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ABSTRACT

114 lakes treated with aluminum (Al) salts to reduce internal phosphorus (P) loading were analyzed to identify factors driving longevity of post-treatment water quality improvements. Lakes varied greatly in morphology, applied Al dose, and other factors that may have affected overall treatment effectiveness. Treatment longevity based on declines in epilimnetic total P (TP) concentration averaged 11 years for all lakes (range of 0–45 years). When longevity estimates were used for lakes with improved conditions through the end of measurements, average longevity increased to 15 years. Significant differences in treatment longevity between deeper, stratified lakes (mean 21 years) and shallow, polymictic lakes (mean 5.7 years) were detected, indicating factors related to lake morphology are important for treatment success. A decision tree developed using a partition model suggested Al dose, Osgood index (OI, a morphological index), and watershed to lake area ratio (related to hydraulic residence time, WA:LA) were the most important variables determining treatment longevity. Multiple linear regression showed that Al dose, WA:LA, and OI explained 47, 32 and 3% respectively of the variation in treatment longevity. Other variables (too data limited to include in the analysis) also appeared to be of importance, including sediment P content to Al dose ratios and the presence of benthic feeding fish in shallow, polymictic lakes.

1. Introduction

Excess phytoplankton biomass, caused by elevated levels of anthropogenically derived nutrients, is common in many limnic systems during the growing season. This condition often leads to degraded water quality, impaired aesthetics and recreation opportunities, odor problems, and byproduct formation during drinking water treatment. To address these problems, water quality standards related to nutrient status (water transparency and algal biomass) have been developed, requiring reductions in phosphorus (P, generally the limiting nutrient in lakes) and occasionally nitrogen. Even after external sources of P are reduced, however, accumulated legacy P in sediment can continue to drive algal blooms (Welch and Jacoby, 2001) and delay water quality recovery for decades (Sas, 1990; Jeppesen et al., 2005). Thus, the recycling of legacy P in the sediment must be considered when designing and implementing measures to meet water quality standards.

Aluminum (Al)-salts have been used to reduce P cycling in lakes around the world for nearly half a century (Landner, 1970; Kennedy et al., 1987; Cooke et al., 2005; Jensen et al., 2015). The success of past treatments has varied greatly, with studies showing longevity of water quality improvements ranging from months to 20 years (Welch and Cooke, 1999). While changes in nutrient-related surface water quality indicators are often determined following Al treatment (see review in Cooke et al., 2005), there are few quantitative analyses of the effects of Al dose, sediment mobile P (including pore-water, reductant soluble, and labile organic fractions), lake morphology, or other causal factors potentially affecting treatment longevity.

Multiple factors can positively (or negatively) affect longevity of improved water quality after Al addition to reduce internal P

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cycling. Water residence time, water column stability, and the relative magnitude of internal to external P loads can all affect the perceived effectiveness of internal P loading management (Sas, 1990). Al dose calculations based on lake water alkalinity, as opposed to sediment P pools that contribute directly to internal loading, affect treatment longevity due to the potential for over- or more likely under-dosing (Cooke et al., 2005). Furthermore, longevity of Al treatment in shallow, polymictic lakes is generally lower than in deeper, stratified systems (Welch and Cooke, 1999). It has been suggested that macrophyte presence (Welch and Kelly, 1990) and invertebrate bioturbation (Nogaro et al., 2006; Steinman and Ogdhal, 2012) can affect the efficacy of alum treatment in shallow systems, but recent work has shown that short-term binding efficiency between Al and P may be improved via increased mixing of the treatment layer (Huser et al., 2015). Shallow lakes, however, have received generally lower Al doses because alkalinity available to buffer acid producing Al hydrolysis reactions is limited simply due to the lower volume of water (i.e. alkalinity) available relative to sediment surface area.

Benthic feeding fishes, such as the common carp (*Cyprinus carpio*), have been shown to degrade water quality (Parkos et al., 2003; Weber and Brown, 2009) and deepen the sediment mixing zone, thereby increasing the mass of sediment P available for release to the water column (Huser et al., 2015). Aging and crystallization of newly formed amorphous $Al(OH)_3$ may reduce binding capacity, especially in the absence of P (de Vicente et al., 2008a). Steeply sloping lake bathymetry or large fetches can lead to focusing of added Al, increasing amounts of Al in one location and leading to crystallization in the absence of P (Huser, 2012; Egemose et al., 2013). Organic carbon (de Vicente et al., 2008b) and pH (Egemose et al., 2009) may also limit P binding by Al in lake water, but no significant relationship was found when comparing Al to Al bound P (Al:P_{Al}) ratios and organic matter content of sediment in Al treated lakes (Huser, 2012).

In order to determine factors related to longevity of water quality improvement in Al-treated lakes, we examined 114 lakes previously treated with Al to reduce internal P loading from the sediment. Both short-term post treatment changes and total longevity for total P (TP), water clarity (Secchi depth), and biomass of planktonic algae (as chlorophyll *a*; Chl *a*) were determined from available data. Lake chemical characteristics, Al dose, morphology, mobile sediment P, and other variables were then evaluated to determine the main factors influencing treatment longevity and effectiveness. A decision tree based on critical thresholds and a multiple linear regression model were developed for predicting the probable success of future Al treatments.

2. Methods

2.1. Data collection

The final dataset includes chemical data from readily available online databases collected during 2014 and early 2015, except where noted. Specific information relating to lakes included in this study is shown in Table 1 and further described below.

2.1.1. US lakes

The US lakes are temperate lakes located in Florida, Maine, Michigan, Minnesota, New Hampshire, Vermont, Washington, and Wisconsin. All lakes were treated with aluminum sulfate (Alum) or a combination of Alum and sodium aluminate (SAl).

2.1.2. Florida

The study lakes in Florida are moderately alkaline and a mix of polymictic and stratified systems. The lakes are generally small in size (mean area = 32 ha) and have relatively high surface water pH (mean > 8). Data were collected from the Water Atlas database hosted by the University of South Florida (USF, 2015) or the Southwest Florida Water Management District website (SWFWMD, 2015), where analytical methods can be found as well.

2.1.3. Michigan

The two lakes in Michigan are alkaline, non-humic, and have relatively high epilimnetic pH (mean > 8). Spring Lake is a drowned river mouth lake (444 ha), whereas Byram Lake is a small, dimictic lake (53 ha). Data availability was limited; during some years only two sample collections occurred and thus these two lakes were given lower weight during modeling (Supplementary Table 1). Analytical methods for the two lakes can be found at the Michigan Clean Water Corps website (MCWC, 2015) and as reported by Steinman and Ogdahl (2008).

