield favorable economies of scale, cost per unit area declines but will be no less than $100/g/m^2 per ha treated.

Treatment planning attempts to balance the minimum necessary reduction in internal loading with cost. Empirical models (e.g., Kirchner and Dillon 1975, Jones and Bachmann 1976, Larsen and Mercier 1976) are used to predict lake TP response to loading changes and translate these into expected Chl-*a*, Secchi transparency, and bloom probability (Oglesby and Schaffner 1978, Vollenweider 1982, Walker 1984). Possible target conditions (e.g., P concentration $<$20 μg/L, water clarity $>$3 m, probability of Chl-*a* $>$10 μg/L at $<$5%) must be evaluated in light of the potential to reach them with internal load reductions and the associated cost. For the Cape Cod lakes, we suggest doses based on a ratio of Al added to target P (by weight) $>$10:1, assay results with remaining Fe-P $<$100 mg/kg, and an internal load reduction of $>$75%.

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