A Review on the Use and Monitoring of Alum Treatments to Control Algal Blooms

Environmental Assessment Program

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Publication 25-03-009

July 2025

Abstract

For lakes impacted by excessive algal growth and harmful algal blooms (HABs), reducing external phosphorus inputs may not be enough to reverse the nutrient-rich state. It may be necessary to employ an engineered solution for the in-lake reduction or inactivation of phosphorus in the water column. There are a number of products used for the inactivation or sequestration of phosphorus; however, aluminum sulfate (alum) is most commonly used for lakes with HABs. The goal of this literature review is to provide an overview of phosphorus inactivation through the use of alum as a lake management tool. The main objectives are to (1) review basic limnology and algal growth, (2) describe the use of alum as a restoration tool, (3) provide an overview of the chemistry and possible impacts of using alum, and (4) explore potential parameters that can be useful in monitoring to identify and minimize unintended ecosystem impacts.

The goal of an alum treatment is to rapidly alter the lake ecosystem by limiting primary production; as such, there are intended ecosystem shifts. Deciphering unintended ecosystem shifts is often difficult to quantitatively disentangle from intended shifts . Phosphorus inactivation treatments are permitted under Ecology's Aquatic Plant and Algae Management (APAM) General Permit, which expires on March 21, 2026. As part of the renewal process for the permit, Ecology can consider changes to the monitoring requirements associated with phosphorus inactivation products. The water quality monitoring currently required under the permit is worth continuing, particularly collecting water samples analyzed for aluminum, dissolved organic carbon, and pH. There are a number of supplemental ecosystem indicators that could be monitored before and after alum treatments, depending on the availability of funding and community interests. Under the permit, Ecology should focus on requiring monitoring that documents the concentrations of active chemicals being added as phosphorus inactivation agents.

Publication Information

This report is available on the Department of Ecology's website at:
https://apps.ecology.wa.gov/publications/SummaryPages/2503009.html.

The Activity Tracker Code for this study is: 25-016.

Suggested Citation:

Hobbs, W., and M. Rosewood. 2025. A Review on the Use and Monitoring of Alum Treatments to Control Algal Blooms. Publication 25-03-009. Washington State Department of Ecology, Olympia. https://apps.ecology.wa.gov/publications/SummaryPages/2503009.html.

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Background

Lakes have a natural seasonal succession of phytoplankton or algae communities throughout the year. Phytoplankton are microscopic aquatic plants that form the base of the food web in lakes. However, excessive algal growth in lakes impacts the recreational value of these waterbodies and can have potential human and wildlife impacts if cyanobacteria are producing toxins. Harmful algal blooms (HABs) are often driven by an over-abundance of growth-limiting nutrients, mainly phosphorus (P) (Schindler 1974; Vollenweider 1975; Schindler et al. 2016). The sources of P to lakes vary from lake to lake and mainly depend on the physical setting (surface geology and hydrology) and the surrounding watershed land use (urban, agricultural, or undeveloped).

For lakes impacted by excessive algal growth and HABs, reducing external P inputs may not be enough to reverse the nutrient-rich state (Scheffer and Jeppesen 2007; Søndergaard et al. 2003). It may be necessary to employ an engineered solution for the in-lake reduction or inactivation of P in the water column. Several products are used to inactivate P; however, applying aluminum sulfate (Alum; $Al_2(SO_4)_3$) to lakes with HABs is the most common approach (Table 1). Alum is generally applied as a liquid, often with sodium aluminate ($Na(AlO_2)$), a buffering agent. The goal of any restoration management action attempting to inactivate water column P and sediment P is to decrease the growth and productivity of algae. It is important to remember that these engineered approaches are essentially large-scale ecosystem experiments, and their outcomes may vary significantly.

Table 1. Number of Ecology-permitted phosphorus inactivation treatments.

Year	Aluminum sulfate	Sodium aluminate	Iron filings	Lanthanum-modified clay
2006	2	—	—	—
2007	1	—	—	—
2008	2	—	—	—
2014	1	—	—	—
2015	2	—	—	—
2016	4	2	—	—
2017	3	1	—	—
2018	5	2	—	—
2019	1	1	—	—
2020	3	1	—	3
2021	3	3	1	4
2022	3	1	1	2
2023	2	1	—	4
Total	32	12	2	13

The goal of this literature review is to provide an overview of phosphorus inactivation through the use of aluminum sulfate (alum) as a lake management restoration tool. The main objectives are to (1) review basic limnology and algal growth, (2) describe the use of alum as a restoration tool, (3) provide an overview of the chemistry and possible impacts of using alum, and (4) explore potential parameters that can be useful in monitoring to minimize unintended ecosystem impacts. This review will not cover the details in assessing whether a lake is suitable for treatment or the work of considering available restoration approaches. More detailed overviews of short-term lake management restoration tools and alum treatments can be found in Cooke et al. (2005), Welch and Cooke (1999), and Osgood et al. (2016). In addition, the North American Lake Management Society is a professional association dedicated to lake management and has a position paper on their website[1]; they offer workshops at their annual conference and have also produced guidance manuals on the restoration of lakes for the USEPA (Olem and Flock 1990; Wedepohl et al. 1990).

Limnology and Phosphorus Cycling

Limnology is the study of the physical, chemical, and ecological aspects of inland waters. The origin of a lake can determine its hydrologic balance (Figure 1). Three major types of lakes in the Puget Sound Lowlands and northern Washington are kettle lakes, glacially formed drainage lakes, and reservoirs. Kettle lakes are formed by glacial scour or remnant ice deposits and tend to have small stream inputs or outputs. They rely heavily on groundwater inputs, precipitation, and evaporation to maintain lake levels. Glacially formed drainage lakes have a dominant inlet and outlet that feed and drain back into the watershed. Reservoirs are engineered basins used for water storage. Reservoirs have an upstream basin maintained by stream and/or river inputs and a downstream dam on the output used for water release. Many reservoirs in eastern Washington are used for power generation and irrigation as part of the Columbia River Basin Project.

[1] https://www.nalms.org/nalms-position-papers/the-use-of-alum-for-lake-management/

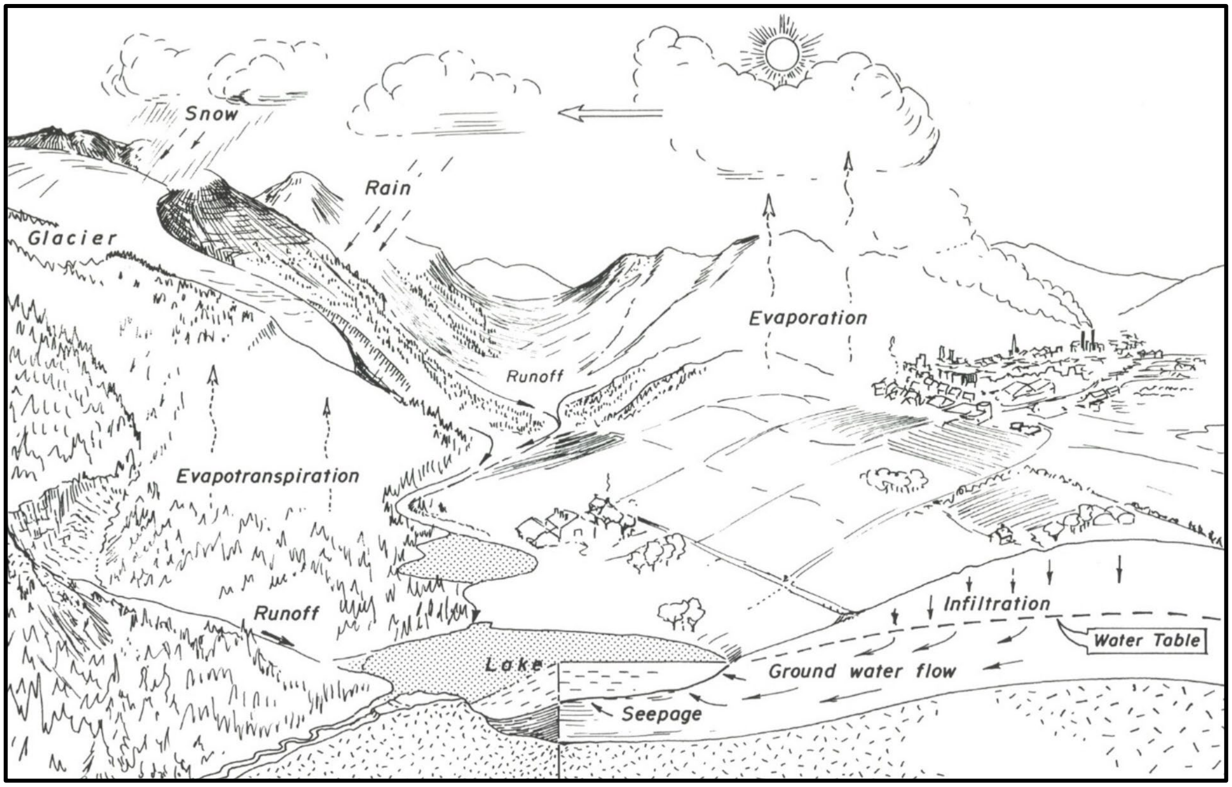

Figure 1. The hydrologic cycle as it affects lakes. Drawn by John Glew (Smol 2009).

External hydrologic and atmospheric inputs to lakes can carry nutrients and pollutants from the upland watershed into the water column (Figure 2). Watershed land use can therefore have a dramatic impact on the health and productivity (rate of algal growth) of a lake. The origin of a lake does not determine the productivity; however, the hydrology of different lake types will affect the inputs and cycling of nutrients.

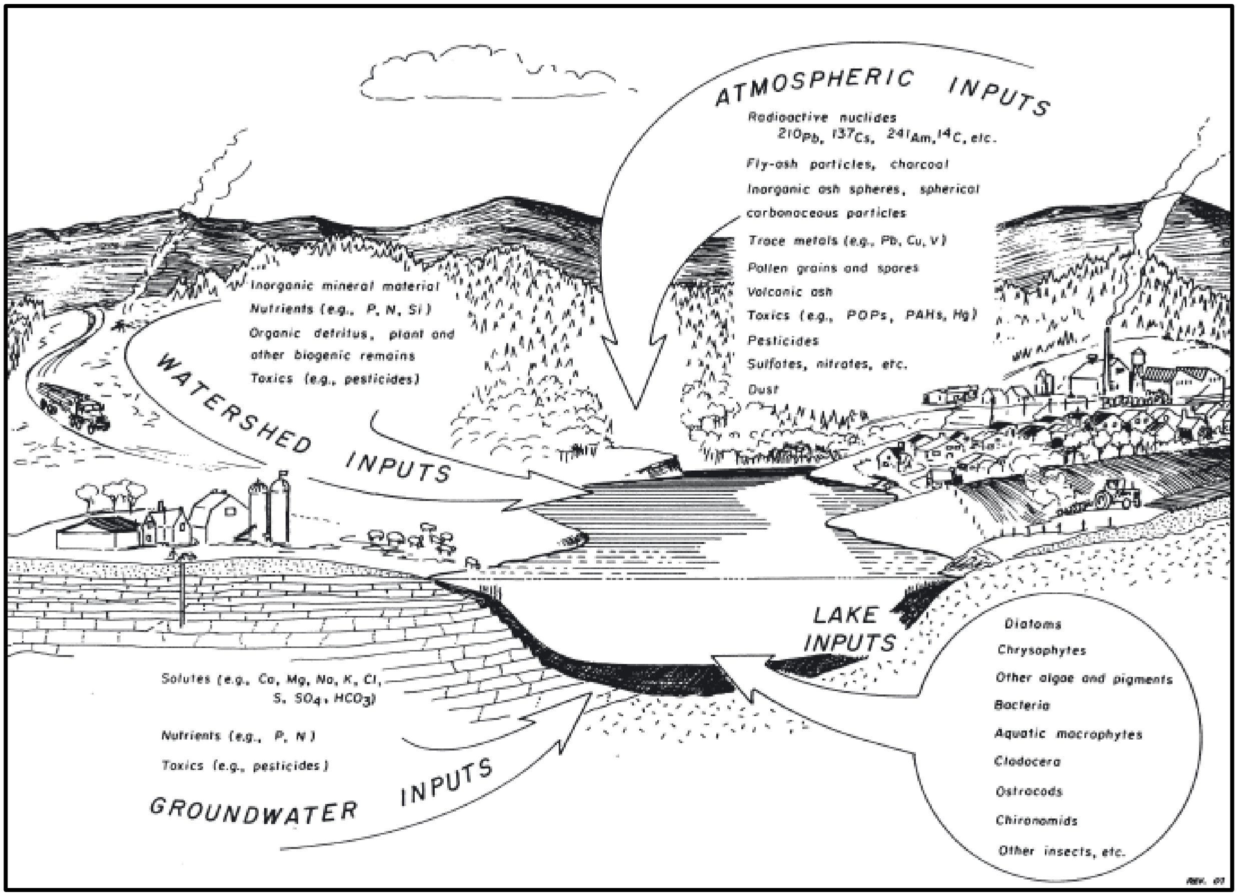

Figure 2. Depiction of inputs to lake ecosystems. Drawn by John Glew (Smol 2009).

The physical mixing of a lake's water column depends on the heating and cooling of the water throughout the year and is an important process for the redistribution of nutrients. Seasonal warming and cooling create a period of water column stratification and periods of mixing as the heat is redistributed from upper to lower waters. Most lakes in Washington are dimictic (mixing twice per year) and turn over in the spring and fall. Shallow lakes can be polymictic, with near-continuous mixing due to wind. The physical structure of a lake's water column can affect a lake's chemical and biological characteristics.

Phytoplankton (algae) growth and productivity are dependent on light and nutrients. Light penetration through clear water can be limited by compounds such as tannins (i.e., stained water), and phytoplankton itself can attenuate light and limit the depth of solar radiation. Nitrogen (N) and P are essential elements for primary productivity; however, when these nutrients are high, they can lead to excessive algae growth. Lake productivity can be either oligotrophic, having low nutrient enrichment and low algal production, mesotrophic with moderate nutrient enrichment, or eutrophic with high nutrient enrichment. Most lakes with excessive algal growth and impairments are eutrophic or hypereutrophic systems.