2.1.4. Minnesota

The Minnesota lakes are generally clear water, non-humic, low turbidity lakes. Data were collected from the Minnesota Pollution Control Agency database (MPCA, 2015), except for Susan (Barr Engineering, 2013), Schwanz, Heine, Fish, Carlson, Blackhawk (City of Eagan 2015), Langdon and Long (Hennepin) lakes (MCWD, 2015), and Centerville Lake (Rice Creek Watershed District, 2015). Analytical methods are described at the respective websites. Lakes were sampled at least monthly and in most cases bi-weekly. Sediment data were either unpublished (Barr Engineering Co. unpublished) or from Huser et al. (2011) and Huser (2012). Benthic fish abundance data were obtained from the Minnesota Dept. of Natural Resources (MNDNR, 2015).

2.1.5. East Coast lakes

The Maine, New Hampshire, and Vermont lakes are low alkalinity, clear water lakes and range from polymictic to dimictic. Data were collected from the New Hampshire Dept. of Environmental Services (NHDES, 2015), Connor and Martin 1989, and the Maine Dept. of Environmental Protection (MEDEP, 2015) or obtained from the Vermont Watershed Management Division (VWMD, 2015). Analytical methods are available at the respective websites.

2.1.6. Washington

Washington lakes are generally low in alkalinity (except for Medical and Liberty lakes) and either polymictic or monomictic. Monitoring occurred throughout the entire year and data sources were different for most lakes (Supplementary Table 1). In some cases, data from the Dept. of Ecology website (Dept. of Ecology, 2015) were used if data were not available from local organizations (Supplementary Table 1). Analytical methods are listed at the above website and references contained in Supplementary Table 1. Sediment data are found in Rydin et al. (2000).

2.1.7. Wisconsin

The Wisconsin lakes are generally alkaline and are either polymictic or dimictic. The Wisconsin dataset includes some of the first Al treated lakes in the country (Horseshoe and Snake, 1970–1972). Water chemical data were collected from the Wisconsin DNR (WIDNR, 2014), the USGS (USGS, 2015), and from published studies (listed in Supplementary Table 1).

2.1.8. Denmark

The Danish lakes are temperate, either polymictic or dimictic, and are alkaline except for one, non-humic and shallow lake. Data were collected every second week or monthly during most of the year. The lakes were treated with poly-aluminum chloride (PAC) and in two cases treatments were buffered with SAl during the

Table 1
Al treatment information (dose based on mobile sediment P (sediment) or either alkalinity or water column P (water)), longevity, morphology (Osgood Index, Eq. (1)), watershed to lake area ratio (WA:LA) and mean water chemical data (2 years prior and 2 years post-treatment) for each individual lake during the growing season for stratified (A) and polymictic (B) lakes. Longevity estimates were determined using either Man Kendall trend analysis (*) or the MLR model developed herein (**).

Lake	State	Country	Treatment year	pH	Alkalinity (mg L^{-1})	Surface area (Ha)	Mean depth (m)	Al dose (g Al m^{-2})	TP longevity (Yr)	Osgood Index	WA:LA	Treatment method
A												
Lake Sønderby[a]	Fyn	Denmark	2001		150	8	2.9	31	17	10.3	15.4	Sediment
Lake Nordborg	Jutland	Denmark	2006	8.3	175	56	5.0	44	6	6.7	21.1	Sediment
Lake Vedsted	Jutland	Denmark	2009		15	7	5.0	26		18.9	0.6	Sediment
Lake Frederiksborg Castle	Sealand	Denmark	2005		125	22	3.5	10	1	7.4	20.2	Sediment
Lake Kollelev	Sealand	Denmark	2003	7.6	125	3	1.5	54		8.7	16.7	Sediment
Großer Weißer See	MeckPomm	Germany	2002			27	6.1	15	7	11.7	7.3	Sediment
Lake Tiefwarensee[a]	MeckPomm	Germany	2001–2005			141	9.6	59	25	8.1	14.5	Sediment
Flaten[a]	Stockholm	Sweden	2000			63	7.4	60	41	9.3	6.4	Sediment
Lejondal	Upplands-Bro	Sweden	1993			272	7.1	25	6	4.3	6.1	Sediment
Anderson	Florida	US	2006	8	38	5	3.9	22		17.4	18.1	Sediment
Arnold	Florida	US	1999			8	6.3	32	11	22.3		Sediment
Daniel	Florida	US	1998			3	3.6	14	3	20.3		Sediment
Gatlin[a]	Florida	US	2004–2005			25	4.8	24	17	9.7	1.9	Sediment
Mizell[a]	Florida	US	1997	8.7	39	24	3.4	45	45	7.0	4.4	Sediment
Silver	Florida	US	1998			28	15.7	30	9	29.7	10.4	Sediment
Tyler	Florida	US	2005			9	3.8	47		12.2		Sediment
Annabessocook	Maine	US	1978	7.7	15	575	5.2	22	13	2.2	9.5	Water
Chickawaukie[a]	Maine	US	1992	7.1	13	145	7.3	29	39	6.1	7.3	Water
Cochnewagon	Maine	US	1986	7	16	156	7.0	31	10	5.6	21.8	Water
Byram[b]	Michigan	US	1990	8.6	125	53	4.8	95	95	6.5	3.3	Sediment
Calhoun[a]	Minnesota	US	2001	8.5	98	180	10.6	42	33	7.9	6.7	Sediment
Carlson	Minnesota	US	1994	8.9	48	5	3.6	7	3	17.0	11.3	Water
Cedar	Minnesota	US	1996	8.8	85	68	6.1	27	11	7.4	11.7	Water
Fish Lake	Minnesota	US	2011	8.3	59	12	2.5	6	1	7.3	6.0	Water
Harriet	Minnesota	US	2001	8.6	106	138	9.8	32	4	8.3	24.4	Sediment
Heine	Minnesota	US	1996	8.7	56	3	3.0	6	3	16.6	1.3	Water
Langdon[a]	Minnesota	US	1998	8.6	106.7	58	2.5	70	26	3.3	7.3	Water
Long (Washington co.)[a]	Minnesota	US	2008–2009	8.2	100	24	3.4	109	16	6.9	53.4	Sediment
McCarrons[a]	Minnesota	US	2004	8.6	96.5	33	7.6	60	25	13.3	13.2	Sediment
Schwanz	Minnesota	US	1997	8.5	75	5	2.1	10	1	9.2	58.7	Water
Morey[a]	Vermont	US	1986	8.03	35	220	8.4	45	42	5.7	8.6	Water
Ballinger	Washington	US	1990	7.8		41	5.0	23	2	7.9		Water
Medical[b]	Washington	US	1977	9.25	142	64	9.7	122	113	12.1	5.4	Water
Pattison north	Washington	US	1983	7.9	52	33	4.3	31	2	7.5	8.8	Water
Bass Lake[b]	Wisconsin	US	1999			15	7.0	97	50	18.1	12.2	Sediment
Bullhead	Wisconsin	US	1978			27	4.0	42	13	7.7	9.5	Water
Horseshoe	Wisconsin	US	1970		230	9	4.0	10		13.4	78.7	Water
Mirror[b]	Wisconsin	US	1978			5	7.8	15	19	34.9	2.6	Water
Shadow[b]	Wisconsin	US	1978			17	5.3	9	6	12.9	3.3	Water
Silver	Wisconsin	US	2004			28	4.2	82	15	8.0	7.2	Sediment
Snake[a]	Wisconsin	US	1972		13	5	2.0	24	19	8.9	5.2	Water
B												
Lake Glumsø	Sealand	Denmark	2006		100	25	1.3	30		2.6	28.5	Sediment
Schwandter See	MeckPomm	Germany	2002			19	1.6	16	7	3.7	12.4	Sediment
Långsjön	Stockholm	Sweden	2006			29	2.2	75	8	4.1	8.4	Sediment
Banana Lake	Florida	US	2007	9.5	86	98	1.3	104	3	1.3	55.9	Sediment
Bay Lake	Florida	US	2006			15	2.3	20		5.9	6.4	Sediment
Conine[b]	Florida	US	1995			96	3	32	46	3.1	1	Sediment
East Lake. Tampa	Florida	US	1999.2001	8.2		40	1.7	30	4	2.6	11.4	Sediment
Three Mile	Maine	US	1988	7.2	12	259	5.2	20	4	3.2	9.3	Water
Spring	Michigan	US	2005	8.87	145	444	5.2	80	6	2.4	27.5	Sediment
Anderson SW	Minnesota	US	2012	7.8	104	23	1.2	51		2.6	8.1	Sediment
Blackhawk	Minnesota	US	1996	8.5	80	19	1.5	10	1	3.5	4.9	Water
Bryant[a]	Minnesota	US	2008	8.16	144	72	4.6	37	9	5.4	18.3	Sediment
Ceneterville	Minnesota	US	1998	8.7	134.5	200	3.7	18	0.5	2.6	0.9	Water
Clear	Minnesota	US	1988	8.18	141	263	4.1	33	9	2.5	5.8	Water
Isles	Minnesota	US	1996	8.4	75	42	2.7	18	4	4.2	7.1	Water
Kohlman	Minnesota	US	2010	8.18	112	30	1.2	78		2.2	101	Sediment
Long (Hennepin co.)	Minnesota	US	1996		160	115	4.3	26	0.5	4	28.8	Water
Olson	Minnesota	US	2005	8.5	79	81	2.1	8	0.5	2.4	23.1	Water
Powderhorn	Minnesota	US	2003	7.87	86	5	1.2	45	6	5.7	25.7	Sediment
Rebecca	Minnesota	US	2011	8.4		106	4.3	81		4.1	4.7	Sediment
St. Clair	Minnesota	US	1998			65	1.5	26	2	1.9	46.1	Water
Sunfish	Minnesota	US	2008	8.74		25	1.2	8	0.1	2.4	8.5	Sediment
Susan	Minnesota	US	1998	8.3	130	36	3	30	2	5.1	27.3	Sediment
Kezar	New Hampshire	US	1983–1984	6.5	4.8	74	2.8	24	2	3.3	37.8	Water
Campbell	Washington	US	1985	8	85	150	2.4	26	7	2	7.1	Water
Erie	Washington	US	1985	8.91	85	45	1.8	20	14	2.7	7.9	Water
Green	Washington	US	1991	7.4	24.7	100	3.9	34	0.5	3.9	7.6	Water

