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SUMMARY

The purpose of this project was to evaluate the release of dissolved phosphorus (P) from bottom sediment at Lake Fayetteville, and the potential use of aluminum sulfate ($Al_2(SO_4)_3$) to remediate the P stored and released by bottom sediments. Intact sediment cores (n=18) were taken at three locations, named inlet, mid and dam sites at Lake Fayetteville. The cores were incubated with 1 L of overlying water with light excluded and bubbled with air (half, aerobic treatment) and N_2 (other half, anaerobic). Water samples were pulled and analyzed for soluble reactive P (SRP), and that water was replaced with filtered lake water with SRP less than the lab's method detection limit (MDL, ≤ 0.005 mg L^{-1}). The SRP mass accumulating in the overlying water was used to estimate SRP release rates from the sediment, and mean rates were compared by treatments, sites and before and after alum dosing. Sediment SRP release rates were significantly greater under anaerobic conditions (mean=7.22 mg m^{-2} d^{-1}) than aerobic (mean=0.85 mg m^{-2} d^{-1}), and within those conditions rates were not different between sites. The addition of alum to the overlying water reduced SRP concentrations near the MDL in most cores, and sediment SRP release rates were significantly less after alum dosing, except for the cores from the mid lake site under aerobic conditions. Overall, it likely that this internal SRP source is an important factor in the development and occurrence of harmful algal blooms (and likely microcystin production) at Lake Fayetteville. Alum might be a means to successfully reduce this internal SRP source.

KEY WORDS: Harmful Algal Blooms, Sediment Phosphorus Release, Lake Management, Alum

CORE IDEAS

Despite little to no dissolved phosphorus supply in the water, sediments have the ability to release lots of phosphorus into the overlying water.

Sediment phosphorus release from lake-bottom sediments is still an important piece of the watershed management puzzle.

Sediment treatment with alum would reduce internal phosphorus.

INTRODUCTION

Sediment phosphorus (P) sources can fuel algal growth in inland waters, when nitrogen in not limiting (Austin et al., 2020), and algal P demand can drive sediment P release in eutrophic waters (McCarty, 2019). This internal P source may result in seasonal nitrogen (N) limitation of harmful algal blooms (HABs; Ding et al., 2018), especially when episodic N inputs cease during prolonged droughts. Nitrogen availability to HABs can limit or trigger production of toxins like microcystin (Wagner et al., 2020). However, dual N and P management of internal and external sources will likely be needed to control HABs in lakes and protect downstream waterbodies (Jankowiak et al., 2019; Paerl et al., 2016).

If internal P sources are a potential threat to water quality and HABs driver, then the first step is quantifying release rates from bottom sediments under aerobic and anaerobic conditions. The goal of this study is to understand the importance of sediment P release rates at Lake Fayetteville, which experiences annual HABs. The specific objectives were to estimate sed-

iment P release rates in intact sediment-water cores under aerobic and anaerobic conditions from three sites, treat the overlying water with aluminum sulfate ($Al_2(SO_4)_3$, hereafter alum) to reduce dissolved P, and then evaluate the influence of alum on sediment P release rates. This information should help inform the City of Fayetteville on the importance of sediment P release (or internal P sources) and possible management option to remediate this water quality problem.

MATERIALS AND METHODS

Lake Fayetteville is the surface impoundment of interest in northwest Arkansas, where cyanobacterial HABs have occurred resulting in public advisories being issued. In 2019, microcystin concentrations were measured up to 15 μg L^{-1} at the lake (Wagner et al., 2021), which was above the recommended guidelines for recreational contact waters (8 μg L^{-1}, EPA, 2019). Since the Arkansas Water Resources Center (AWRC) began routine monitoring at this lake in December 2018, microcystin has been observed in measurable concentrations (i.e., greater than typical reporting limits, 0.3 μg L^{-1}) throughout the year (Haggard, B. E., unpublished data). Lake Fayetteville is an important recreational re-

source for the City of Fayetteville, including almost 80 ha of water surface and 185 ha of land around the lake (City of Fayetteville, 2021).