Phytoplankton succession from the spring through fall in most dimictic lakes generally follows the trend described in Figure 3. In spring, as the daylight hours increase and waters warm, bioavailable nutrients and silica are mixed throughout the water column, promoting early algae blooms dominated by diatoms. During the summer, waters continue to warm, leading to stratification in most lakes; available silica and nutrients decrease as the summer progresses, which promotes a shift in algal communities. In the late summer, cyanobacteria (blue-green algae) can dominate owing to their ability to move throughout the water column and effectively store and scavenge nutrients; cyanobacteria blooms in the late summer or early fall can produce harmful toxins. In the fall or early winter, the water column is mixed, productivity is generally lower, and diatoms become an important part of the phytoplankton community again. In eutrophic lakes, cyanobacteria play a dominant role in the phytoplankton succession throughout the growing season.

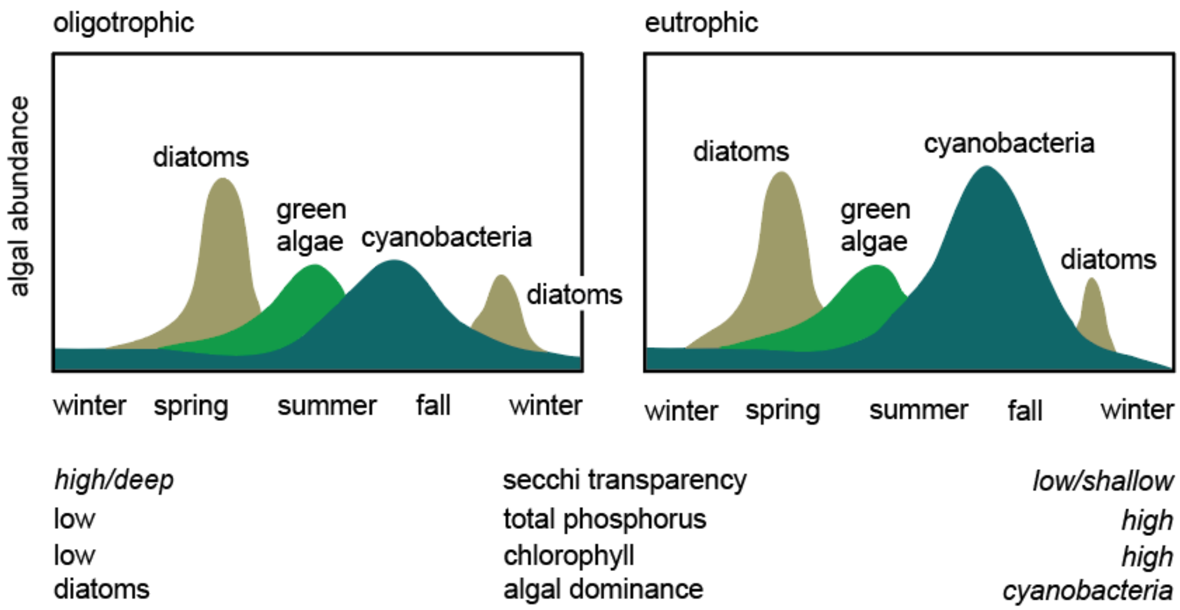

Figure 3. Exaggerated seasonal succession of lake phytoplankton.
Adapted from WOW (2004). The left side is nutrient-poor (oligotrophic), and the right side is nutrient-rich (eutrophic). Common limnological response variables are listed in the lower center below the figure.

Phosphorus is generally the primary limiting nutrient for algal growth in freshwaters (Figure 4; Schindler 1974), so lake management focuses on controlling abundant phosphorus to reduce algal blooms. Sources of P into lakes can come from external inputs, such as runoff, incoming streams, and groundwater. In addition, phosphorus can be cycled internally from the sediments, decaying plants, and animal excretions.

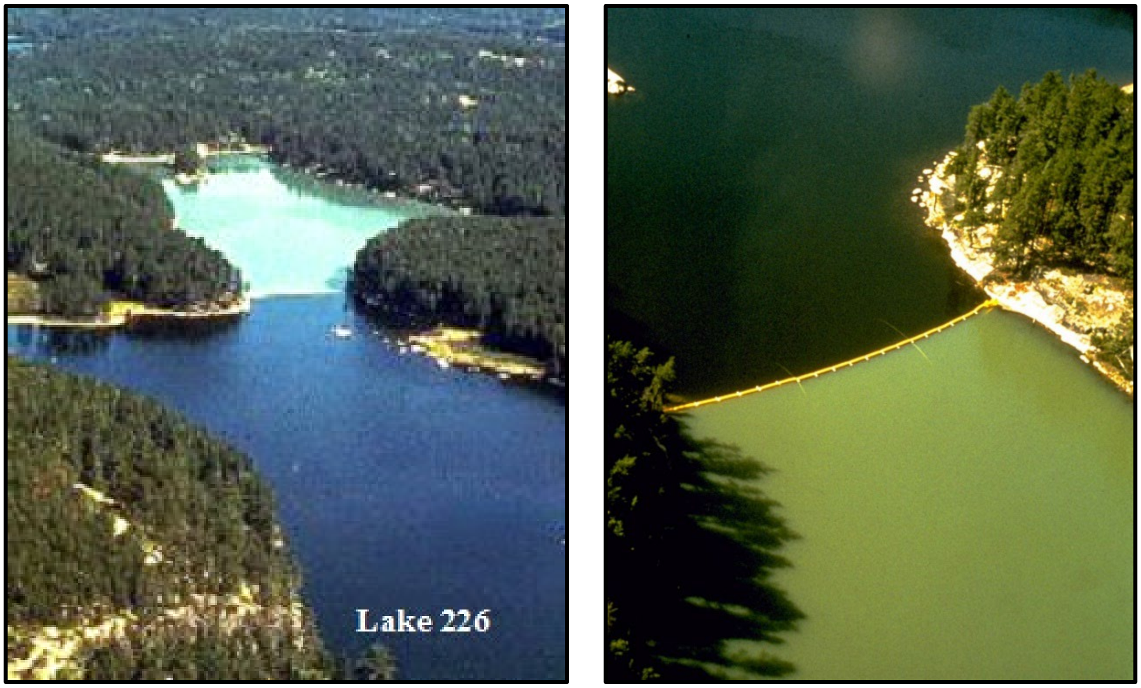

Figure 4. Lake 226 in the Experimental Lakes Area, 1973.
Lake 226 was divided by a curtain and amended with phosphorus (P), nitrogen (N), and carbon (C) additions (green algal growth) and N and C only (clear water). Photographs taken by David Schindler (Schindler 1974).

Phosphorus reductions can be achieved by directly managing nutrients from external sources. However, nutrient-rich conditions can persist as a result of internal sources, largely the recycling of bioavailable P from the sediments (Figure 5). Phosphorus can cycle from soluble to insoluble and organic to inorganic forms, and this cycling can vary from season to season and over different trophic conditions (i.e., oligotrophic vs eutrophic). Inorganic forms of P (orthophosphate, PO_4), which are bioavailable, tend to bind with iron and manganese oxides under oxic (in the presence of dissolved oxygen) conditions. These bonds are redox-sensitive, which means that as the conditions become anoxic (oxygen-devoid) in the bottom waters or sediments, bioavailable P can be liberated. Lake stratification is necessary for this process, and when lakes overturn, or high wind periods create mixing, internal P is available in the water column. The P cycle shown in Figure 5 is fairly simplistic; further detail on the transformations and key solid and aqueous P pools in a lake can be found in Figure A-1.

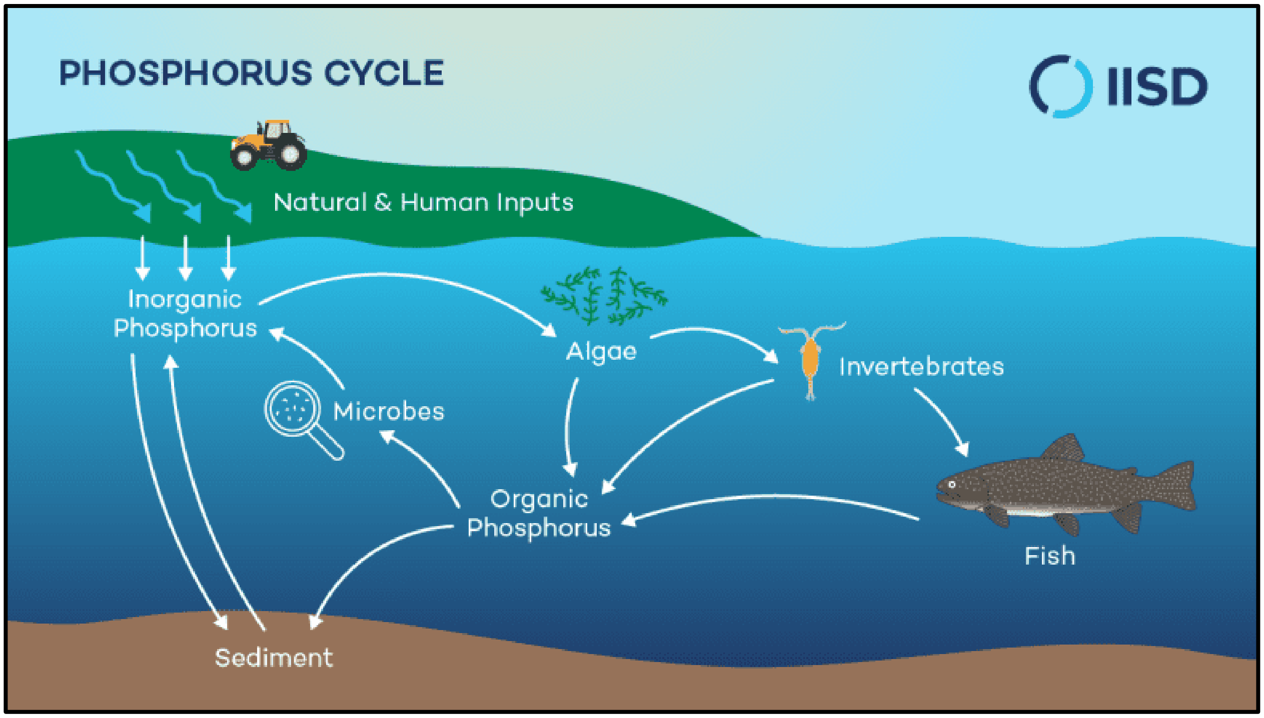

Figure 5. Simplified lake phosphorus cycle.
IISD = International Institute for Sustainable Development (https://www.iisd.org/ela/), which is operating the Experimental Lakes Area

Understanding the cycling and loading of P into and within a lake is an important step in assessing the appropriate restoration tool. Otherwise known as a P budget, modeling and prioritizing the P loads of a lake is necessary to understand the relative inputs of P driving algal growth. A P budget aims to quantify each of the terms in Figure 6. Inputs to the lake include surface flows (streams and runoff), groundwater, atmospheric and internal loading from the sediments under certain conditions. Outflows or losses of P from the lake include surface water flows, groundwater and sedimentation losses. Mathematical models are also used to understand the flux/loading and retention of P in lake ecosystems (Vollenweider 1975; Brett and Benjamin 2008).

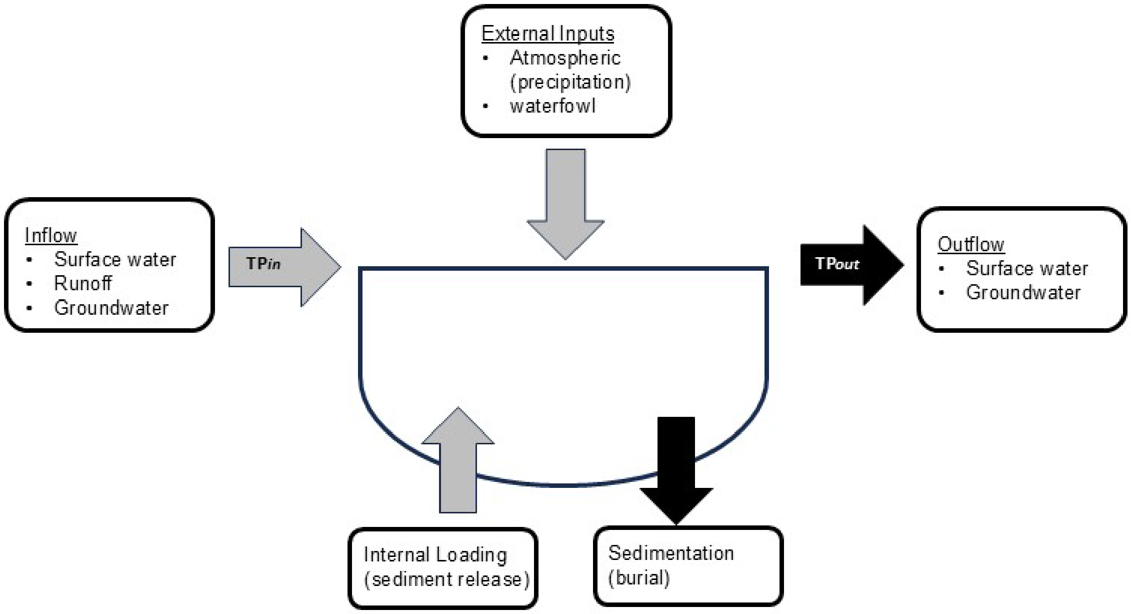

Figure 6. Simplified phosphorus budget for a lake.
Grey arrows are inputs to the lake; black arrows are outputs

Phosphorus inactivation as a restoration tool

Nutrient inactivation is accomplished through the addition of chemical coagulants and flocculants. The chemical additions do not act as an algaecide, and any direct impact on cyanobacteria or algae communities at the time of treatment is due to the physical removal by flocculation. There are multiple chemicals that can be used for P inactivation, including:

- Aluminum-based compounds
  - Aluminum sulfate, $Al_2(SO_4)_3$
  - Sodium aluminate, $Na(AlO_2)$
  - Polyaluminum hydroxychlorides, PACl (numerous compounds)
- Iron-based compounds
  - Ferric sulfate, $Fe_2(SO_4)_3$
  - Ferric chloride, $FeCl_3$
  - Zerovalent iron (ZVI; FeO)
- Calcium (Ca) and lanthanum-based compounds
  - Lanthanum, La (used with bentonite clay)
- Calcium or lime, CaO and $Ca(OH)_2$, sometimes as $CaCO_3$

The availability and quality of chemicals vary and seem to impact the use of inactivation treatments. Chemicals are either supplied and added to the water column as a solid or a liquid. The goal of adding chemicals as an inactivation mechanism is two-fold: (1) to strip or bind available P (mostly as dissolved inorganic P or PO_4, or dissolved organic P) from the water column and (2) to prevent the internal loading or recycling of P from the sediments. Removing P from the water column relies on binding to or in chemical particulates, which are then transported to the lake sediment for burial. If supporting limnological studies show that internal P loading represents the dominant flux of P to the water column, adding an inactivation chemical to the sediment surface can reduce the internal P flux. In the case of inactivating or capping the lake bottom sediment, a higher dose is usually applied.