Table 1 (*continued*)

Lake	State	Country	Treatment year	pH	Alkalinity (mg L^{-1})	Surface area (Ha)	Mean depth (m)	Al dose (g Al m^{-2})	TP longevity (Yr)	Osgood Index	WA:LA	Treatment method
Green[a]	Washington	US	2004	7.4	24.7	100	3.9	96	16	3.9	7.6	Sediment
Liberty	Washington	US	1980–1981		110	288	5.2	52	14	3.1	11.8	Water
Liberty	Washington	US	1974		110	288	5.2	5	0.5	3.1	11.8	Water
Long south (Thurston Co.)	Washington	US	1983	8.2	55	60	3.8	28	5	4.9	7.3	Water
Long south (Thurston Co.)	Washington	US	2008	7.8	55	60	3.8	54		4.9	7.3	Sediment
Long north (Thurston Co.)	Washington	US	1983	8	55	70	3.6	28	12	4.3	6.3	Water
Long (Kitsap co.)	Washington	US	1980		25	140	2	11	4	1.7	6.7	Water
Newman	Washington	US	1989			515	5.6	15	1	2.5	15.1	Water
Pattison south	Washington	US	1983	8.4	58	77	4	31	0.5	4.5	4.9	Water
Phantom	Washington	US	1990	8		24	6.4	28	12	13.1	8.2	Water
Wapato	Washington	US	1984	7.7		12	1.5	12	0.1	4.3	30.4	Water
Bear Trap	Wisconsin	US	1998			96	3.3	35	3	3.4	42	Sediment
Delevan	Wisconsin	US	1991	8.5	230	725	6.4	19	2	2.4	14.5	Water
EauGalle	Wisconsin	US	1986	8.9	132	60	3.2	12	0.1	4.1	276.7	Water
Wind	Wisconsin	US	1998			374	5.8	13	0.1	3	27.1	Sediment

[a] Treatment longevity estimated using Seasonal Mann Kendall Sen's slope ($p < 0.05$).
[b] Treatment longevity estimated using the MLR model developed herein.

period 2001–2009. Data and analytic methods are either unpublished or included in (Reitzel et al., 2005; Egemose et al., 2011; Jensen et al., 2015).

2.1.9. Germany

The German lakes are situated in the federal state Mecklenburg-West Pomerania (Northeast Germany). These hard water lakes were formed following the last continental glaciation 12,000 years ago and are located within a transition zone between temperate-maritime and temperate-continental climates. Due to intensive agriculture in the catchments, about 80% of lakes have a very high trophic state. The monitoring of selected lakes was carried out according to European Water Framework Directive guidelines. Details about Lake Tiefwarensee are presented in Mehner (Mehner et al., 2008; Wauer et al., 2009).

2.1.10. Sweden

The Swedish lakes are non-calcareous, clear water, low turbidity and situated in the Stockholm area on granites and gneisses covered by glacial and post-glacial clay deposits. Lejondalssjön (dimictic) had PAC applied to the bottom water, while Flaten (dimictic) and Långsjön (polymictic) had PAC injected directly into the sediment. Water column data were generally collected monthly. Analytical methods are described by Stockholm City (Stockholms stad, 2015) and Upplands-bro county (Upplands-bro 2015).