In July 2020, a total of 21 sediment cores were collected from Lake Fayetteville from the main channel near the dam, mid-lake and inlet using a UWITEC corer (Figure 1). Plexiglas tubes (length, 0.6 m) were inserted approximately 0.3 m into the sediments, and then the cores were capped on the bottom and stoppered on top. A properly collected sediment-water core had relatively undisturbed sediment at the surface and through the core with relatively clear overlying water.

Upon return to the AWRC water quality lab, the depth of the water and sediments (if needed) were adjusted so that each core had one L of overlying water. Six cores per site (18 total) were wrapped to exclude light and incubated at room temperature (~22°C). The overlying water of each core was bubbled with air for 24 h, then half the cores per site were continued to be bubbled with air (aerobic conditions) while the other half were bubbled with N_2 (anaerobic conditions). The cores were incubated for a month (from July 22, 2020 to August 24, 2020), and alum was added to each core at 23 d into the incubation.

Figure 1. Sediment coring sites at Lake Fayetteville, July 2020 (image from Google Earth, May 6, 2021)

Optimum alum doses were based on a rate of 10:1 Al:P ratio where the P was based on the site average of the SRP release rates estimated across all cores and conditions, and the specific doses were 0.5X optimum, optimum and 2X optimum (see results for alum mass used). Three cores, i.e. one from each site, were incubated at room temperature in the window pane with exposure to natural light.

Water samples (~50 mL) were removed from the overlying water of each core (including those in the window pane exposed to natural light) at almost daily intervals for the first week and then every other day throughout the incubation. The sample was filtered (0.45 µm), acidified with HCl to pH<2, and then analyzed for SRP at the AWRC certified water quality lab. The overlying water in the cores was maintained at a volume of one L using filtered lake water with SRP concentration near method detection limits (MDL, SRP≤0.005 mg L^{-1}).

Sediment P release rates (mg m^{-2} d^{-1}) were calculated as linear changes in SRP mass over eight days during the incubation (mg d^{-1}) divided by the inside core area (0.005 m^2). The SRP mass was corrected for withdrawal and addition of SRP during sampling. In general, the first eight days were used because that represented the linear increase from the start of the incubation and then following alum addition. Analysis of variance was used to evaluate whether sediment SRP release rates were different between sites and treatment (aerobic verse anaerobic), as well as before and after alum addition at sites within a treatment (P≤0.05).

RESULTS

Sediment Core in the Window Sill

Initial SRP concentrations in the overlying waters across all of the collected sediment cores were low (≤0.007 mg L^{-1}), matching that typically observed in the lake water (Haggard, B.E., unpublished data). Two of the three cores sitting in the window sill exposed to light showed increasing SRP concentrations through 6 d (up to 0.179 mg L^{-1}), and then decreased below 0.007

mg L^{-1} through 12 d of the incubation; SRP remained low through the rest of the incubation in these cores. SRP concentrations in the third core in the window were always less than the MDL (≤0.005 mg L^{-1}) throughout the incubation. The addition of alum to these cores at any rate did not influence SRP concentrations, because the measured SRP concentration in the cores incubated in the window sill had concentrations already below the MDL. However, it is important to note that the overlying water of cores exposed to sunlight have little to no SRP in the overlying water likely due to algal uptake.

Sediment Cores under Aerobic Conditions

SRP concentrations in the overlying water of the cores incubated under aerobic conditions (i.e., bubbled with air) slowly increased through the first 8 d (Figure 2), and then leveled off in some cores, continued to increase in others and even slightly decreased in others. The minimum SRP concentration in overlying water of the aerobic treatments was 0.018 mg L^{-1} from 8 to 23 d, and the maximum was 0.058 mg L^{-1}. The average SRP in the overlying water across all cores, sites and from 8 to 23 d was 0.028 mg L^{-1}. The range in average SRP from 8 to 23 d across the aerobic treatments was 0.022 to 0.038 mg L^{-1}, marking likely dissolved P equilibrium between water and sediments.