Calcium additions to lakes as a P inactivation tool rely largely on the formation of calcium carbonates and the incorporation of P into the lattice of precipitates; this reaction is most efficient at a pH >9 (Cooke et al. 1993). P can bind to the outside of calcite particles, but this bond is weak and is not an effective long-term solution (Golterman 1988). In hard-water lakes that experience productivity-driven shifts in pH and authigenic carbonate precipitation events, P can be temporarily removed from the water column. In general, the addition of Ca as an inactivation tool is seldom used in Washington State lakes.

Lanthanum (La) is a relatively new chemical that is being used in inactivation treatments. It is currently being incorporated into proprietary formulations with bentonite clay and used as a flocculent and binding agent. In a study by Lürling and Tolman (2010), the lanthanum-modified clay (PhosLock®) at a high concentration (250 mg/L) was subjected to a leachate experiment and released only small quantities of total La, on the order of 0.13 – 2.13 µg/L. Lanthanum has been shown to bioaccumulate in fish organs following treatments, but no acute or chronic effects were noticed (Waajen et al. 2017). In bioassay experiments with PhosLock ® using the common zooplankter, *Daphnia magna*, Lürling and Tolman (2010) found growth endpoints (EC_{50}) of 871 mg PhosLock/L for weight and 1557 mg PhosLock/L for length. A typical lake treatment application with Phoslock is 2 – 3 tonnes per hectare. Given a lake that is 5 hectares with a mean depth of 10m, this would result in a water concentration of roughly 65 mg/L of Phoslock. Lürling and Tolman (2010) present an average application dose of 84 ± 24 mg/L over four different lake treatments. Typically, it appears that application doses are well below lab-derived toxicity thresholds.

Another brand of La-modified bentonite with a higher La concentration is Eutrosorb, which contains 10% La versus 5% found in Phoslock. In a recent treatment of Spanaway Lake in the Puget Sound Lowlands, Eutrosorb was applied at a concentration of approximately 3.5 mg/L (Aquatechnex 2024). The Spanaway La treatment was orders of magnitude below the toxicity thresholds found by Lürling and Tolman (2010).

Lanthanum-modified clays bind with phosphate to form the mineral rhabdophane ($LaPO_4 \cdot nH_20$), which is highly stable. However, the scavenging efficiency of lanthanum-modified clays appears to be impacted by pH, where adsorption capacity decreases at pH>9 (Ross et al. 2008) and at higher dissolved organic carbon concentrations (Li et al. 2020). Few studies look at the total and available lanthanum in the water column during treatments. Lürling et al. (2024) measured La in dissolved and total form before and after a large treatment to both the sediment (14 tonnes) and the water column (13.65 tonnes) in a Dutch lake and found measurable concentrations following treatments (Figure 7). Based on the amount of lanthanum-modified clay (PhosLock) added and the lake volume, there would be a theoretical concentration of 1050 mg PhosLock/L in the water, which is quite high.

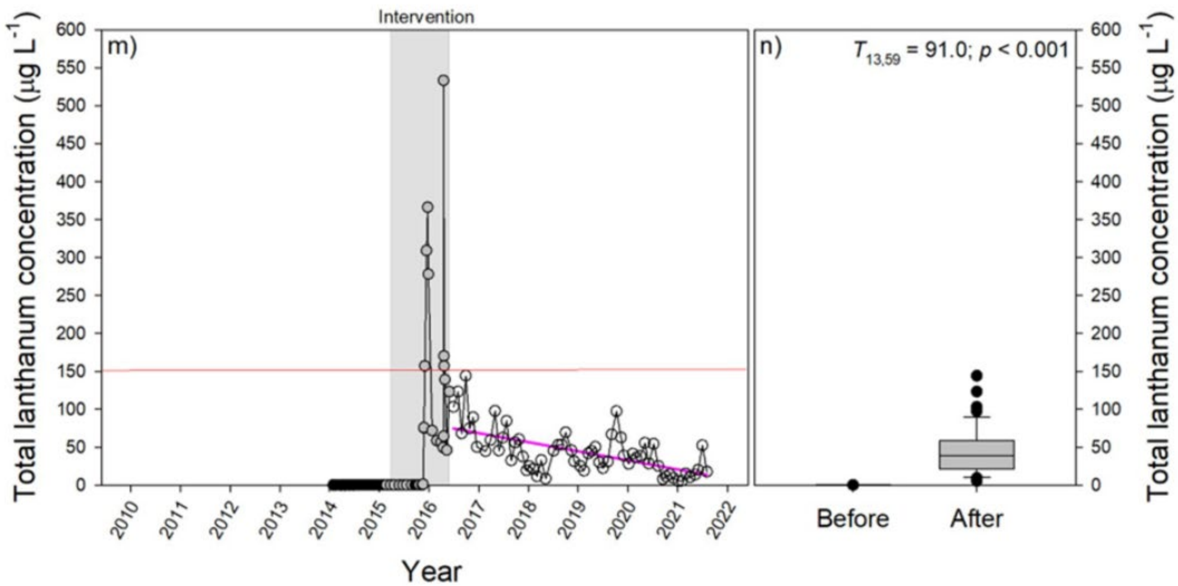

Figure 7. Lanthanum monitoring data from Lürling et al. (2024) before and after the 2016 PhosLock treatment.

The use of iron and iron salts has a long history in wastewater treatment (Jenkins 1971). In lakes, the addition of iron in various forms has been employed as an inactivation tool. Iron (oxy)hydroxides ($Fe(OH)_3$ are very effective at binding PO_4 and forming complexes. Examples and reviews on the use and effectiveness of iron treatments are available from North American (Walker et al. 1989; Orihel et al. 2016; Engstrom 2005; Natajaran et al. 2021) and European lakes (Foy 1985; Boers et al. 1994; Deppe and Benndorf 2002; Kleeberg et al. 2013). Owing to the redox sensitivity of $Fe(OH)_3$, the long-term effectiveness of iron treatments can be impacted in deeper lakes by seasonally anaerobic bottom waters and sediments (Nurnberg 2009). The internal loading of P from anaerobic sediments is largely a function of the iron content of the sediments. This observation also holds true for shallow lakes with aerobic waters (Jensen et al. 1992; Søndergaard et al. 2003). The potential instability of chemical treatments using iron has led many practitioners to incorporate hypolimnetic aeration devices into the restoration plan or instead rely on aluminum for chemical treatment. It should be noted that this paradigm of redox-sensitive Fe species leading to internal P loading has been challenged by some (Hupfer and Lewandowski 2008; Kleeberg et al. 2013).

Aluminum sulfate (alum) and sodium aluminate are the dominant chemicals used in the US for treatment and have a history of ~50 years of use (Cooke et al. 2005). This report will focus on aluminum as an inactivation chemical. Readers interested in the use and application of other chemicals should consider the references earlier in this section and the chapter on "*Phosphorus Inactivation and Sediment Oxidation*" in Cooke et al. (2005) and references therein.

Chemistry of Alum Treatments

Alum is applied to lake water as aluminum sulfate, or $Al_2(SO_4)_3$ 14 H_2O. When aluminum sulfate is added to water, it forms aluminum ions, which are then hydrated (combined with water) (eqn 1). Liquid aluminum sulfate has a pH of 2.2 and an aluminum content of 4.4% Al^{3+} by weight.

$$Al^{+3} + 6\ H_2O \rightleftarrows Al\ (H_2O)_6^{3+} \qquad \text{(eqn 1)}$$

Aluminum and hydrogen ions are liberated during the chemical hydrolysis steps. The aluminum ions lead to the formation of aluminum hydroxide ($Al\ (OH)_3$), and the hydrogen ions cause a decrease in lake water pH (eqn 2):

$$Al^{3+} + H_2O \rightleftarrows \text{intermediate reactions} \rightleftarrows Al\ (OH)_3(s) + H^+ \qquad \text{(eqn 2)}$$

Aluminum hydroxide $Al\ (OH)_3(s)$ is a solid precipitate that forms a flocculent material, referred to as a floc, that has a high capacity to adsorb phosphates (PO4); physical entrapment of P-containing organic matter also occurs within the floc. Phosphorus can also precipitate as $AlPO_4$ (eqn 3).

$$Al_2(SO_4)_3 + 2PO_4 \rightarrow 2AlPO_4 + 3SO_4 \qquad \text{(eqn. 3)}$$

The buffering capacity or acid-neutralizing capacity of the water will dictate the resulting pH after the addition of aluminum sulfate. A lake water pH range of 6 – 8 is optimal to maintain aluminum hydroxide in a solid precipitate form (Figure 8). Above this pH range, aluminate ($Al(OH)_4^-$) predominates, and below pH 6, $Al(OH)_2^+$ will dominate, followed by Al^{3+} or Al (III) below pH 4. High pH and predominance of aluminate will reduce the effectiveness of the alum floc to bind P (Cooke et al. 1993).

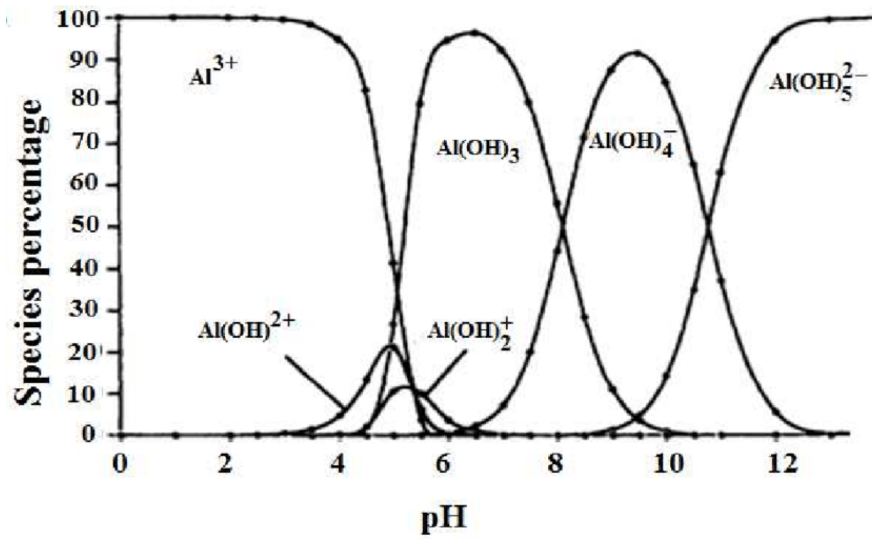

Figure 8. Distribution of aluminum species as a function of pH.

Most waters contain sufficient alkalinity so that large doses will not lower the pH below 6.0 – 6.5. However, if water alkalinity is naturally low, the potential pH reduction may require a buffering agent — sodium aluminate (Cooke et al. 2005). The buffering agent is typically added in combination with aluminum sulfate. A 38% liquid sodium aluminate solution has a pH of 11.5. The use of sodium aluminate is crucial to buffer hydrogen ions and can play a significant role in reducing the toxicity of aluminum (EPA 2020). Continuous monitoring of the pH during application will prevent over- or under-buffering the lake.

Alkalinity is the measure of the buffering capacity of water, and both hardness and alkalinity play important roles in alum applications. Hardness is the measure of dissolved minerals, such as calcium or magnesium, in water. Water hardness determines the alkalinity of a lake and, therefore, the planned dosage for each unique alum lake application. The greater the hardness values, the more ions present that compete and make aluminum less bioavailable.

Another factor that influences aluminum bioavailability is dissolved organic carbon (DOC). Aluminum can form strong complexes with humic and fulvic acids in water (Angel et al. 2015; Campbell et al. 1983), which are what is being measured as DOC. At pH values between 6.5 and 9.0, aluminum complexes readily bind with humic substances. Therefore, in lake waters undergoing alum treatments with higher concentrations of humic substances, there is a tendency for higher doses of aluminum sulfate to bind phosphate ions because of the competition for binding sites.

The aluminum floc that sinks to the lake sediment surface can continue to bind PO_4 from interstitial waters and recycled internal P after treatment. Indeed, some treatments have the main goal of negating the internal P loading by adding a layer of aluminum hydroxide to the surface sediment. It has also been observed that the deposited $Al(OH)_3$ floc will work its way into the upper sediments as a result of the density of the floc compared to the bulk density of the upper sediments (Rydin et al. 2000). Post-depositional sediment records documenting the $Al(PO_4)$ layers have demonstrated the retention and burial of P following treatment (Rydin et al. 2000). If sufficient capacity in the sediment exists, then continued binding of P can take place; this is often described by the Al:P ratio (James and Bischoff 2015). In cases where internal P loading is the largest source, a large P immobilization dose may be applied to the lake by injection in the bottom waters, with the goal of providing an $Al(OH)_3$ sediment cap.

Alum Dose Determination

The dose required for an alum treatment depends on the chemistry of the receiving waters and whether the goal is to prevent internal P loading from the sediments. For a treatment of precipitating P from the water column, practitioners will typically complete a sequential test on water samples from the water body to decipher the appropriate ratio of Al:P. The dose depends on the P concentration, pH, and alkalinity of the water. During the initial range finding or testing of the water, the threshold for alum additions is a 90% reduction in P without reducing the pH to below 6 or reducing the alkalinity to below 95% of the pre-treatment concentration (Kennedy and Cooke 1982; Osgood et al. 2016). This initial range-finding procedure is called "jar test" by practitioners and is required as part of Ecology's General permit. This Al:P ratio will then be scaled up to the lake for the treatment.

If the goal of the treatment is to prevent internal P loading by injection of aluminum sulfate into the bottom waters, then additional methods are required. This dose is dependent on the estimate of internal P loading from the sediments, which can be assessed in several ways: (1) mass balance approach based on characterization of the P in the water column (e.g., Nürnberg 1998), (2) using sediment cores by measuring the loosely-bound or mobile P fractions of the upper sediments and calculating a P release rate (Pilgrim et al. 2007), and (3) incubating sediment cores and measuring P release under oxic and anoxic conditions (Nürnberg 1987). Other methods that are seldom used include benthic flux chambers and in situ porewater profiles. There is a great deal of inconsistency in how internal P loading is defined, measured, and assessed, which introduces uncertainty into these estimates and treatment dose calculations (Orihel et al. 2017).