2.2. Data handling and statistics

Treatment longevity for all nutrient related water quality variables was calculated using a minimum post-treatment improvement of 50% (either a reduction of epilimnetic TP and Chl *a* or an increase in Secchi depth) compared to a minimum of 2 years (within 5 years prior to treatment) of pre-treatment growing season data (May–September). Treatment longevity was defined as the time between treatment and the last year of 50% or greater improvement that preceded at least two successive years (to account for extreme years) of less than 50% improvement. We used this approach because lake-specific management targets were often not available. The method is likely conservative because treatment longevities for lakes that were included in both this and a similar study conducted by Welch and Cooke (1999) were the same or slightly lower using the 50% improvement for determining treatment longevity. In cases where specific goals were set that were lower than state defined water quality standards and improvement was less than 50% compared to pre-treatment conditions, these goals superseded the 50% improvement criteria (Lake Calhoun, Cedar Lake, and Lake McCarrons, Minnesota).

When treatment longevity exceeded the data record (i.e. improvement remained 50% or more over pre-treatment conditions in the last observation year available), a Seasonal Mann–Kendall test was used to determine if significant, monotonic trends ($p \leq 0.05$) could be determined (Loftis et al., 1991). The Sen's-slope (unit yr^{-1}) trend estimator was used to estimate longevity for each lake based on an average of the final three years of data for each lake (Huser et al., 2012). For cases where positive treatment effects continued through the end of the dataset and no significant trend was detected, the lake was excluded from model development and then longevity was estimated using the model developed herein (Medical, Byram, Bass, Conine, Mirror, and Shadow, Table 1). Longevity in these lakes was then estimated using the regression model developed herein (see below). In one case hypolimnetic P was used instead of epilimnetic P due to substantial external loading reductions directly before Al treatment (Annabessocook, Maine).

Because some lakes were recently treated and longevity could not be estimated, two-year pre- and post-treatment means for TP, Secchi depth, and Chl *a* were calculated for all lakes. Data were either log transformed (TP and Chl *a*) and standardized through calculation of Z-scores, or Z-score standardized (Secchi depth), to account for skew in the data and allow for between lake comparisons. Z-scores were calculated for each observation as the difference between the observed value and the mean value for that variable across the four-year monitoring period for each lake, divided by the standard deviation. Means for water chemistry (pH, alkalinity) used in modeling were calculated using the four years of data closest to treatment (2 years before and 2 years after treatment) to account for conditions pre- and post-treatment for each lake.

Statistical analyses were conducted in JMP (SAS Institute Inc., version 11.0.0). Before modeling, all predictor variables, except percentage internal P load (pre-treatment), were log transformed to improve normality. Variables with high bivariate correlations (pH and alkalinity as well as Al dose and Al:mobP) were analyzed separately to reduce problems of multi-collinearity. Stepwise multiple linear regression (MLR) was conducted using forward selection of variables that most improved fit. Model fitting ended when significance of additional variables was greater than 0.05 and

Table 2
Explanation of different potential confounding factors and the weights given to each lake based on these factors.

Weight	Description
5	Good data series (at least monthly data) Minimal changes to P loading besides Al treatment
4	Minor reductions to external P load (10–25%) Final longevity estimated
3	Poor quality pre- or post-treatment data (e.g. less than monthly but consistent data) Moderate external P load reduction (25–50%)
2	Major external load TP reduction (>50%) or major in-lake changes (e.g. dredging)
1	Missing years in data record

explained variation was less than 2%. Because Chl *a* and Secchi depth data were more limited than those available for TP, models for longevity were developed using only TP data. Model development was also conducted after excluding lakes with moderate to high biomass densities of common carp estimated using a relationship between trap net catch rates, mean fish weight, and biomass densities of carp based on electro-fishing mark and recapture (Supplementary Table 1; Bajer and Sorensen, 2012). Results were verified using a threshold capture rate greater than 0.6 fish per net, which has been shown to correlate with degraded water quality (Weber and Brown, 2011).

A decision tree identifying critical thresholds for predicting treatment longevity was developed using a partition model. A partition model (also known as a regression tree) recursively performs dichotomous splits according to a relationship between predictor and response variables by finding a set of cuts or groupings of predictor values that minimize within group variance. Partitioning trees have been successfully used to identify potential causal relationships in a variety of environmental datasets (Dobbertin and Biging, 1998; Rothwell et al., 2008). All r^2 coefficients of determination were adjusted for the number of explanatory terms by using the degrees of freedom in each model, unless otherwise indicated.

A weighting factor (1–5, with 1 being the lowest weight) was used to control for other factors that may affect calculation of treatment longevity (Table 2). The weighting factor scales a lake contribution to the loss function by $(\text{weight})^{-0.5}$ using least squares estimation. Thus, rows with minimal potential confounding factors contribute more to the loss function. For example, lakes with less than monthly monitoring data (but still seasonally consistent) or lakes with a moderate reduction in external P loading (25–50%) were given a weight of 3, but lakes having a complete dataset (at least monthly data) and minimal changes to other potential P sources, were given a weight of 5 (Supplementary Table 1).

2.3. Sediment analysis

Sediment from some of the study lakes ($N = 44$) was analyzed in previous studies (see Supplementary Table 1) for P fractions determined using the Psenner et al. (1988) wet sediment P extraction technique as modified by Hupfer et al. (1995), except for Florida lakes where a modified Chang and Jackson (1958) extraction scheme was used. An Al to mobile sediment P ratio (Al:mobP) was estimated using the mass of Al applied to the lake and the mass of mobile (pore-water, loosely bound, and reductant soluble) P that directly leads to internal sediment P release (Pilgrim et al., 2007) in the upper 6 cm of sediment. Al dose calculations were generally based on sediment depths from 6 to 10 cm for available data, thus a sediment depth of 6 cm was used to standardize ratio estimates, as information was generally not available for depth of the active sediment layer.

2.4. Calculated variables

The Osgood index was used to assess the effects of lake morphology on treatment longevity:

$$\text{Osgood Index} = \text{Zm} \Big/ (\text{A})^{0.5} \quad (1)$$

where Zm is the mean water column depth (m) and A is the surface area of the water body (km^2). An Osgood Index of 6 generally represents the point where lakes are stratified or polymictic, with lower ratios being polymictic (Osgood, 1988). Watershed to lake area ratios (WA:LA) were used to assess the influence of hydrology on treatment longevity because water residence time was available for only 32 lakes. WA:LA ratios correlated negatively and significantly with residence time ($r^2 = 0.39$, $p < 0.0001$).

3. Results

Of the 114 lakes in the database, only 83 lakes had sufficient data to be included in the analyses. Both lake surface area (3–725 ha) and WA:LA ratios varied great for the lakes in the dataset (Table 1). Both soft and hard water lakes were included and mean growing season pH in the epilimnion ranged from 6.5 to 9.5. The dataset was nearly evenly split between stratified ($N = 41$) and polymictic ($N = 42$) systems and between lakes with Al doses calculated using sediment mobile P (Rydin and Welch, 1999; Huser and Pilgrim, 2014; $N = 42$) and non-sediment based methods (Kennedy and Cooke, 1982; Kennedy et al., 1987; $N = 41$).