The mass of SRP accumulating in the overlying water significantly (R^2≥0.88, P<0.01) increased over time the first 8 d of the incubation across all aerobic cores (Table 1). This resulted in SRP release rates that were not significantly different between the inlet, mid and dam sites at Lake Fayetteville (Figure 3A). The SRP release rates ranged from 0.46 to 1.20 mg m^{-2} d^{-1} across all cores where the overlying water was bubbled with air (i.e., aerobic conditions), and the mean was 0.88, 0.69, and 0.99 mg m^{-2} d^{-1} of the cores from inlet, mid and dam sites, respectively.

At 23 d, the overlying water of all cores was treated with alum, where the doses ranged from 0.225, 0.450 and 0.900 g alum per core across the replicates at each site. The SRP conc-
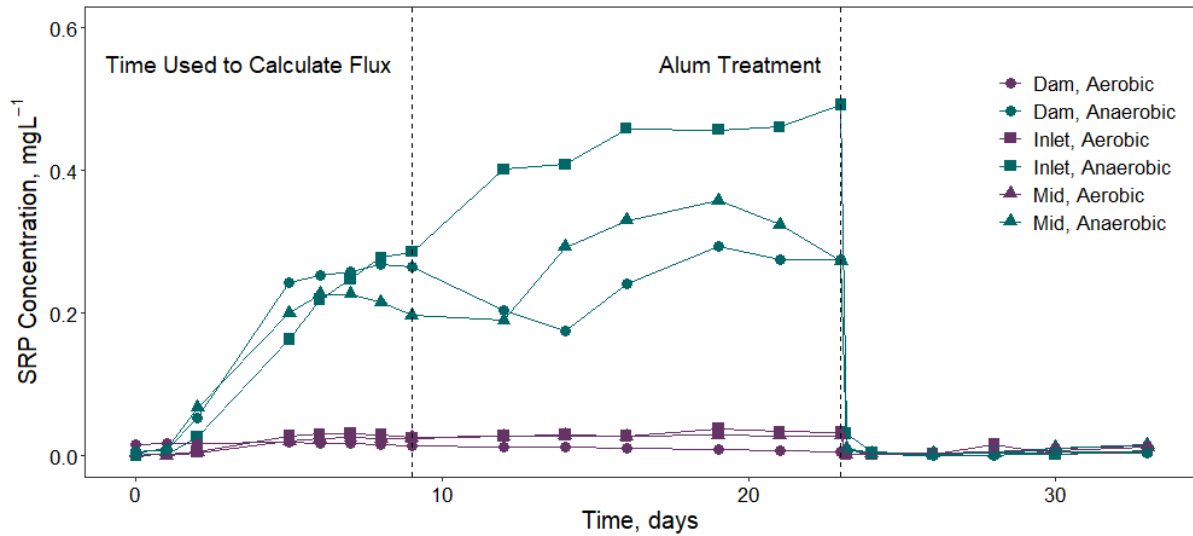
Figure 2. Soluble reactive phosphorus (SRP) concentrations in the overlying water of cores incubated under aerobic and anaerobic conditions from three sites at Lake Fayetteville (dam, inlet, and mid-lake) over time; approximately 8 d were used to calculate SRP fluxes, and the aluminum sulfate (alum) treatment occurred at 23 d.

entrations in overlying water of all cores incubated under aerobic conditions dropped to below the MDL after alum treatment (≤0.003 mg L^{-1}, Figure 2). The SRP release rates were significantly less under aerobic treatment following alum treatment, except at the mid lake site (Figure 3B). The SRP release rates following alum treatment were near zero and not significantly different between sites, where release rates from individual cores ranged from -0.26 to 0.33 mg m^{-2} d^{-1} (Table 1).