Rydin and Welch (1999) conducted some sediment benchtop experiments and suggested that a ratio of 100:1 is necessary between the amount of Al added: the amount of mobile P (Fe-bound P and loosely bound P) in the sediments to convert the available P to Al-bound P. Post-treatment measurements of added Al: Al-P in the sediments of several Washington lakes was roughly 11:1 in the sediment layers corresponding to the treatment window (Rydin et al. 2000).

The actual volumetric and aerial doses of alum applied to Washington lakes are summarized in Table 2 (Herrera 2024). There is a great deal of variability in the magnitude of the dose, and generally, the earlier doses in the 1980s are much lower because they tended to be calculated based on the alkalinity of the receiving water and the dose required to shift to pH 6.0 (Figure 9). As dose calculations began to focus on preventing internal loading and estimating the dose based on the Al:P required to immobilize mobile P (Rydin and Welch 2000), alum doses applied to lakes increased (1990s through 2000s).

Table 2. Summary of Alum Treatment Doses in Washington State (Herrera, 2024).

Lake (County)	Treatment Date	Vol. Dose (mg Al/L)	Aerial Dose (g Al/m2)	Longevity (years)a	Reference
Blackmans Lake (Snohomish)	*2025 Proposed*	20.5	86	unknown	Herrera 2024
Heart Lake (Skagit)	April 2018	12.9	32.1	>5	Herrera 2019
Lake Campbell (Skagit)	October 1985	10.9	26	7	Huser et al. 2016
Lake Erie (Skagit)	September 1985	10.9	20	14	Huser et al. 2016
Lake Ketchum (Snohomish)	May 2014 March 2015 Annual 2016–2024	19.5 20.4 3.0–6.1/yr	71.3 74.6 11–22/yr	NA NA NA	M. Burghdoff (pers. comm. 2024)
Lake Stevens (Snohomish)	Annual 2013–2020 2022, 2024	0.15/yr 0.15/yr	3.0/yr 3.0/yr	NA 2	Tetra Tech 2022 S. Farrant (pers. Comm. 2024)
Long Lake (Kitsap)	September 1980 September 1991 August 2006 April 2007 April 2019	5.5 5.5 2.5 17.5 5.0	10.7 10.7 4.6 36.2 9.8	>11 >11 NA 10 unknown	Rydin et al. 2000 Rydin et al. 2000 Tetra Tech 2010 Tetra Tech 2010 Tetra Tech 2019
Lake Ballinger (King)	June 1990	5.0	23	2	Huser et al. 2016
Green Lake (King)	October 1991 April 2004 April 2016	8.6 24 8.2	34 94 32	3 >10 >8	Herrera 2003 Herrera 2004 Herrera 2016
Phantom Lake (King)	September 1990	4.4	28	12	Huser et al. 2016
Hicklin Lake (King)	April 2005	22	60.4	3	King County 2006
Lake Fenwick (King)	May 2011 October 2023	2.5 11.7	9 42.1	stripping unknown	S. Brattebo (pers. Comm. 2023) Tetra Tech 2024
Wapato Lake (Pierce)	July 1984 July 2008 April 2017	7.8 67.7 56.3	12 108 90	<1 5 >6	Huser et al. 2016 Herrera 2017a Gawel and Oliva-Membreno 2018
Waughop Lake (Pierce)	March 2020 July 2020/2023	40 40/20	84 84/42	unknown	Tetra Tech 2023
Long Lake (Thurston)	N: Sept. 1983 S.: Sept. 1983 2008	7.8 7.4 15.2	28 28 54.9	12 5 unknown	Huser et al. 2016 Huser et al. 2016 Tetra Tech 2006

Lake (County)	Treatment Date	Vol. Dose (mg Al/L)	Aerial Dose (g Al/m2)	Longevity (years)a	Reference
Pattison Lake (Thurston)	S/N: Sept. 1983	7.8/7.2	30.8/31	12/<1	Huser et al. 2016
Black Lake (Thurston)	April 2016 May 2021	1.9 54.5	13 317	>5 unknown	Herrera 2017b Herrera 2021
Liberty Lake (Spokane)	1974 1980–1981	1 10	5 52	0.5 14	Huser et al. 2016 Huser et al. 2016
Medical Lake (Spokane)	Aug.–Sept. 1977	12.6	122	>10	Huser et al. 2016
Newman Lake (Spokane)	1989 Annual 2021–2023	2.8 1.5/yr	15 12.7/yr	1 NA	Huser et al. 2016 D. Vilar (pers. Comm. 2023)

[a] Longevity reported by reference or observed through 2023

mg Al/L = milligrams of aluminum per liter; g Al/m^2 = grams of aluminum per square meter; NA = not applicable

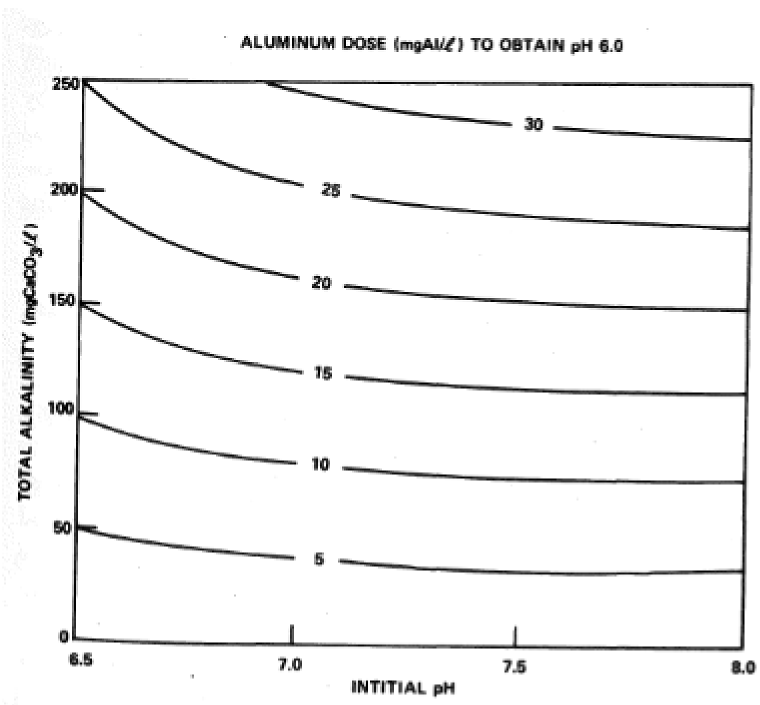

Figure 9. Estimated alum dose to shift pH to 6.0 as a function of water alkalinity (from Cooke et al. 2005).

Effectiveness of Alum Treatments

The effectiveness of alum treatments has been assessed by long-term monitoring of water column reductions in PO_4 and total P, in addition to post-treatment sediment coring to assess the binding and burial of P. There are several examples discussed in Cooke et al. 2005 covering lakes that stratify and those that do not. Welch and Cooke (1999) give an overview of 21 lakes that have received treatments, both water column and hypolimnetic treatments to reduce internal loading; aluminum sulfate doses varied from 2.6 to 12.0 g Al/m^3 for water column treatments and 4.5 to 30.0 g Al/m^3 for hypolimnetic sediment treatments. A treatment is generally considered successful if the post-treatment TP and chlorophyll a levels in the water column and internal sediment P loading rate remain below the pre-treatment levels. Thirteen out of 21 of the treatments in the Welch and Cooke (1999) study were considered successful, lasting for 4 – 21 years. Three shallow polymictic lakes in the study had macrophyte beds that interfered with the alum treatments, while several dimictic lakes had external P loading, land use changes in the watershed, and low alum doses that led to short periods of treatment effectiveness.

The long-term effectiveness of a hypolimnetic alum treatment to control internal P loading can be impacted by the higher-density floc sinking into lighter surface sediments, bioturbation of the floc, and the deposition of new sediment with higher P concentrations (Rydin et al. 2000; Lewandowski et al. 2003; Cooke et al. 2005). Generally, in lakes where successful treatments have occurred, the layer of Al-bound P can be measured and documented in sediment cores collected several years following treatment (Figure 10).

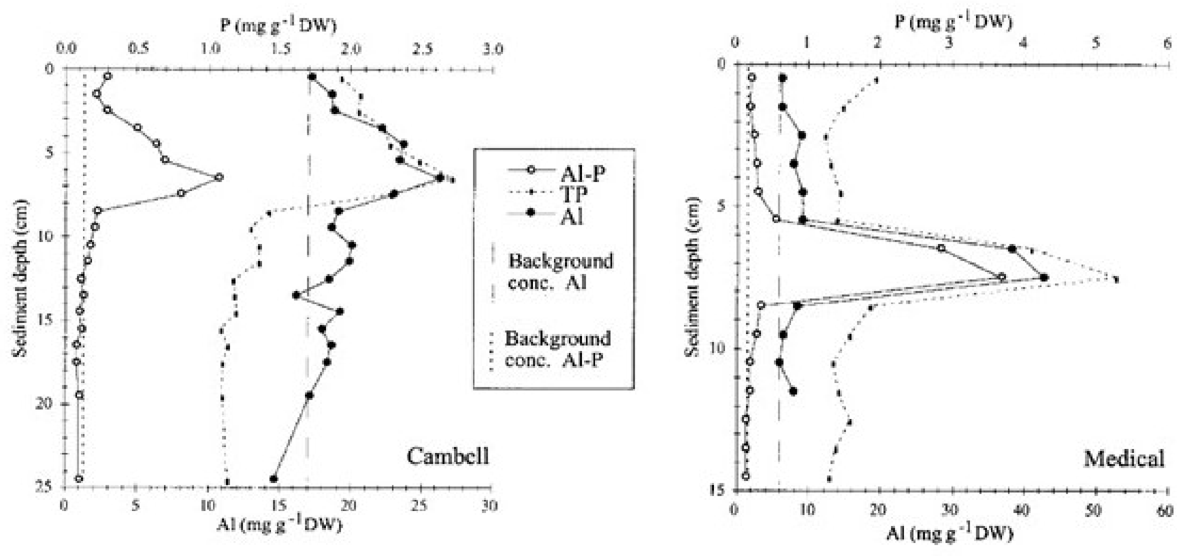

Figure 10. Sediment core profiles of Al-bound P in two lakes from Rydin et al 2000.

Sediment-P can be bound in different fractions ranging from strong recalcitrant bonds (e.g., minerals such as vivianite) to redox-sensitive P (e.g., Fe-bound P) to loosely-bound P (e.g., porewater P or loosely bound to organics); both redox-sensitive and loosely-bound P are susceptible to remobilization (Psenner et al. 1988). Sediment-P content is usually higher near the sediment-water interface and decreases with depth (Engstrom and Wright 1984). This observation is a function of loosely-bound P and redox-sensitive P being mobilized from Fe and Mn oxides in the upper sediments and new sediment being deposited. The goal of an Alum treatment is to bind and retain P in a more recalcitrant form (Al oxides and organic P) to encourage longer-term burial of P (Lukkari et al. 2007). Lewandowski et al. (2003) found a molar ratio for Al:P of 2.1 in the sediment of Lake Susser See about 10 years after treatment with Al salts, and the P-adsorption capacity of the Al-containing sediment layer was not exhausted in this lake.

In Washington State, Ketchum Lake in Snohomish County stands out as an informative case study in alum treatment effectiveness and adaptive management. Lake Ketchum is a relatively shallow, formerly hypereutrophic dimictic lake. The hydrology is dominated by groundwater inputs and a small inlet stream. Early monitoring data showed that approximately 73% of the annual P load was contributed by internal loading, with 23% contributed from the inlet stream (Brattebo et al 2017). An algae control plan for the lake was published in 2012 that described a collaborative approach with residents within the watershed and the county (Burghdoff and Williams 2012). An initial in-lake alum (and sodium aluminate) treatment was applied mostly in 2014 and 2015, with smaller maintenance treatments targeting the inlet stream applied from 2016 onwards. The main metric of success for this treatment has been the reduction in water column P (~95% reduction in the surface waters) and algal biomass (~85% reduction); water clarity has increased, and qualitative observations of macrophyte communities have suggested an increased diversity.

In a study of 83 lakes dosed with alum, Huser et al. (2016) found that the main variables influencing the long-term reduction in water column TP were alum dose, lake water residence time (estimated as the flushing ratio, watershed area: lake area; WA:LA) and the lake morphology (described as the Osgood index; Oi = mean depth/lake area$^{0.5}$) (Figure 11). The decision tree modeling in Figure 11 describes the thresholds for the Al dose (15.1 g Al/m^2), WA:LA (27) and the Osgood ratio (5.7). Subsequent thresholds within the decision tree were deemed not as significant (Huser et al. 2016). Overall treatment longevity averaged 11 years for all lakes (15 years in stratified lakes and 5 years in polymictic lakes) (Huser et al. 2016); effectiveness was based on 50% declines in epilimnetic total P (TP) concentration over the 2 years following treatment.