Average longevity of TP reduction for all lakes was 11 years (15 years using modeled longevities, see below), whereas Secchi disk and Chl *a* based longevities were 9.7 and 8.8 years, respectively. Deeper, stratified lakes had an average treatment longevity based on TP of 15 years, ranging from 0 to 45 years (113 estimated maximum), whereas shallow, polymictic lakes had an average TP-based treatment longevity of 4.6 years, ranging from 0 to 14 years (16 estimated maximum). Mean treatment longevities for improvements in water clarity (as Secchi depth) and Chl *a* were similar to TP-based values for shallow lakes, but longevity of improvement for both Secchi depth and Chl *a* were less in stratified systems (13 years for both) when compared to TP-based longevity (Table 3). When modeled longevities for TP reduction were used for the six lakes with continued improvement through the end of the dataset but without significant increasing TP trends (see below), treatment longevity in deep and shallow lakes increased to 21 and 5.7 years, respectively.

3.1. Short-term pre- and post-treatment effectiveness of Al treatment

Significant ($p < 0.0001$) differences in Z-scores were detected for pre- and post-treatment average TP, Secchi depth, and Chl *a* (Fig. 1). Paired t-tests also showed TP decreased significantly from a pre-treatment average of 0.101 to 0.036 mg L^{-1}. Chl *a* also decreased, dropping from a pre-treatment average of 42.7 to 16.3 $\mu\text{g L}^{-1}$ and Secchi depth increased from 1.6 to 2.4 m. Percent change after treatment was greater for both TP and Chl *a* for lakes that were dosed according to sediment-based methods but only the difference in TP response was statistically significant ($p < 0.05$). No significant difference in post-treatment Secchi depth response was detected between sediment and non-sediment based dosing methods.

3.2. Threshold development for factors relating to treatment longevity

To help determine factors controlling longevity of TP reduction and to identify critical thresholds, a decision tree partition model was developed (N = 68 lakes). A dose of 15.1 g Al m^{-2} was the first threshold (Fig. 2), above which mean treatment longevity for TP improvement was approximately 8 years. From this group, lakes having OI greater than 5.7 had a mean treatment longevity of 15 years. The next threshold was a WA:LA of 8.8; for lakes below this level, treatment longevity increased to 26 years. Finally, the lakes from this group that also had an Al dose of greater than 28 g Al m^{-2} had an average longevity of 36 years. On the other side of the partition tree, lakes receiving an Al dose lower than 15.1 g Al m^{-2} and having a WA:LA ratio greater than 27 had less than one year of reduced TP concentration. Further splits did not improve predictability and the final partition model had an r^2 of 0.69.

When the Al:mobP ratio was substituted for Al dose, the critical thresholds differed somewhat. OI was the first threshold split (5.5), above and below which mean longevities were 9.7 and 2.3 years, respectively (Supplementary Fig. 1). The second split was at Al:mobP = 9.9, above which longevity averaged just over 12 years. The third threshold split was at WA:LA = 9.5, below which longevity averaged 22 years. Lakes with an OI less than 5.5 and Al:mobP ratio less than 10.5 had an average longevity of less than 1 year. Further splits did not improve model prediction ($r^2 = 0.44$), likely due to the low number of lakes with adequate information for the included variables (N = 34).

3.3. Predicting longevity in lakes treated with aluminum

Stepwise MLR was conducted using either Al:mobP or Al dose, pH or Alkalinity, OI, WA:LA and percent internal P load as candidate predictors. Only Al dose, WA:LA, and OI were significant ($p \leq 0.05$). Al:mobP was also significant, but because of the low number of lakes with information on sediment P-fractions and the other model parameters, Al dose was used instead. A MLR model was developed using Al dose, WA:LA, and OI that explained 62% of the variation in TP treatment longevity among the study lakes (Table 4). An obvious group of lakes with poorly predicted model results was detected; these lakes had an OI < 6 with populations of benthic feeding fish (Fig. 3). An alternate model was developed omitting these lakes that explained 82% of the variation in treatment longevity with Al dose, WA:LA, and OI explaining 47, 32 and 3%, respectively (Table 4, Fig. 3). The model was applied to the six lakes where treatment longevity could not be estimated because

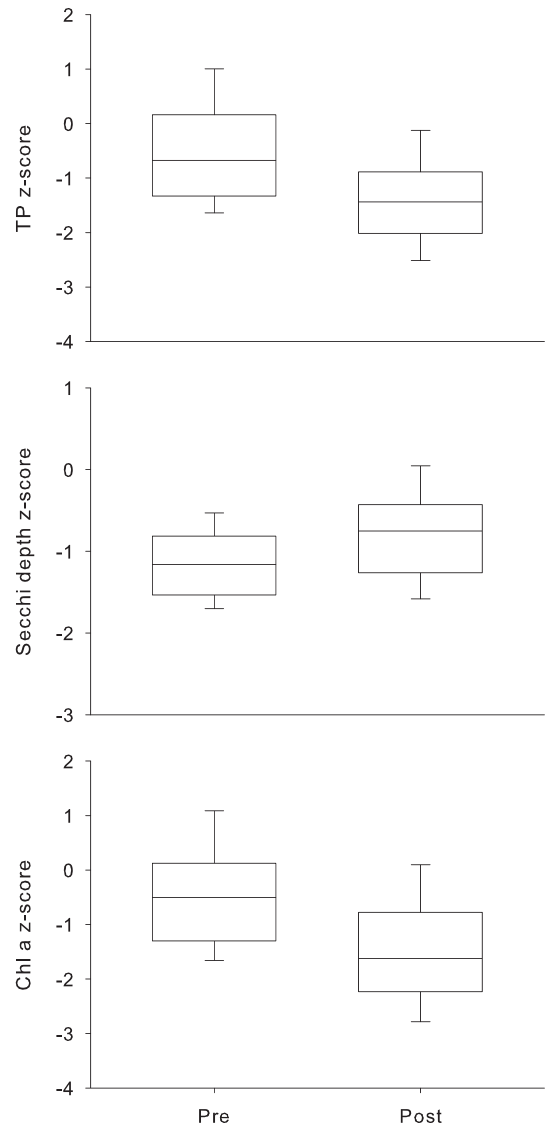

Fig. 1. Box plots for z-scores (TP and Chl *a*) and standardized z-scores (Secchi depth) for two-year pre- and post-treatment growing season means. All differences were significant ($p < 0.0001$).

Table 3
Means for water chemical data, Al dose, lake morphology, and hydrology for the study lakes. Numbers in parentheses represent means including longevities (6 lakes) estimated using the MLR model.