Sediment Cores under Anaerobic Conditions

On the other hand, SRP concentrations in the overlying water of the cores incubated under anaerobic conditions (i.e., bubbled with N_2) increased to 0.254 mg L^{-1} on average, ranging from 0.130 to 0.398 mg L^{-1} across all these cores (Figure 2). SRP concentrations in the overlying water continued to increase in some cores, while concentrations in the overlying water of other cores leveled off and even decreased in the later part of the 23 d incubation. The maximum SRP concentration in the overlying water of the anaerobic cores was 0.727 mg L^{-1} between 8 and 23 d.

The mass of SRP accumulating in the overlying water significantly ($R^2 \geq 0.81$, $P < 0.01$) increased over time in the cores the first 8 d of the incubation under anaerobic conditions (Table 1). The rate of increase, or the slope of these relations, was much greater under anaerobic conditions compared to aerobic. The SRP release rates were not significantly different across the inlet, mid or dam sites averaging 7.61, 5.15, and 7.70 mg m^{-2} d^{-1}, respectively (Figure 3A). The SRP release rates ranged from 3.56 to 11.27 mg m^{-2} d^{-1} across all cores under anaerobic conditions, where this minimum and maximum were both from cores collected at the dam site.

At 23 d, alum treatment at the same doses were effective at reducing SRP in the overlying water of the cores incubating under anaerobic conditions, almost immediately knocking SRP down to 0.061 mg L^{-1} or less (Figure 2). The next day after alum treatment SRP concentration in the overlying waters was less than the MDL on average (0.004 mg L^{-1}) across all cores, and the maximum concentrations was more than an order of magnitude less (0.010 mg L^{-1}) than prior to alum dosing. SRP concentration in some anaerobic cores started slowly increasing, but

Table 1. Soluble reactive phosphorus (SRP) slopes, R^2, P-values, and fluxes for each core at the inlet, mid-lake, and dam at Lake Fayetteville, pre-aluminum sulfate (alum) and post-alum treatment; average SRP fluxes are shown below each site and treatment.

Core	Site	Treatment	Pre-Alum				Post-Alum			
			Slope, mg d^{-1}	R^2	P value	SRP Flux, mg m^{-2} d^{-1}	Slope, mg d^{-1}	R^2	P value	SRP Flux, mg m^{-2} d^{-1}
1	Inlet	Aerobic	0.006	0.90	<0.01	1.13	0.000	0.37	0.20	-0.03
2	Inlet	Aerobic	0.003	0.89	<0.01	0.59	0.001	0.16	0.44	0.29
3	Inlet	Aerobic	0.005	0.96	<0.01	0.91	-0.001	0.29	0.27	-0.26
Average SRP Flux, Inlet, Aerobic						0.88				0.00
4	Inlet	Anaerobic	0.052	0.97	<0.01	10.35	-0.002	0.37	0.20	-0.35
5	Inlet	Anaerobic	0.022	0.93	<0.01	4.45	-0.003	0.19	0.39	-0.56
6	Inlet	Anaerobic	0.040	0.97	<0.01	8.04	-0.002	0.36	0.21	-0.41
Average SRP Flux, Inlet, Anaerobic						7.61				-0.44
7	Mid	Aerobic	0.005	0.94	<0.01	0.91	0.001	0.49	0.12	0.19
8	Mid	Aerobic	0.004	0.91	<0.01	0.71	0.002	0.96	<0.01	0.33
9	Mid	Aerobic	0.002	0.88	<0.01	0.46	0.000	0.04	0.72	0.02
Average SRP Flux, Mid, Aerobic						0.69				0.18
10	Mid	Anaerobic	0.032	0.97	<0.01	6.43	0.000	0.00	0.91	-0.02
11	Mid	Anaerobic	0.026	0.81	<0.01	5.15	0.002	0.36	0.21	0.41
12	Mid	Anaerobic	0.037	0.93	<0.01	7.43	-0.001	0.51	0.11	-0.18
Average SRP Flux, Mid, Anaerobic						6.33				0.07
13	Dam	Aerobic	0.005	0.92	<0.01	0.97	0.000	0.04	0.72	0.01
14	Dam	Aerobic	0.006	0.91	<0.01	1.20	0.001	0.54	0.10	0.20
15	Dam	Aerobic	0.004	0.95	<0.01	0.80	0.000	0.37	0.20	-0.04
Average SRP Flux, Dam, Aerobic						0.99				0.06
16	Dam	Anaerobic	0.057	0.94	<0.01	11.27	-0.002	0.37	0.20	-0.50
17	Dam	Anaerobic	0.018	0.96	<0.01	3.56	0.001	0.40	0.18	0.19
18	Dam	Anaerobic	0.042	0.91	<0.01	8.29	0.000	0.60	0.07	-0.03
Average SRP Flux, Dam, Anaerobic						7.70				-0.11