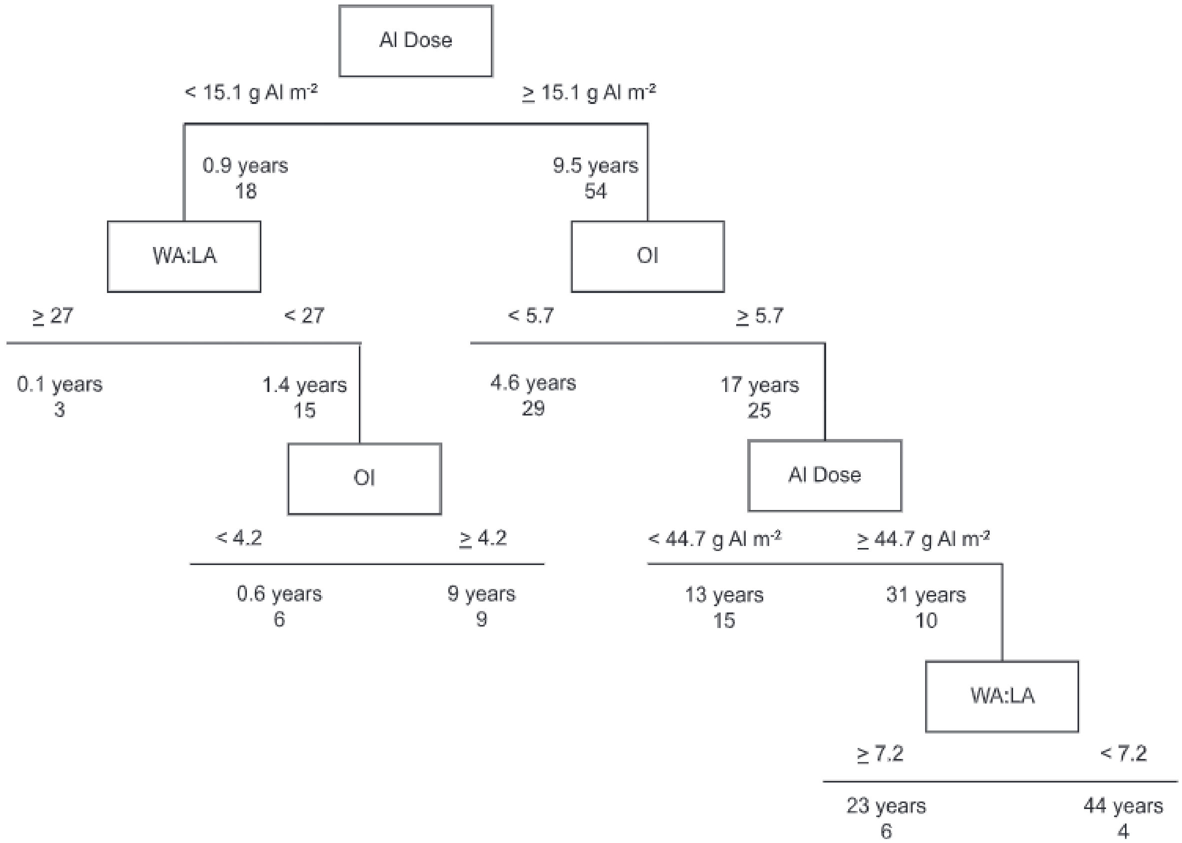

Figure 11. Decision tree partition model from Huser et al. (2016) on alum treatment effectiveness in years.
OI = Osgood index or lake morphology; WA:LA = watershed area:lake area.

The above-mentioned study by Huser et al. (2016) defined a metric of "longevity" or effectiveness by calculating 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 before 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. This approach provides a simple threshold, but very little statistical rigor in defining measurable reductions from the pre-treatment monitoring and nutrient concentrations.

Possible negative impacts

Direct organism impacts (toxicity)

The main concern with the addition of aluminum sulfate to the water column is the potential reduction in lake water pH (< 6) and dissolution effects resulting in available Al (III) ions. The toxicity of Al (III) is well studied, and elevated levels of aluminum can affect some aquatic species' ability to regulate ions, like salts, and inhibit respiratory functions (EPA 2018). Aluminum can also accumulate on the surface of fish gills, leading to respiratory dysfunction and possibly death. Aquatic invertebrates with gills are susceptible to Al through endpoints similar to fish; sediment-dwelling invertebrates (e.g., mollusks) can be less sensitive. Aquatic plants are generally less sensitive to aluminum than fish and other aquatic life.

In early alum treatments, Kennedy and Cooke (1982) and Cooke et al. (1978) suggested that a concentration of 50 µg Al/L be adopted as a safe upper limit for post-treatment dissolved aluminum concentrations. Dose was therefore defined as the maximum amount of aluminum which, when added, would still ensure low (<50 µg AI/L) aluminum concentrations. The EPA first recommended total recoverable Al criteria of 87 µg AI/L (chronic exposure) and 750 µg Al/L (acute exposure) in 1988. The 1988 criteria were not adopted in Washington State. More recently, the toxicity of aluminum to aquatic life is understood to depend not only on pH, but also dissolved organic carbon (DOC) and hardness (EPA 2018). The protection of aquatic life criteria is now site-specific, based on multiple linear regression (MLR) models that incorporate the relevant parameters.

The Washington State criteria for the protection of aquatic life in ambient waters were recently updated to adopt the latest EPA criteria; the proposed rulemaking is under review by EPA and not final. The MLR approach[2] was recommended for site-specific total recoverable aluminum criteria. Default criteria were also calculated based on representative data from Ecology's Environmental Information Management (EIM) database, stratified into east and west of the Cascade Mountains divide: West = 270 µg Al/L (chronic exposure) and 510 µg Al/L (acute exposure) and East = 480 µgAl/L (chronic exposure) and 820 µg Al/L (acute exposure).

Direct impacts on benthic macroinvertebrate larvae have been observed at concentrations similar to those of the revised state criteria. Senze et al. (2024) found an acute toxicity LC_{50} (where 50% of the test individuals die) concentration for Chaoborus at 646 µg Al/L and for chironomids at 208 µg Al/L. The toxicity of porewater aluminum to benthic larvae has not been explicitly tested and is likely to vary somewhat based on the importance of DOC and pH to the availability of dissolved Al.

In addition to DOC and hardness or alkalinity of the receiving water, temperature is also important and affects the rate at which aluminum toxicity can harm aquatic organisms. At lower temperatures, the solubility of aluminum becomes lower, which lowers the toxicity of

[2] https://www.epa.gov/wqc/aquatic-life-criteria-aluminum

total aluminum. At decreased water temperatures, the chemical/biochemical reaction rates and the metabolic rates also become slow, which means lower rates of consumption and utilization by aquatic organisms and therefore lower toxicity of total aluminum. For every 10 degrees C of temperature increase, the reaction rate doubles (EPA 2020).

Alum treatments resulting in an acute toxicity event to fish appear rare and are likely the result of an incorrect calculation of dose or a lack of adaptive management and monitoring during the treatment to maintain buffering capacity. Smeltzer et al. (1999) felt that the decline in yellow perch health and density following an alum treatment in Lake Morey, VT, was likely attributable to short-term chronic exposure to dissolved aluminum in the water column. The recovery of the yellow perch community took approximately 3 years (Smeltzer et al. 1999).

In Washington State, Heart Lake in Skagit County was treated in 2018 with alum and sodium aluminate at a volumetric dose of 12.9 mg Al/L (Herrera 2019). The initial pH of the lake was 8.8, decreasing to 8.2 during the treatment; dissolved oxygen was high throughout the pre- and post-treatment period. Unfortunately, the treatment of the lake resulted in total aluminum concentrations in the water column of 0.99 – 1.7 mg/L after the first day of treatment and 2.0 – 5.16 mg/L after the second day of treatment, followed by concentrations of 1.17 – 2.65 mg/L two days after treatment. Subsequently, a fish mortality event occurred with 32 trout dying in the 5-day period after treatment, presumably from chronic exposure to aluminum. A qualitative amphibian survey conducted before and after did not find any impact on egg clusters.

Indirect ecosystem impacts

The benthic macroinvertebrate communities of lakes have been studied in response to the application of hypolimnetic alum treatments that blanket the lake bottom. Generally, there have been a number of observations of macroinvertebrate richness and densities decreasing following treatments, followed by recovery within ~2 years and possible increases thereafter (Smeltzer et al. 1999). A whole-lake alum treatment was applied to eutrophic Spring Lake, Michigan, during October and November 2005. An ecological assessment of the lake was performed eight months following treatment (2006) and again in 2010 and 2016; data were compared with pre-treatment assessments in 2003 and 2004. Total macroinvertebrate density declined significantly in 2006 compared with 2004, but then increased significantly in 2010 and 2016 (Steinman et al. 2018).

In addition to macroinvertebrate densities, community structure can change in response to changes in water clarity and productivity. A common response following alum treatments has been the greater presence of chaoborids (*phantom midges*; predatory pelagic invertebrates) as water clarity increases and prey such as rotifers increase in abundance (Schumaker et al. 1993; Doke et al. 1995; Smeltzer et al. 1999; Steinman et al. 2018). It should be acknowledged that benthic macroinvertebrate communities in profundal (deep locations) areas of lakes are generally low in diversity and often dominated by species or genus that can tolerate pollution or low oxygen environments (e.g., midge larvae). The use of benthic macroinvertebrate communities in profundal sediments as a biotic index is therefore relevant to changes in oxygen deficiencies, whereas littoral communities can be more indicative of impacts from acidification and habitat changes (Poikane et al. 2016). Lastly, it is worth mentioning that continuous flow alum treatments on lake inflow streams can impact (toxic effects and smothering) stream and lake littoral communities depending on the injection concentration (Barbiero et al. 1988; Pilgrim and Brezonik 2005).

Zooplankton density has been observed to decline immediately following treatments, possibly in response to the physical action of a settling aluminum floc, toxicity of aluminum, or predation (Schumaker et al. 1993). Declines are often temporary (on the order of months), and rotifers seem to become more dominant following an alum treatment. Daphnia have also been observed to decline following treatments, likely in response to predation with increased water clarity (Doke et al. 1995). Opposite trends in biomass and Daphnia densities have also been observed immediately following treatment (Lund et al. 2010).

Very little work has been completed on the impacts on macrophytes following alum treatments and floc deposition. There is some work on the uptake of dissolved Al by aquatic plants, particularly in the context of phyto-remediation of effluent or constructed wetlands (e.g., Goulet et al. 2005). The phyto-remediation work does not demonstrate an impact on the plants, but rather the effectiveness of uptake. It may also be the case that following an alum treatment, the density of macrophytes increases in response to improved water clarity as phytoplankton biomass decreases (e.g., Welch and Kelly 1990). This has also been observed in Lake Ketchum, Snohomish County, in response to the annual treatments with alum (pers. comm. M. Burghdoff, 2025).

Some recent work in Minnesota on the sensitivity of the native wild rice (*Zizania palustris*) to levels of sulfide in the sediments is worth acknowledging (Myrbo et al. 2017). Sulfide forms in the sediments through the anaerobic process of microbial sulfate reduction, and sulfide can have toxic effects on rooted plant growth (Lamers et al. 2013; Simkin et al. 2013). However, to date, the addition of sulfate as part of an alum treatment (as $Al_2(SO_4)_3$) and the production of sulfide in the sediment have not been explicitly tested.

Grand Lake St. Mary's in Ohio received an alum treatment in 2010, and several studies were conducted before and after the treatment. Nogaro et al. (2013) investigated the indirect effects of the treatment on sulfur cycling within the lake, including the upper sediment layers. They measured an increase in sulfate (SO_4^{2-}) in the porewaters and surface waters at several locations in the lake, and while sulfides in the sediments were not measured, they speculated that an increase in sulfide production could occur, impacting benthic invertebrates. In addition, the authors measured an increase in dissolved Al in the sediment porewaters.

Impacts on fish communities can be a concern for lake managers conducting alum treatments. The direct impacts on fish respiration from exposure to Al ions were noted at the beginning of this section. Indirect impacts on fish are possible through the alteration of the lake food web and productivity, potentially reducing food availability. A study in Lake Nordberg (Germany) concluded that after an alum treatment, fish responded rapidly to changes in nutrient state, both in terms of community structure and habitat use; improved oxygen concentrations in the deeper profundal zone were likely responsible for the observed habitat shift (Lund et.al 2010).

Monitoring of Alum Projects

Lake water quality ecosystem monitoring before, during, and after alum treatments varies with the lake management goals and the applied science interests of the communities near the lake, as well as potentially academic interests.

Pre-treatment monitoring (diagnostic)

The decision to treat a lake with alum is based on data showing ample available phosphorus in the water column that is directly related to algae and cyanobacteria growth, and most often, the internal P loading is a relevant contribution to the annual lake P budget. Therefore, there is a general understanding of the chemical limnology before treatment. Under Ecology's Freshwater Algae Control Program,[3] regional entities have been partially funded to work towards a Lake Cyanobacteria Management Plan, which entails a characterization of the lake's annual P budget. Historically, in Washington State, there were a number of large lake management plans carried out in the late 1970s and 1980s under the EPA's Clean Lakes Program under section 314 of the Clean Water Act, with additional funding from the State of Washington. Occasionally in more recent years, large lake management plans have been appropriated state funding (e.g., City of Lakewood 2017).

The amount of diagnostic monitoring before an alum treatment or other lake management restoration efforts varies with the complexity of the impacts to the lake and the funding budget available (Table 3). The frequency of monitoring the lake should be sufficient to capture the seasonal variability and spatial variability across the lake basin, and at least over one year, preferably two or more. Samples should be collected at least monthly during the monitoring period. In some cases, the phytoplankton community has been characterized quantitatively or qualitatively, with suitable repetition to provide an indicator of measurable change following the treatment. However, it is surprising how seldom the phytoplankton community is assessed before and after treatments in both Ecology-funded projects and elsewhere.

[3] https://ecology.wa.gov/about-us/payments-contracts-grants/grants-loans/find-a-grant-or-loan/freshwater-algae-program-grants

Table 3. Monitoring parameters commonly measured during the diagnostic phase of a lake study (pre-treatment).

Parameter	lake inlet	epilimnion	hypolimnion	lake outlet	lake sediment
Total phosphorus (TP)	X	X	X	X	—
Ortho-phosphate (PO_4)	X	X	X	X	—
Total persulfate nitrogen (TPN)	X	X	X	X	—
Nitrate-nitrite (NO_3-NO_2-N)	X	X	X	X	—
Ammonia (NH_3-N)	X	X	X	X	—
Dissolved and total aluminum and iron	—	X	X	—	—
Cations (calcium, potassium, magnesium, and sodium)	—	X	X	—	—
Anions (bromide, chloride, fluoride, and sulfate)	—	X	X	—	—
Phytoplankton (possibly cyanotoxins)	—	X	X	—	—
Zooplankton	—	X	X	—	—
Phosphorus fractions (loosely bound, Fe-P, Al-P, NaOH-P, Ca-P, and inorganic-P)	—	—	—	—	X
Total recoverable aluminum and iron	—	—	—	—	X
Total organic carbon	—	—	—	—	X
Secchi disk (in situ)	—	X	—	—	—
Temperature (in situ)	X	X	X	X	—
Specific conductance (in situ)	X	X	X	X	—
Dissolved oxygen (in situ)	X	X	X	X	—
pH (in situ)	X	X	X	X	—

Note. Summarized as a general list from projects funded by Ecology and from the larger historical lake management plans.