Variable	Units	N	All lakes			Stratified	Polymictic
			Mean	Min	Max	Mean	
Al dose	g Al m^{-2}	83	36	5.2	122	39	33
TPlong	Year	68(74)	11(15)	0	45(113)	15(21)	4.6(5.7)
SDlong	Year	55	9.7	0	27	13	6.5
Chlalong	Year	42	8.8	0	27	13	4.5
pH Epi		51	8.2	6.5	9.5	8.3	8.2
pH Hypo		40	7.3	6.7	8.9	7.2	7.5
Alkalinity	mg L^{-1} $CaCO_3$	49	87	4.8	230	82	91
Lake area	ha	83	96.2	3	725	63.9	128
Mean depth	m	83	4.4	1.2	15.7	5.5	3.2
OI	m km^{-1}	83	7.4	1.3	34.9	11	3.6
WA:LA		79	18.7	0.6	276.7	13	24
Int P load	%	34	41.5	3.1	88	42	41
Al:mobP		44	25.2	1.6	107.4	27	24

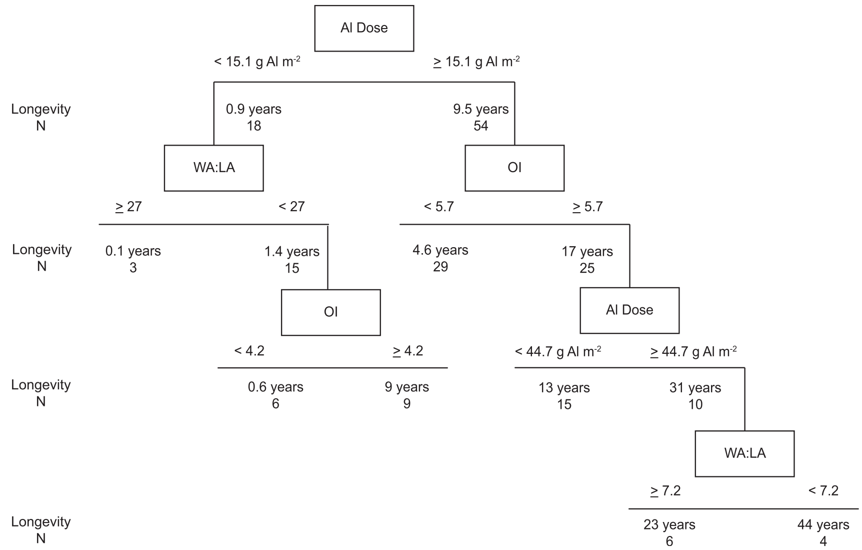

Fig. 2. Partition model ($r^2 = 0.69$) with threshold values for longevity of reductions in TP concentration. N = number of lakes.

improved conditions still existed throughout the period of observation and there was no increasing trend for epilimnetic TP (Table 1). Estimated longevities ranged from 6 years (Shadow) to over 100 years (Medical).

The two-year post treatment TP response to treatment was related to treatment longevity (Fig. 4). Lakes with longevities of 40 years or more had two-year mean TP reductions of at least 70%, whereas lakes with less than a 20% reduction all had longevities of less than three years. In between these two boundaries there was more variability. Obviously no or negative improvement during the first two years resulted in even lower longevity (i.e. less than one year) but even some lakes having an average improvement of 50% or more for TP after treatment had longevities as short as two years. Some of these lakes are infested with carp, and others were likely influence by high WA:LA, low Al doses, or other factors limiting treatment as discussed below.

4. Discussion

The addition of P-binding metals to sediment is a common management technique used to reduce sediment P cycling and restore water quality in lakes (Cooke et al., 2005). Although such techniques have been used for decades, reports of variable results have led to uncertainty in current applications (Mackay et al., 2014; Spears et al., 2014). To improve predictability, we quantitatively defined the physical and chemical factors that relate to longevity of improved water quality conditions after addition of Al to lake sediment.

Table 4
TP longevity model estimates and variable interaction predictors for all lakes, and for lakes with medium to high densities of carp excluded.

Model	Term	Estimate	p
Carp lakes included	Intercept	−1.3	<0.001
	Log(Al dose)	1.4	<0.0001
	Log(WA:LA)	−0.59	<0.0001
	Log(OI)	0.75	<0.01
Carp lakes excluded	Intercept	−0.50	<0.05
	Log(Al dose)	1.3	<0.0001
	Log(WA:LA)	−0.79	<0.0001
	Log(OI)	0.37	<0.05

The lakes included in this study ranged from small to large with widely varying morphology, hydrology, water chemistry, and applied Al doses. Improvements in water quality were not detectable after treatment in some cases, whereas in other cases improvements lasted decades (Table 1). Of the predictor variables included in this study, WA:LA ratio, Al dose, and lake morphology (OI) appeared to drive both short-term changes in water quality and treatment longevity. The ratio of Al dose to sediment mobile P was also a significant explanatory variable, but the low number of lakes for which sediment P-fractions were available limited its usefulness here.

4.1. *Short-term pre- and post-treatment effectiveness of Al treatment*

Approximately 90% of the study lakes showed some water quality improvement in the two years following Al treatment (Supplementary Table 1). This was shown by using both a comparison of z-scores, which reduced variability between treatments (Fig. 1), and in pairwise testing of percent change between pre- and

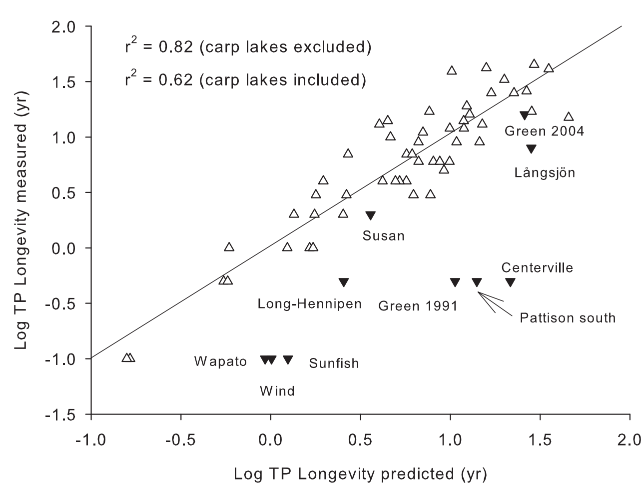

Fig. 3. Measured versus modeled treatment longevity based on TP concentrations. Filled triangles represent lakes with medium to high densities of common carp. Only the regression line for the model where carp lakes were excluded is shown.