the rate of increase was not significantly different than zero (P≥0.07). Alum treatment significantly reduced SRP release rates across all sites under anaerobic conditions (Table 1, Figure 3C).

APPLICATION OF RESULTS

Like many other reservoirs, Lake Fayetteville accumulates and stores large amounts of the P inputs from its watershed (approximately 90% of the P inputs; Grantz et al., 2014). This stored P accumulates in the bottom sediment, and then it can be released by two mechanisms: sediment water equilibrium and reductive dissolution.

These two processes dominate under the different conditions that the lake expresses at the sediment-water interface both spatially and temporally.

Sediment equilibrium P concentrations (EPC$_0$) are the concentrations at which dissolved P in the overlying water are in dynamic equilibria with sediments, that is the net adsorption of SRP to the sediments and release from the sediments is zero (Froelich, 1988; Haggard et al., 1999). This is the process that likely results in the sediment P release rates in our aerobic cores, and it is likely that sediment EPC$_0$ at Lake Fayetteville is ~0.03 mg L^{-1} based on these
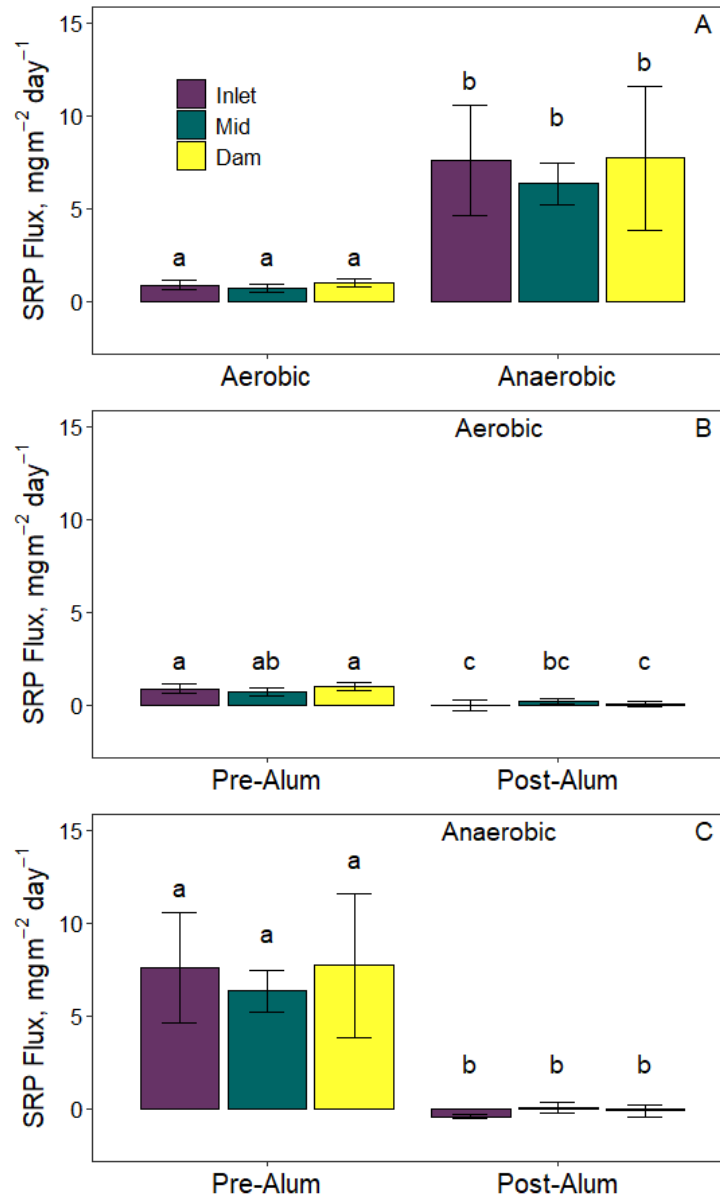
Figure 3. Soluble reactive phosphorus (SRP) fluxes in sediment cores under aerobic and anaerobic conditions from three sites at Lake Fayetteville (dam, inlet, and mid-lake) prior to aluminum sulfate (alum) treatment (A), SRP fluxes under aerobic (B) and anaerobic (C) conditions before and after alum treatment across the sites.