Additional media that should be monitored as part of a lake nutrient budget include: groundwater and stormwater inputs; benthic fluxes (or sediment core incubations) can also be used as an alternative means to determine the internal nutrient loading and metals inputs (Pilgrim et al. 2007; Nürnberg 2009). Ecosystem indicators that can be monitored before a treatment include: macrophyte density and community structure, benthic macroinvertebrate density and communities (profundal and littoral), waterfowl habits and populations, and fish communities.

In situ monitoring during treatment (adaptive)

Water quality monitoring that takes place during the alum treatment is primarily intended to prevent toxicity events and adaptively manage the treatment. The main parameter of interest is pH, which should be continuously measured before and during the treatment to prevent the alum from lowering the pH below 6.0. Aluminum toxicity also depends on DOC and hardness of the water; however, pH is the only parameter that can be measured in situ, in real time. In addition, field probes with dissolved oxygen and pH can be used to profile the water column periodically.

To date, permittees have also successfully collected continuous in situ pH data during the treatments (Figures 12 and 13). This parameter seems informative and not onerous to collect. It does require a deployed water quality sonde, and the data are downloaded following the treatment. Additionally, the collection of the pH data before the treatment allows for an understanding of the diurnal fluctuations in pH in the lake water driven by in-lake algal production.

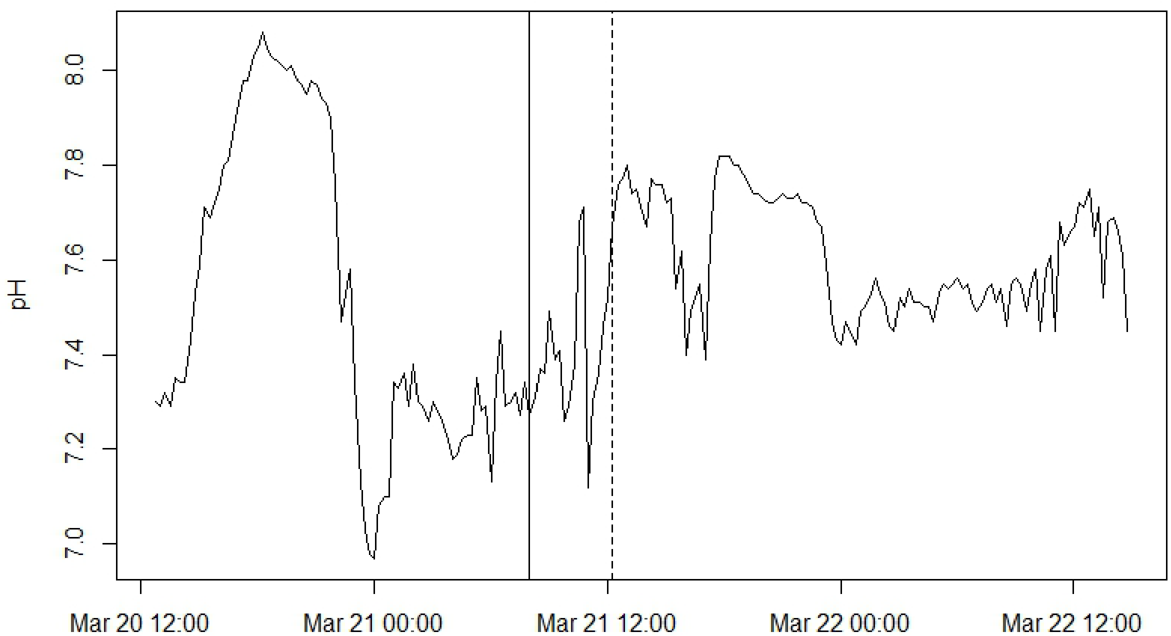

Figure 12. Continuous in situ pH data collected at Ketchum Lake (2023) before, during, and after treatment.
Vertical solid line is the start of the alum treatment; vertical dashed line is the end of the treatment.

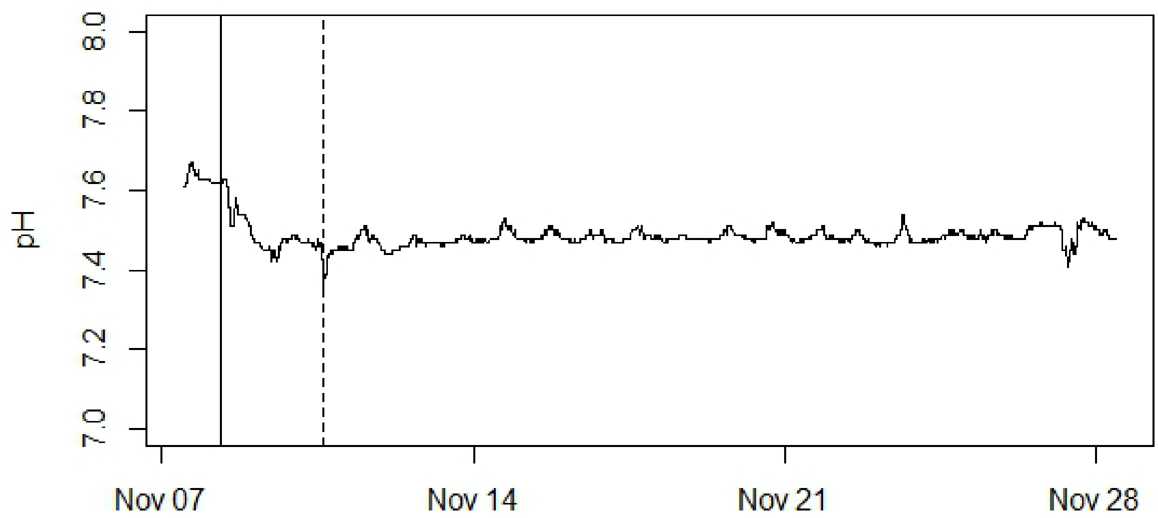

Figure 13. Continuous in situ pH data collected at Stevens Lake (2022) before, during, and after treatment.
Vertical solid line is the start of the alum treatment; vertical dashed line is the end of the treatment.

Post-treatment monitoring (effectiveness)

Several studies have been highlighted in this report that cover long-term monitoring post-treatment to document the effectiveness (Smeltzer et al. 1999; Huser et al. 2016; Lewandowski et al. 2003; Cooke et al. 2005). With any long-term monitoring effort, the key and most difficult aspect is often the consistent funding to complete the study. Many of the parameters described in the pre-treatment section should be continued through to the post-treatment monitoring period, the most important of these being phosphorus (total and dissolved as orthophosphate). In order to detect measurable reductions in phosphorus or algal biomass from the baseline or pre-treatment sampling, sufficient repetition is necessary to achieve a suitable level of statistical power. In addition, the dose calculations for the alum treatment are often based on a calculated reduction in water column total P concentrations that can be verified post-treatment.

Water quality conditions in hypereutrophic Lake Ketchum, Snohomish County, Washington, improved to mesotrophic conditions after alum treatments in 2014 and 2015. From 2013 pre-treatment conditions, summer mean epilimnion total phosphorus (TP) declined from 289 to 34 µg/L in 2014 and then to 15 µg/L in 2015 (a total reduction of 95%). Hypolimnetic TP declined 99% overall, and chlorophyll *a* dropped 70% while there was 135% improvement in transparency over the 2 years. (Brattebo et.al 2017). As highlighted by the Lake Ketchum example, the reduction in P is often dramatic following treatment, and it is common to rely on a mean of water column concentrations over the stratified growing season to describe the long-term trend in P.

Post-treatment monitoring of aluminum in the water column is not often carried out because it is assumed that the aluminum sulfate, added in liquid or hydrated form, is quickly precipitated to aluminum hydroxide that flocs and sediments to the lake bottom (Cooke et al. 2005). However, residual dissolved and particulate aluminum can persist in the water column for months (Figure 14). Unfortunately, the monitoring of dissolved and total Al requires analytical instruments in the lab, and the results are therefore post-hoc verification; in addition to the supplemental parameters DOC and water hardness to infer Al water quality criteria.

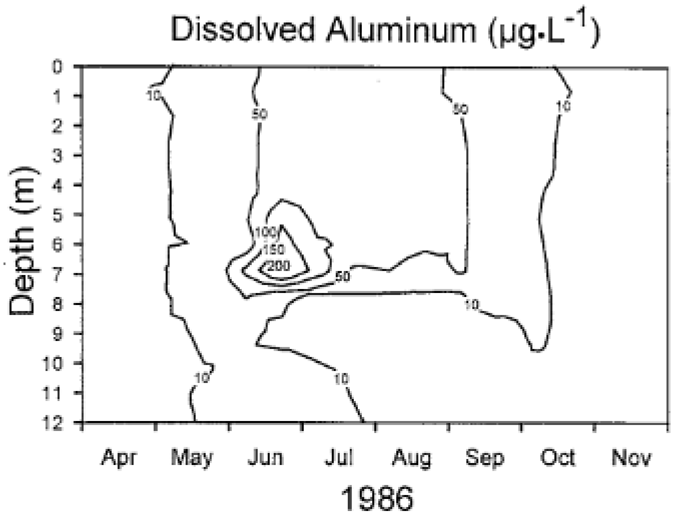

Figure 14. Profiles of aluminum in Morey Lake, VT, following treatment in June 1986 (Smeltzer et al. 1999).

Ecosystem indicators used in monitoring of alum treatments tend to be semi-quantitative, and there is seldom enough repetition, seasonally or spatially, to detect statistical change in community structures. The previously cited study by Smeltzer et al. (1999) presented benthic macroinvertebrate data from 1986 to 1993 and 1997, sampled each year during the winter at two depth contours using six replicates of a sediment dredge. The Smeltzer et al. (1999) dataset highlights the necessary commitment to measuring ecosystem indicators; however, the resulting datasets for ecosystem indicators are often highly variable, making it difficult to detect trends (Figure 15).

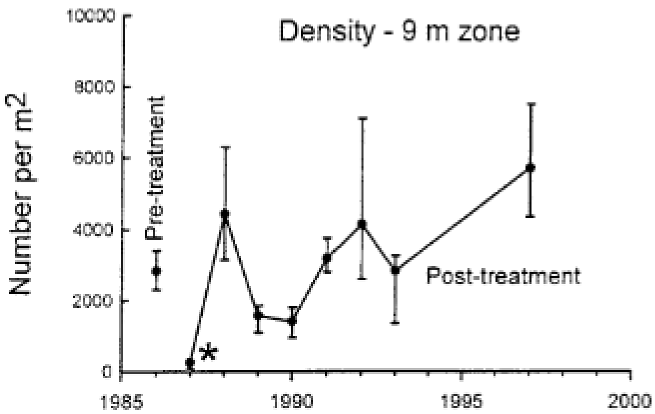

Figure 15. Benthic macroinvertebrate data from Morey Lake pre- and post-treatment (Smeltzer et al. 1999).
The asterisk next to the 1987 sample point denotes a significantly lower abundance.

Permit Requirements in Other States

To our knowledge, the only other states that require some form of permit for alum treatments are Minnesota and New Jersey. The New Jersey Department of Environmental Protection's (NJPDES) Permit No. NJ0356531 is the master general permit for HAB — Harmful Algal Bloom (HAB) Management (GP). Under Appendix B of the permit are the monitoring requirements (Table 4).

Table 4. Permit requirements under New Jersey's HAB — Harmful Algal Bloom (HAB) Management General Permit.

Parameters measured	Pre-treatment	During treatment	Post-treatment (within 1 week)	Within 1–4 weeks after application	Within 2–11 months after application
In situ: pH, dissolved oxygen, conductivity, temperature, Secchi depth	X	pH only (daily)	X	X	X
Total alkalinity	X	—	X	X	X
Total hardness	X	—	X	X	X
Dissolved organic carbon	X	—	X	X	X
Total and dissolved aluminum	X	—	X	X	X
Total phosphorus	X	—	X	X	X
Zooplankton	X	—	X	X	X
Phytoplankton	X	—	X	X	X

Multiple sample locations are also required under the NJ permit, depending on the size of the lake (Table 5). In addition, permittees are required to report the anticipated dose to be applied, all doses tested (e.g., gathered from dosing tests like a jar test), and the water quality results of the required parameters (Table 4). The current permit is effective until 6/30/2029.

Table 5. Permit requirements for number of sample sites under New Jersey's HAB — Harmful Algal Bloom (HAB) Management General Permit.

Waterbody Size (acres)	Number of Sample Locations
≤ 20	3
> 20 and ≤ 50	4
> 50 and ≤ 200	5
> 200 and ≤ 1,000	6
> 1,000	7

The State of Minnesota does not have any monitoring or reporting requirements, but has guidance on maintaining a pH within the range 6.0 – 9.0 during treatment and requires a letter of approval from the Minnesota Pollution Control Agency (MPCA 2020).

Current permit monitoring

The Washington State Department of Ecology has the Aquatic Plant and Algae Management (APAM) General Permit[4] (Ecology 2021) which is a national pollutant discharge elimination system and state waste discharge general permit. The permit is issued under the provisions of Chapter 90.48 Revised Code of Washington (State of Washington Water Pollution Control Act) and Title 33 United States Code, Section 1251 et seq., the Federal Water Pollution Control Act (The Clean Water Act). Specific restrictions are contained in the permit for the application of products for sequestration of phosphorus (Table 6; Table 3 in the permit); among these are the following restrictions for the application of alum (as aluminum sulfate and sodium aluminate):

[4] https://fortress.wa.gov/ecy/ezshare/wq/permits/APAMGeneralPermitFinal.pdf

Table 6. Permit requirements listed in Table 3 of the Aquatic Plant and Algae Management General Permit.

Subject to Timing Restrictions	Restrictions/ Advisories	Treatment Limitations	Other Specific Restrictions
No for fish – check timing window map for other priority species. Timing should address aquatic plant biomass that may interfere with inactivation of sediment phosphorus (requiring early spring or fall treatment).	None	Application must cease when wind speed is greater than 15 miles per hour Powdered alum must be mixed with water to form a slurry before applying to the water surface. The pH of lake water during treatment must remain between 6.0 and 8.5 based on lake average. Only aluminum compounds suitable for water treatment may be used. Buffering materials must be available for use.	A jar test must be completed before whole lake treatments only if a buffer other than sodium aluminate is used or a ratio of liquid alum to liquid sodium aluminate differs from 2:1 by volume. An on-site storage facility is required for any treatment requiring 9,000 gallons of alum or more, or the project proponent must have a plan to store any unused alum or buffering products. Follow the monitoring requirements in S6.B.