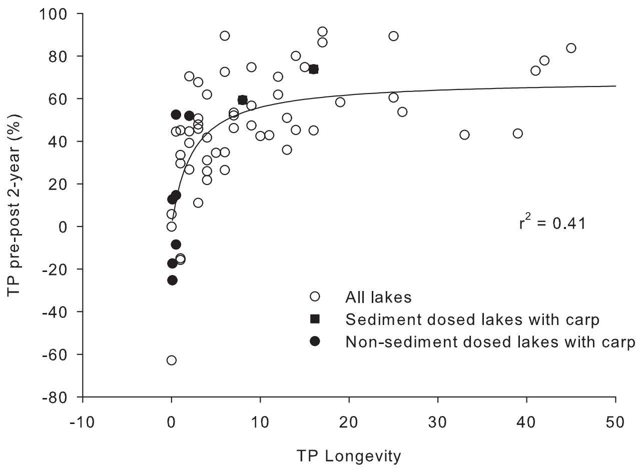

Fig. 4. Comparison of the percentage change between two-year averages for pre- and post treatment TP concentrations and longevity of treatment (TP). The best model fit was found using a Langmuir equation ($r^2 = 0.41$). Solid squares and circles indicate shallow lakes with carp and where Al dose was calculated either using sediment or non-sediment based methods, respectively.

post-treatment trophic state indicators for each lake. Dosing method appeared to have some effect when comparing pre- and post-treatment data. Al doses (51 versus 23 g Al m^{-2}), as well as percentage improvement in TP were greater in lakes where Al doses were calculated using mobile sediment P ($p < 0.05$) than for other methods. However, pre- and post-treatment mean Secchi depth and Chl *a*, were not significantly different between the two dosing methods. This result is not entirely surprising because other factors besides P may control phytoplankton growth, including nitrogen, inorganic particulate matter in the water column caused by bioturbation (Breukelaar et al., 1994), wind induced sediment resuspension (Hilton et al., 1986), and trophic structures leading to imbalances in grazing pressure on phytoplankton (Horppila and Nurminen, 2003).

4.2. Critical thresholds and factors controlling Al treatment longevity

Al dose (15 g Al m^{-2}) was the first threshold in the decision tree model (Fig. 2). That is not surprising, as more Al will bind more P. OI, however, had a break point of 5.7, very close to the value of 6, below which lakes are generally considered weakly stratified (e.g. Osgood, 1988). With an OI greater than 5.7, mean treatment longevity increased from 8 to 15 years. Thus, greater water column stability aided treatment longevity. Other factors including lake shape and fetch also affect water column stability, but the fact that the threshold developed herein is very close to the empirical results of other studies is interesting. Previous research suggested that lakes with an OI less than 6 would have greater transport of sediment derived P to the epilimnion and thus show the greatest effect of Al treatment on Chl *a* and Secchi depth (Cooke et al., 1982). Both resuspension and elevated pH in shallow lakes can increase the likelihood for release of sediment P, but can occasionally limit treatment effectiveness as well (Egemose et al., 2009; Reitzel et al., 2013). Furthermore, lakes with OI values greater than 6, and even much greater, can have vertical P transport similar to shallower lakes with weaker stratification (Mataraza and Cooke, 1997).

WA:LA ratio was also an important threshold value, with lower values correlating to greater treatment longevity. For lakes having WA:LA ratios less than 8.8, average longevity increased to 26 years. Low WA:LA ratios indicate both a longer residence time and a higher percentage of P load from internal sources, both of which would increase the influence of sediment P sources on productivity and water quality (Welch and Jacoby, 2001). On the other hand, productivity in lakes with higher WA:LA ratios tends to be driven by external P sources, even when internal sources of P are high (Vollenweider, 1975; James et al., 1991).

When Al:mobP ratios were used instead of Al doses for modeling, the first threshold in the partition model changed to an OI of 5.5, similar to that for the Al dose based model. The next threshold split was an Al:mobP ratio of 9.9, above which longevity averaged 12 years. This threshold was similar to the Al:mobP dosing ratio suggested as important for long term success of Al treatment (Jensen et al., 2015). It is also similar to binding ratios between Al added and P_{Al} in lake sediments after Al treatment (Rydin and Welch, 1999; Rydin et al., 2000; Huser et al., 2011; Huser, 2012; Jensen et al., 2015).

Surprisingly, pH was not an important factor in the partition model given that the soluble anionic form of Al dominates above pH 9 (Stumm and Morgan, 1996), and it has been suggested that a sustained pH at this level can affect P release from sediment and effectiveness of Al treatment (Welch and Cooke, 1999; Niemisto et al., 2011; Reitzel et al., 2013). Although it is likely that sustained high pH at the sediment–water interface will limit treatment effectiveness in lakes, available data suggested that the mean bottom water pH near the sediment surface of the study lakes was nearly a unit lower than surface water pH (8.2 versus 7.3, Table 3). The lower pH near the sediment surface is likely due to sediment buffering processes such as alkalinity generation via nitrate and bicarbonate reduction (Gahnström, 1985) and light limitation of phytoplankton production. These factors can limit pH deviation from neutral conditions, helping to sustain the chemical conditions needed for binding between Al and P (Huser and Rydin, 2005). Furthermore, effective Al treatment should reduce lake water pH as algal driven productivity and CO_2 consumption will decline after treatment. Even if Al-bound P is released due to re-suspension into high pH water during periods with limited through flow, when pH drops $Al(OH)_3$ will precipitate and can adsorb even more P than was initially released (Reitzel et al., 2013). This may explain the low Al:P_{Al} binding ratio of ~2 in Süsser See, Germany (Lewandowski et al., 2003).

The same variables from the partition analysis were important in MLR modeling, and Al dose was the most important factor controlling treatment longevity. Sediment Fe and Al content generally controls P cycling in lakes (Mortimer, 1942; Kopacek et al., 2007) and excess inputs have even led to lower P in the hypolimnion compared to the epilimnion in stratified lakes (Huser et al., 2011; Hu and Huser, 2014). WA:LA ratio, a proxy for water residence time, was nearly as important as Al in the model. As noted above, a high WA:LA ratio will limit the effect of internal P loading due to short water residence time and a relatively higher proportion of P input from external sources (Vollenweider, 1975), whereas a low WA:LA ratio means internal cycling of P likely plays a more important role in in-lake nutrient dynamics and primary production (Welch and Jacoby, 2001).

4.3. MLR model application

Of the six lakes to which the MLR model was applied to estimate longevity (6–113 years), Medical Lake had the highest Al dose (120 g Al m^{-2}) and a WA:LA ratio less than a third of the mean (5.4 vs. 18.7), resulting in the highest predicted treatment longevity (113 years). Because this case likely represents near optimal conditions for Al treatment, it is perhaps not surprising that beneficial effects are expected to last so long. Medical Lake received direct inputs of municipal wastewater for decades prior to treatment. Even after this load was controlled, 60% of the lake volume was anoxic during summer (Bauman and Soltero, 1978) and massive cyanobacterial blooms occurred regularly (Kettelle and Uttormark, 1971). After Al treatment, over 7 g of P_{Al} m^{-2} was detected in the sediment, which was generally an order of magnitude higher than other Al treated lakes in the region (Rydin et al., 2000). Both epilimnetic and hypolimnetic TP decreased after Al treatment, from growing season averages of 0.45 and 1.3 mg L^{-1}, to 0.044 and 0.11 mg L^{-1} when comparing pre-to post-treatment two-year means, respectively. It should be noted that aeration was initiated 10 years after Al treatment to improve the conditions for game fish and prevent a return of internal P loading. Aeration is believed to have had only a minor influence on internal P cycling. Average hypolimnetic P concentrations decreased significantly from 0.13 to 0.051 mg L^{-1} after aeration was initiated (Soltero et al., 1994), but they returned to just over 0.10 mg L^{-1} by 1989 and have stabilized between 0.1 and 0.2 mg L^{-1} since then.