incubation. This would mean that the sediments would try to release SRP into the overlying water until that EPC_0 is reached, resulting in the measured sediment P release rates (0.85 mg m^{-2} d^{-1}, on average) under aerobic conditions at Lake Fayetteville. However, we do not typically see SRP in the lake water overlying the sediments

typically reach the EPC_0 because of algal uptake (McCarty, 2019; Austin et al., 2020). So in theory, bottom sediments at Lake Fayetteville are continually leaking out SRP trying to reach equilibrium with the overlying water (i.e., EPC_0).

Table 2. Average soluble reactive phosphorus (SRP) fluxes under aerobic and anaerobic conditions at local reservoirs measured using intact sediment cores or hypolimnion accumulation over the summer.

Reservoir	Average SRP Flux, mg m^{-2} d^{-1}		Source
	Aerobic	Anaerobic	
Lake Eucha, OK	1.03	4.40	Haggard et al. 2005
Lake Frances, OK	4.00	15.00	Haggard and Soerens, 2006
Lake Wister, OK	1.13	3.30	Haggard et al. 2012
Lake Tenkiller, OK	1.20	13.77	Lasater and Haggard, 2017
Lake Elmdale, AR		5.21[*]	Grantz et al. 2014
Lake Fayetteville, AR		5.48[*]	Grantz et al. 2014
Lake Wedington, AR		1.37[*]	Grantz et al. 2014
Beaver Lake, AR (Transition and Riverine Zones)	0.50	1.19	Sen et al. 2007

[*]SRP fluxes estimated using accumulation of SRP in the hypolimnion, whereas other studies were conducted using methods similar to that in this study.

Most reservoirs like Lake Fayetteville, especially deeper ones, stratify thermally – that is, warm water sits in the upper layers (i.e., epilimnion) and colder water sinks into the lower layers (i.e., hypolimnion). This happens in Lake Fayetteville each year, where stratification starts at the beginning of the growing season (e.g., April) and thermal stratification strengthens throughout the summer. When this happens, the dissolved oxygen is depleted in the lower water (i.e., hypolimnion) shifting the sediments into anaerobic conditions. This is where reductive dissolution is the primary mechanism responsible for SRP release from the sediment, and this is what we measured in the lab. Sediment SRP release rates are usually several times greater under anaerobic conditions than aerobic in eutrophic to hypereutrophic lakes (e.g., see Table 2 giving SRP release rates from local reservoirs), and the rates were almost ten times greater under anaerobic conditions (7.22 mg m^{-2} d^{-1}, on average) than aerobic in the cores from Lake Fayetteville.