The “jar test” in Table 6 refers to the range-finding procedure that is used by practitioners to determine the Al dose for the desired P reduction. While there is no standard method for the jar test, the procedure outlined in Kennedy and Cooke (1982) is generally followed. There are no reporting requirements for the jar test under the current permit. It is also likely that total alkalinity (mg/L) of the water column is being assessed before the alum treatment as part of the jar test, but is likely not being continued following treatment.

Under the permit restrictions are monitoring requirements, detailed in Sec 6B of the permit that pertain specifically to the application of alum (Table 7). The permit states that “water samples must be representative of the treatment area, with at least one shoreline sample and one open water sample” collected.

Table 7. Monitoring requirements under the Aquatic Plant and Algae Management General Permit for an alum treatment.

Frequency	pH (in situ)	Hardness (as CaCO3)	Dissolved organic carbon (mg/L)	Total aluminum (µg/L)
Pre-treatment	X	—	—	—
During	Continuous (minimum 15 min intervals for 24 hrs)	—	—	—
Post-treatment (immediately following)	X	X	X	X
2 weeks	X	X	X	X
1 month	X	X	X	X
2 months	X	X	X	X
6 months	X	X	X	X
9 months	X	X	X	X
12 months	X	X	X	X

Recognizing the reality of applying aluminum sulfate to a lake in large quantities and then measuring total aluminum in the water, permittees are allowed a short-term and long-term exceedance of the water quality standards as long as the permittee complies with the provisions of WAC 173-201A-410. The current version of the permit with the above monitoring requirements has been effective since April 2021.

To date, there have not been prolonged exceedances of the aluminum criteria, as calculated using the proposed site-specific MLR approach. As described in Figure 16, there have been exceedances of chronic exposure criteria immediately following the treatment as permitted under the APAM General permit. In all cases, total aluminum concentrations have decreased by the follow-up sampling event two weeks after the treatment, and in some cases, the decrease has been monitored 24 hours after treatment. Despite the analysis of total aluminum being a post-hoc monitoring tool (i.e., results are not available in real time), the data collected under the permit is useful should there be a need to demonstrate or investigate potential toxicity events. It is also useful to demonstrate the dissipation or sedimentation of aluminum from the water column.

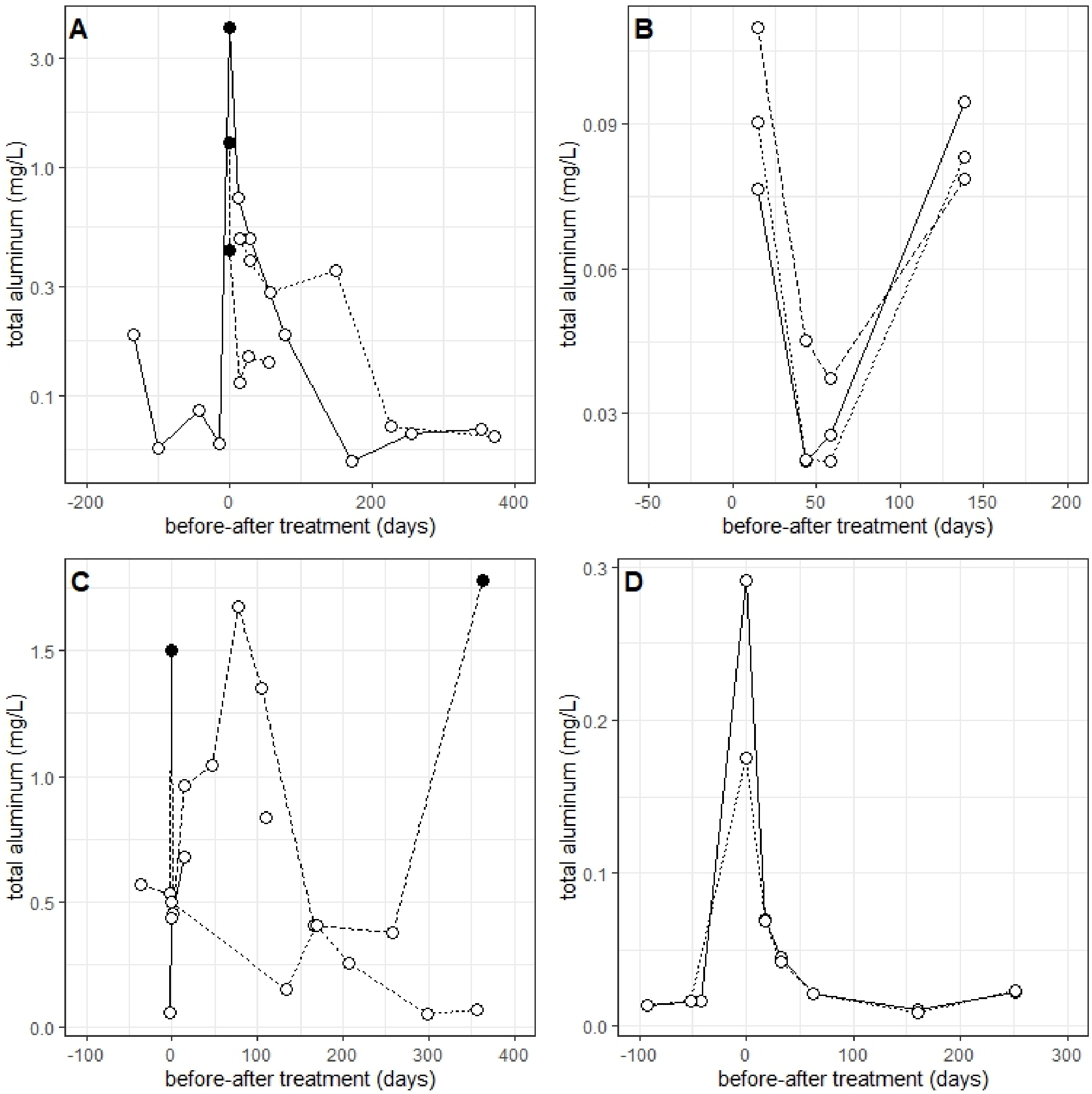

Figure 16. Total aluminum measured pre- and post-treatment in Ketchum Lake (A), Newman Lake (B), Waughop Lake (C), and Stevens Lake (D).
Black dots are total aluminum concentrations above the Washington State chronic criteria; white dots are concentrations below the criteria. Criteria are calculated based on the DOC, pH, and hardness measurements for each sampling event, and therefore criteria will differ. Line type differs to help differentiate between treatments.

The permit monitoring also requires the analysis of dissolved aluminum immediately following the treatment, in addition to total aluminum concentrations. It appears that there is a much higher proportion of dissolved aluminum at higher pH in the lakes treated under the permit since 2021 (Figure 17). As the formation and precipitation of aluminum hydroxide proceeds during the treatment, with the associated lowering of the water pH, a much lower proportion of the total aluminum is dissolved.

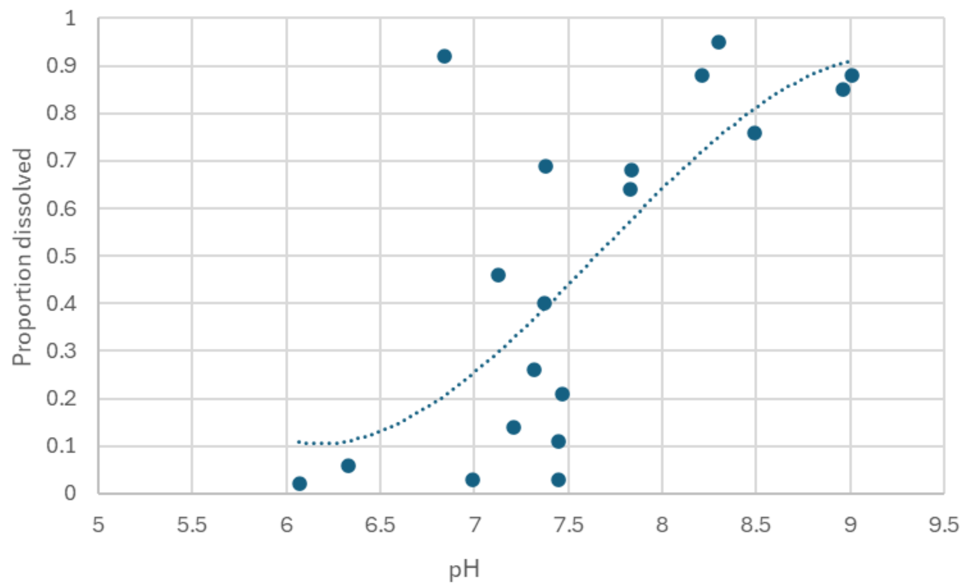

Figure 17. Proportion of dissolved aluminum over a range of lake water pH.
Polynomial regression fit (3rd order) accounts for 50% of the variability in the data.

Additional Monitoring Considerations

Waughop Lake in Pierce County, Washington, underwent three high-dose alum treatments: two in 2020 (March and July) and one in June 2023 (Tetra Tech 2025). In 2023, the monitoring associated with the alum treatments on Waughop Lake suggested that the lake alkalinity and sulfate concentrations were increasing well above the 2020 pre-treatment concentrations. In response, Ecology and the City of Lakewood agreed to expand the suite of monitoring parameters under Sec 6B of the permit to temporarily include a broader suite of anions and cations (as per Table A-2), in addition to sulfate and sulfides (S_2) — the reduced form. Waughop Lake water level is heavily influenced by groundwater inputs, and therefore, the question was whether the changes in alkalinity are being driven by external inputs or the result of increased sulfate from the 2020 treatment.

The monitoring data from Waughop Lake showed that a temporary decrease in alkalinity occurred following the alum treatments, and the previously observed increases in alkalinity (2021 – 2022) were likely in response to groundwater inputs. Sulfate concentrations remained high after the 2023 treatment, becoming similar to pre-treatment levels after a year. Sulfide concentrations measured in the surface and bottom waters were elevated pre-treatment and then close to non-detect 2 – 3 months after the treatment (Table 8). The low concentrations of sulfides may be related to the aerobic conditions throughout the water column before and after the treatment. The additional monitoring parameters collected by the City of Lakewood did not differ between the pre- and post-treatment collections, with the exception of sulfate and bicarbonate values (Table A-2).

Table 8. Total sulfides (mg/L) in 2023/24 grab samples from Waughop Lake.

Collection Date	1.8m depth (bottom)	1m depth	Time period
6/27/2023	2.8	1.6	Day Before
6/30/2023	1.2	4	Day After
7/13/2023	2.4	0.8	2 weeks Post
8/15/2023	0.128 (<MDL)	0.6	Two Months after
9/14/2023	0.0139 J	0.0138 (<MDL)	Three Months after
10/11/2023	—	0.0138 (<MDL)	Four Months after
12/12/2023	0.0383 J	0.0336 J	Five Months after
3/13/2024	0.107	0.232 J	Eight Months after
6/27/2024	0.0117	0.0508	One year after

J = result is an estimate; <MDL = less than the method detection limit.

The long residence of residual sulfate in the Waughop Lake water column, attributable to the treatment, demonstrates a need to understand changes to the sulfur cycle and possible ecological impacts (Lamers et al. 2013; Nogaro et al. 2013). The State of Washington does not have surface water criteria for sulfides or sulfates, and there is not a lot of research on ambient thresholds or porewater thresholds for aquatic life. Relying on ecosystem indicators (e.g., benthic macroinvertebrate communities and aquatic plant surveys) may provide a useful metric of potential changes associated with alterations in lake productivity, sulfur cycling, and porewater aluminum concentrations.

Permit Monitoring Revisions

The data gathered under the current permit for total and dissolved aluminum has documented the potential temporary impact of aluminum toxicity and subsequent loss of aluminum from the water column following treatment. Despite the sample results not being known until analyzed by a qualified lab, the data are important in documenting the lack of or potential cause of a toxicity event. This monitoring requirement should remain in the permit, but the frequency could be shortened, and additional samples could be taken before the treatment (Table 9).

The buffering capacity of the lake water column is pivotal to the assessment of the amount of alum and buffer necessary for the treatment. In all likelihood, this parameter is being measured before the treatment and during the early lake assessment phase of the project. However, it is likely not being measured following the treatment to assess whether alkalinity has returned to pre-treatment levels. Total alkalinity (measured in mg/L) would be a justifiable addition to the alum monitoring requirements (Table 9).

Large amounts of sulfate added to the lake during treatment could alter the sediment S cycle, with the possibility of influencing the production of sulfides (S_2). There are no state water quality criteria for sulfides and no guidance on sediment porewater sulfides. Monitoring sulfate concentrations in the water column could provide an indication of whether the S cycle is being significantly impacted by an alum treatment. Sulfate should return to pre-treatment levels within the monitoring period; if it doesn’t, there may be cause to investigate further.

Table 9. Recommended changes to the current monitoring requirements under the Aquatic Plant and Algae Management General Permit for an alum treatment.

Frequency	pH (in situ)	Hardness (as CaCO3)	Total alkalinity (mg/L)	Dissolved organic carbon (mg/L)	Total aluminum (µg/L)	Sulfate (mg/L)
Pre-treatment	X	**Add**	**Add**	**Add**	**Add**	**Add**
During	Continuous (minimum 15 min intervals for 24 hrs)	—	—	—	—	—
Post-treatment (immediately following)	X	X	**Add**	X	X	**Add**
2 weeks	X	X	**Add**	X	X	**Add**
1 month	X	X	**Add**	X	X	**Add**
2 months	X	X	**Add**	X	X	**Add**
6 months	X	X	**Add**	X	X	**Add**
9 months	X	X	**Add**	X	X	**Add**
~~12 months~~	~~X~~	~~X~~		~~X~~	~~X~~	

Note. Red text indicates changes to current monitoring requirements.

The recommendations in Table 9 are specific to alum treatments. There are total recoverable metals that are part of the active chemicals being added as other P inactivation agents, such as La for lanthanum-modified clay treatments and Fe for zero-valent iron. No other states are requiring monitoring of these elements; however, New Jersey requires monitoring of all other parameters (as listed in Table 4) under treatments with all other P inactivation treatments.

Currently, under Washington's APAM general permit, permittees are required to report the chemical applied, amount (lbs), acres treated, date of treatment, plants targeted, and the results of the monitoring for alum treatments. It would also be useful for permittees to report the calculated volumetric dose (i.e., the results of the jar test, in mg/L) and the actual aerial dose applied (g/m^2).