4.4. Other factors potentially affecting longevity of treatment

Several lakes had significantly lower measured treatment longevities compared to model predictions. All of these were shallow and had at least some evidence of moderate to high carp densities (Fig. 3). Sediment disturbance by benthic invertebrates or bottom feeding fishes (e.g. *C. carpio* and bream, *Abramis brama*) can affect water quality (Breukelaar et al., 1994; Driver et al., 2005), and more importantly for Al treatment longevity, the depth of the sediment mixed layer. Huser et al. (2015) showed that moderate densities of large carp increased the sediment mixed layer from 5 cm without carp, to 16 cm with carp. Because the depth of sediment interacting with the water column increased, the amount of mobile sediment P potentially available for release to the water column also increased.

Green lake has high carp densities and visible feeding pockets of up to 10 cm sediment depth (Seattle Parks and Recreation, 2005; Dugopolski et al., 2008). Green lake received two Al treatments; the first (1991) based on alkalinity and buffering, and the second (2004) based on mobile P in the upper 10 cm of sediment. The longevity of the first treatment was substantially less than predicted (Fig. 3), whereas longevity (estimated) of the second treatment was near the predicted value. This shows the importance of dosing Al relative to mobile sediment P to an adequate depth when benthic feeding fish are present. Lake Susan, another lake with moderate densities of carp, had a predicted longevity only slightly above actual. The lake had the highest OI of the carp excluded lakes (5.1) and was partially stratified during most years (Bajer and Sorensen, 2014). Bajer and Sorensen (2014) suggested that benthic feeding fish play a relatively minor role in nutrient availability in deeper lakes like Susan that become stratified during the growing season, because no effect was seen on TP after carp removal from this lake in 2009.

Young benthic fish (i.e. before they switch to foraging for food in the sediment) can also affect perceived treatment effectiveness. Lakes with high densities of young carp and bream (or other zooplanktivorous fish) often have high resilience against water clarity improvements due to the influence on pelagic trophic structure of the youngest year classes predation on zooplankton (e.g., Jeppesen et al., 2012). This means that even if lake water TP decreases after Al treatment, there may be little or no improvement in Chl *a* or Secchi depth. One such example is Lake Kollelev, which after Al addition showed a strong response in TP but no improvement in Secchi depth until fish removal occurred two years later (Jensen et al., 2015).

Al treatments have been short-lived in a few shallow lakes (Cooke et al., 2005) and the inclusion of OI in both the decision tree and MLR models also indicates shorter treatment longevity in shallow lakes. OI, however, explained only 3 percent of across lake variation in longevity. The weak relationship between OI and treatment longevity suggests vertical P transport may have been similar in both weakly and strongly stratified lakes (Mataraza and Cooke, 1997). The OI may also be an indirect indication that shallow lakes are more likely to be affected by other processes that can affect water quality and the translocation of P from sediment to water, including sediment disturbance by benthic feeding species (Breukelaar et al., 1994), macrophyte growth and lower Al doses (Welch and Cooke, 1999). Unfortunately, data needed to adequately assess how such factors may affect longevity of Al treatment in the study lakes were lacking in most cases.

4.5. Modeling limitations

Several assumptions in the modeling procedure may have added uncertainty to the results. Estimates of carp biomass density (low to high, Supplementary Table 1) were based on only two lakes that had the necessary gill or trap net data and quantitative determinations for fish biomass. Trap net data for the shallow lakes in the current study ranged from 0.2 to 3.4 fish per net with a range of mean fish weight of 1.4 kg–2.9 kg, respectively. All lakes excluded from the final MLR model were above the 0.6 fish per trap net suggested by Weber and Brown (2009) to negatively impact water quality in lakes, suggesting that carp also negatively affect water quality in the lakes included herein.

The sediment depth of mobile P used herein (6 cm) may not be an adequate predictor of the mass of mobile P potentially available to the water column for all lakes. The active sediment layer is variable and can be affected by e.g., stratification, internal waves and benthic feeding fish (Rydin and Welch, 1999; Lewandowski et al., 2003; Huser et al., 2015). The fractionation method used may also cause differences in what is termed the 'mobile' P pool, given that all such methods are operationally defined. In addition, recent research has shown that the inclusion of labile organic P in Al dosing methods may increase longevity of treatment (Jensen et al., 2015), indicating the importance of this fraction when using Al to immobilize potentially available P in the sediment. The weighting factors used in this study may also add some uncertainty to model predictions, but even if the weighting system was removed from the model entirely, the r^2 of model fit decreased from 0.82 to 0.80.

4.6. Implications for future management

Clearly, correct Al dose calculation and application technique are prerequisites for lasting improvements in water quality (Brattebo et al., 2015). Therefore, lake managers need good estimates for the mobile sediment P pool and appropriate in-lake treatment conditions. The WA:LA (a proxy for water residence time) also had a strong influence on treatment longevity, and applying a loading/retention model (e.g. Vollenweider, 1975) should allow for prediction of post treatment lake water TP independent of WA:LA. This will, however, require a good knowledge of P and water inputs to the lake.

Most reports on lake restoration successes and failures cite a lack of sufficient understanding of the system as the main issue leading to perceived failure of a restoration project. To minimize the likelihood of failure, it is important to combine both high quality monitoring data and expertise in lake functioning. Unfortunately for the present study, monitoring was in some cases highly variable with limited pre-treatment data and intensive monitoring occurring for only a few years after treatment. This is the main reason more than 30 Al treatment cases were excluded from the study. Even in lakes where basic water chemical data are available, other potential treatment limiting factors are generally neglected (sediment P-fractions, macrophytes, fish community), limiting post-treatment analysis. Finally, other factors (e.g. actual P and water balances and trophic interactions) should be incorporated into future monitoring efforts to determine the factors controlling longevity of lake restoration measures. This may be difficult, however, until adequate funding is provided allowing monitoring programs to 'catch-up' with these needs.

5. Conclusions

- Clear differences were seen between pre- and post-treatment growing season means for TP, Secchi depth, and Chl *a*.
- Threshold criteria for treatment longevity indicated the importance of Al dose, watershed to lake area ratio, and lake morphology as important factors for treatment success
- The above three variables explained 82% of the variation in treatment longevity based on post-treatment changes in TP concentration.
- The presence of moderate to high densities of benthic feeding fish negatively affected treatment longevity, but the effects were weak in lakes that stratify or those where Al doses were based on mobile P in at least the upper 10 cm of sediment.
- Adequate monitoring programs are needed for future treatments in order to better assess the multitude of factors that may affect longevity of previous treatments and to improve future treatment outcomes.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.watres.2015.06.051.

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