The thermal stratification at Lake Fayetteville separates the upper (i.e., epilimnion) and lower (i.e., hypolimnion) layers with a thermocline (e.g, where the big change in water temperature is). In theory, the [upper and lower] layers don't mix across the thermocline, so the SRP concentrations build up in the lower layers like we observed in the overlying water of the cores incubated under anaerobic conditions. The SRP in the lower layers can diffuse to the upper layers across the thermocline due to the gradient in SRP concentrations, or the fact that SRP is negligible in the upper layers and much greater in the lower layers during stratification. So, SRP can slowly move from the lower layers when released under anaerobic conditions to the upper layers.

The thermocline in shallow reservoirs is not always stable under extreme weather events with wind and potentially large amounts of rain resulting in runoff from the watershed. Under these extreme weather conditions, the thermocline breaks down and allows the upper and lower layers to potentially mix a little bit before the thermocline sets back up. Currently, how often this occurs at Lake Fayetteville is not known, but the current research platform

installed by Baylor University, in collaboration with the AWRC, will help us answer this question (Figure 4). This platform allows very frequent measurements of water temperature and dissolved oxygen with depth at Lake Fayetteville.

Algal growth at Lake Fayetteville is fueled by both N and P, and it is likely that both of these nutrients influence HABs and the production of cyanotoxins like microcystin (Wagner et al., 2020). If sediment SRP release is sustaining these nuisance and potentially HABs, then there are management options to reduce SRP release from bottom sediments in lakes. For example, alum and other Al products have been used to remediate internal P sources from bottom sediments for many decades (Welch and Cooke, 1995; Welch and Schrieve, 1994). The addition of alum to lake waters results in the dissolution and disassociation of the Al salt, forming Al flocs which adsorb SRP from the water and settle to the bottom coating the bottom sediments like an Al floc blanket. Based on our core experiments and the observed decrease in SRP in overlying waters under both aerobic and anaerobic conditions, alum would be a potential man-agement option for Lake Fayetteville. A con-servative 10:1 ratio of Al to P was selected for this study, which translates to an average of 16 g Al m^{-2}, and it successfully reduced SRP concentration and fluxes in Lake Fayetteville cores. Alum doses have ranged between 2:1 and 100:1 ratios of aluminum (Al) to P by mass and 6

Figure 4: Research platform on Lake Fayetteville for frequent measurements of water temperature and dissolved oxygen with depth.

to 122 g Al m^{-2} (Rydin et al. 2000; Dugopolski et al. 2008; Huser 2012; Huser et al. 2016; Kuster et al. 2020).

While alum treatments have reduced water column P concentrations, sediment P fluxes and algal abundances in many reservoirs (Smeltzer 1990; Welch and Cooke 1999; Steinman et al. 2004; Huser et al. 2011), the longevity of alum treatments have been variable across the literature. The average longevity for TP reductions after alum treatments was 11 years across numerous lakes and reservoirs in the U.S., Denmark, Germany, and Sweden, and ranged from 0 to 45 years (Huser et al. 2016). The variability in alum treatment longevity is often attributed to insufficient alum doses, saturation of alum flocs, and/or the magnitude of external sediment and P loading (Welch and Cooke 1999; Lewandowski et al. 2003; James and Bischoff 2019).

While immediate effectiveness of alum treatments are often seen in sediment-core experiments (Steinman et al. 2004; Sen 2005; Pilgrim et al. 2007), the long-term effectiveness is difficult to assess with short-term core incubations. In Green Lake, Washington, sediment P fluxes immediately decreased foll-owing alum treatments to cores, but fluxes began to increase again after 32 days of incubation (Degasperi et al. 2009). Therefore, it may be necessary to lengthen core incubation experiments and/or carefully determine alum doses based on reservoir P concentrations and loadings.

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