Conclusions and Recommendations

Alum has been used in lake restoration projects for roughly 40 – 50 years. Ecology has evaluated Alum under an Environmental Impact Assessment[5] (Ecology 2017), and the effectiveness and negative impacts of Alum have been evaluated in the scientific literature (references within this report). However, in recent years, Ecology has received feedback and public concerns about using alum as an inactivation product.

The goal of an alum treatment is to rapidly alter the lake ecosystem by limiting primary production; as such, there are intended ecosystem shifts. Unintended ecosystem shifts are often difficult to decipher and quantitatively disentangle from intended shifts. Alum treatments are regulated under Ecology's Aquatic Plant and Algae Management General Permit (APAM), which expires on March 21, 2026. As part of the renewal process for the permit, Ecology can consider changes to the monitoring requirements associated with phosphorus inactivation products.

The water quality monitoring currently required under the permit is worth continuing. The current suite of chemical parameters seems adequate to provide the necessary data to confirm non-toxic events using the state draft site-specific MLR criteria. However, under the permit, Ecology could consider the following changes to the monitoring requirements:

1. Require that all parameters be measured during the pre-treatment sampling.
2. Reduce the post-treatment monitoring to 9 months if the concentrations are similar to the pre-treatment levels.
3. Add total alkalinity (mg/L) and sulfate (mg/L) to the required parameters for alum treatments.
4. Require monitoring of the total metal concentrations for other P inactivation products (e.g., La for lanthanum-modified clay treatments and Fe for zero-valent iron).
5. Require the reporting of estimated alum (or other P inactivation chemical) volumetric doses (i.e., jar test results as mg Al/L) and the actual dose by area (g Al/m^2) applied.

There have been numerous studies in Washington State that examined the efficacy and long-term effectiveness of alum treatments. While some studies have investigated ecosystem indicators (e.g., benthic and macrophyte communities), long-term monitoring is not routine. As described in this report, monitoring of alum treatments can be binned into pre-treatment, during treatment, and post-treatment, or effectiveness. It is unusual for there to be long-term funding and a commitment to monitoring the efficacy and potential ecosystem impacts. Ecology should evaluate potential funding opportunities under existing programs (e.g., Freshwater Algae Control Program and Water Quality Combined Funding Program) that could be used to supplement and support monitoring and oversight of alum treatments. In addition, there may be utility in Ecology conducting a study to further evaluate chemical and biological

[5] https://apps.ecology.wa.gov/publications/SummaryPages/1710020.html

monitoring parameters to provide more holistic oversight of these restoration actions. Included in Appendix B is a draft study design.

As the current permit undergoes review and potential revision, Ecology should consider convening a workshop with applicators, consultants, and county natural resource departments to discuss phosphorus inactivation treatment monitoring and effectiveness. A review of the current monitoring parameters should be discussed for utility, logistical aspects, and possible changes to improve the protection and restoration of Washington's lake ecosystems.

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Appendix A.

Table A-1. Summary of Ecology-permitted aluminum sulfate treatments 2006 – 2023 under the Aquatic Plant and Algae General Permit.

Year	Permit Number	County	Waterbody	Amount	Acres Treated
2006	WAG994121	Kitsap	Long	15000 gal	340
2006	WAG994121	Kitsap	Long	15000 gal	340
2007	WAG994121	Kitsap	Long	126912 gal	300
2008	WAG994170	Pierce	Wapato Lake	24978.4 gal	34
2008	WAG994131	Thurston	Lawrence (Edited by ECY)	67947 gal	137
2014	WAG994226	Snohomish	Echo Lake (Edited by ECY)	72797 lbs	26
2015	WAG994226	Snohomish	Echo Lake (Edited by ECY)	70200 lbs	26
2015	WAG994235	King	Barbee Mills Retention Ponds	200 lbs	1
2016	WAG994232	Snohomish	Scriber Lake	5346 lbs	2.3
2016	WAG994226	Snohomish	Echo Lake (Edited by ECY)	15660 lbs	26
2016	WAG994246	King	Green Lake	910130 lbs	427
2016	WAG994245	Thurston	Black Lake	609314 lbs	506
2017	WAG994170	Pierce	Wapato Lake	176840 lbs	18
2017	WAG994226	Snohomish	Lake Ketchum	21870 lbs	26
2017	WAG994197	Snohomish	Lake Stevens	271952 lbs	1040
2018	WAG994197	Snohomish	Lake Stevens	43946 lbs	1040
2018	WAG994180	Grant	Moses Lake	196480 lbs	100.3
2018	WAG994380	King	Lake Jeane	270 lbs	8
2018	WAG994226	Snohomish	Lake Ketchum	33000 lbs	26.1
2018	WAG994233	Skagit	Heart Lake	183560 lbs	63
2019	WAG994226	Snohomish	Lake Ketchum	6695 lbs	26
2020	WAG994380	King	Lake Jeane	1000 lbs	10
2020	WAG994380	King	Lake Jeane	999.24 lbs	9
2020	WAG994226	Snohomish	Lake Ketchum	6695 lbs	26
2021	WAG994226	Snohomish	Lake Ketchum	35420 lbs	26

Year	Permit Number	County	Waterbody	Amount	Acres Treated
2021	WAG994245	Thurston	Black Lake	1.17387e+006 lbs	538
2021	WAG994123	Spokane	Newman	76941.5 lbs	141
2022	WAG994123	Spokane	Newman	365970 lbs	141.31
2022	WAG994197	Snohomish	Lake Stevens	37096.8 lbs	653
2022	WAG994226	Snohomish	Lake Ketchum	1797 lbs	26.1
2023	WAG994123	Spokane	Newman	133274 lbs	141.31
2023	WAG994226	Snohomish	Lake Ketchum	28220 lbs	26.1

Fig. 2. A conceptual diagram of the key solid and aqueous P pools that occur in lake sediments and the water column and the processes that cause changes between these pools. Physical processes affecting solid particles are shown as red arrows, physical processes affecting aqueous species are shown as blue arrows, and chemical or biological processes are shown with black arrows. Physical processes: 1, advection or diffusion; 2, external transport; 3, sedimentation; 4, resuspension; 5, burial. Chemical and biological processes: 6, adsorption; 7, desorption; 8, precipitaton; 9, dissolution; 10, recrystallization; 11, assimilation; 12, exudates; 13, hydrolysis or mineralisation; 14, complexation; 15, dissociation; 16, death; 17, migration; 18, intracellular processes.

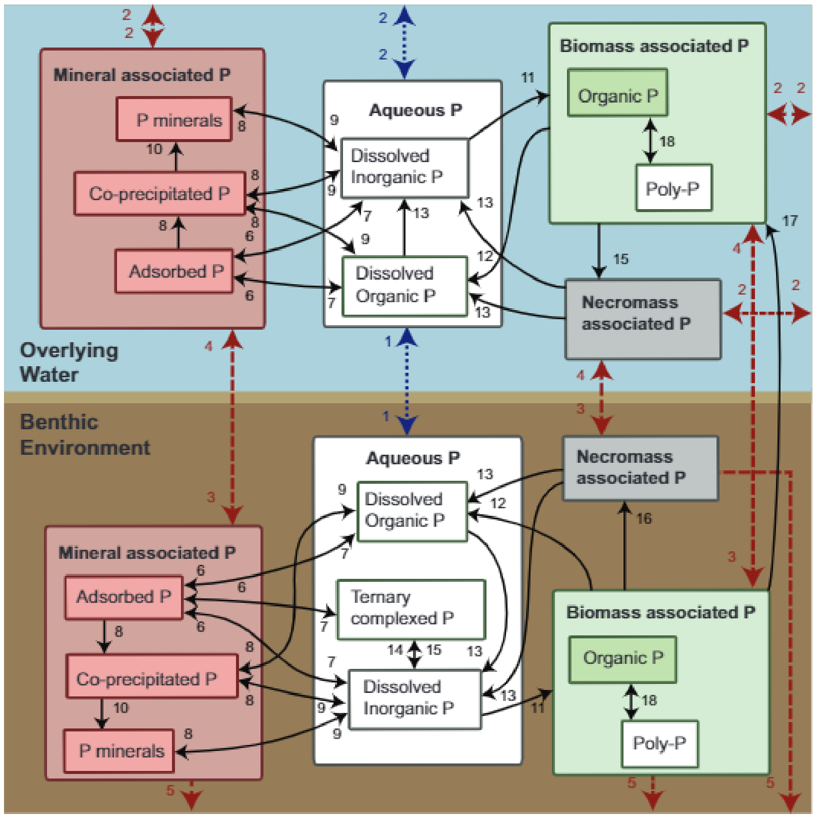

Figure A-1. Detailed phosphorus cycle near the lake sediment-water interface. Directly from Orihel et al. 2016

The detailed P cycle describes the potential complex series of fluxes among mineral-associated P, aqueous P, and biomass-associated P, both in the water column and in sediments. Physical processes, like advection, diffusion, sedimentation, resuspension, and burial, move P among the different compartments. Chemical and biological processes are responsible for a large number of the P fluxes through diffusion, adsorption/desorption, dissolution, assimilation, mineralization, complexation, migration, and intracellular processes.

Table A-2. Summary of the additional monitoring parameters collected at Waughop Lake before and after the 2023 alum treatment.

Sample date	depth (m)	Alkalinity (mg/L)	Chloride (mg/L)	Calcium (mg/L)	Magnesium (mg/L)	Potassium (mg/L)	Sodium (mg/L)	Sulfate (mg/L)	HCO_3 (mg $CaCO_3$/L)	CO_3 (mg $CaCO_3$/L)
6/27/2023	1	53.4	4.62	5.99	1.12	3.37	28.5	25.7	51	<1.00
6/27/2023	1.8	52.7	4.94	5.81	0.913	2.92	28	25.2	51.1	<1.00
6/30/2023	1	38	4.73	5.84	0.804	2.73	48.8	89.5	32.3	<1.00
6/30/2023	1.8	37.2	4.41	5.82	0.816	2.71	48.5	91.9	30.6	<1.00
7/13/2023	1	38	3.99	6.36	0.876	3.05	50.5	94.8	36.9	<1.00
7/13/2023	1.8	39.2	3.99	6.47	0.896	2.98	50.8	97.6	38	<1.00
8/15/2023	1	37.4	4.52	6.74	0.998	4.81	56.6	108.3	36.3	<1.00
9/14/2023	1	37.6	5.15	6.98	1.06	3.46	62.8	107.6	36.7	<1.00
10/11/2023	1	34.8	4.62	6.43	1.03	3.51	58.1	74.7	32.9	<1.00
12/12/2023	1	28.6	4.62	5.74	0.94	3.07	45	98.6	26.5	<1.00
3/13/2024	1	26.5	5.36	5.92	0.90	2.77	34.67	56.18	25.58	<1.00
6/27/2024	1	44.4	4.52	7.23	1.08	3.16	37.10	39.36	25.00	<1.00

Note. Alum treatments took place in 2020 (March and July) and June 2023.

Appendix B. Study Design for Expanded Alum Treatment Monitoring

Goal: Over the last few years, Ecology has received feedback and concerns from citizens over possible negative impacts to lake ecosystems resulting from the short-term application of alum as a restoration tool. The goal of this study is to evaluate the required and possible supplemental chemical and biological monitoring parameters for alum treatments.

Objectives: This study would follow a before-after control-impact (BACI) design, monitoring a control and an impacted (alum-treated) lake. The lakes would be monitored for a suite of chemical parameters that are commonly evaluated during lake nutrient budgets and treatment effectiveness monitoring. The lakes will also be monitored for ecosystem indicators such as benthic macroinvertebrates, aquatic plant communities, zooplankton, and fish communities. Each of the parameters will be statistically evaluated to determine the power to detect the impact (alum treatment); the statistical power represents how useful the parameter is in assessing impact.

Timeline: The study would require two years of pre-treatment and two years of post-treatment monitoring. A year of planning and reporting would be necessary, making this a five-year project.

Sampling Design: Water column sampling protocols would follow the EPA's National Lakes Assessment (NLA) (USEPA 2022) and would be 2 – 3 sample locations, depending on the size of the lake. Sampling frequency would generally follow Table B-1, with the exception that sampling during the treatment would follow the permit required frequency, and nutrients and chlorophyll a would be sampled on a weekly basis for three months following the treatment.

Sampling protocols for phytoplankton and zooplankton would also follow the NLA guidance (USEPA 2022), with sampling taking place at a single index location in the center of the lake. Sampling protocols for the assessment of aquatic plant communities would follow the point-intercept method described by Madsen and Wersal (2017). Sampling protocols for benthic macroinvertebrates would follow the NLA guidance (USEPA 2022) for littoral communities and the approach of Smeltzer et al. (1999) for the benthic communities.

Table B-1: Sampling frequency and parameter list.

Parameter	lake inlet	epilimnion	hypolimnion	lake outlet	lake sediment
Nutrients[a]	monthly	monthly	monthly	monthly	—
Total and dissolved metals[b]	quarterly	quarterly	quarterly	quarterly	—
Cations[c]	monthly	monthly	monthly	monthly	—
Anions[d]	monthly	monthly	monthly	monthly	—
Chlorophyll a	monthly	monthly	monthly	monthly	—
Total/dissolved organic carbon	quarterly	quarterly	quarterly	quarterly	
Secchi depth	—	monthly	—	—	—
in situ profile[e]	monthly	Monthly (bi-weekly April–Nov)	Monthly (bi-weekly April–Nov)	monthly	—
Cyanotoxins (microcystin and anatoxin-a)	—	During bloom	During bloom	—	—
Phytoplankton communities	—	monthly	monthly	—	—
Zooplankton communities	—	monthly		—	—
Phosphorus fractions (loosely bound, Fe-P, Al-P, NaOH-P, Ca-P, and inorganic-P)	—	—	—	—	Start and end of project
Total recoverable aluminum and iron	—	—	—	—	Start and end of project
Aquatic plant communities	—	Annual	—	—	—
Benthic macroinvertebrate communities	—	Littoral (annual)	—	—	Profundal (annual)

[a] Nutrients: total phosphorus (TP), ortho-phosphate (PO4), total persulfate nitrogen (TPN), nitrate-nitrite (NO3-NO2-N), and ammonia (NH3-N).
[b] Total and dissolved metals: aluminum, iron.
[c] Cations (dissolved metals): calcium, potassium, magnesium, and sodium.
[d] Anions: bromide, chloride, fluoride, and sulfate.
[e] In situ profile: temperature, specific conductance, dissolved oxygen, pH, phycocyanin/chlorophyll a.

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