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Chemical Precipitation and Inactivation as a Method to Reduce Internal Phosphorus Loading in Lakes

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Introduction

The primary goal of most lake restoration plans is to improve water quality. Water quality is typically defined in terms of clarity, oxygen content, and amount of algae. Lake users want water to be clear, swimmable, fishable and aesthetically pleasing. In aquatic systems, decline in water quality is frequently linked to excess phosphorus in the water column. Phosphorus is generally the limiting factor for aquatic plant growth. High phosphorous concentrations stimulate algal and macrophyte growth, when they die and decompose oxygen is depleted. In extreme cases dissolved oxygen becomes so low that it can lead to fish kills. High phosphorus levels can also favor invading species, alter species composition and reduce light infiltration due to higher concentrations of plankton (Perry and Vanderklein 1996). The effects of increased phosphorus levels are far reaching and include diminished recreation opportunity, poorer quality drinking water, changes in fisheries, and reduction of biological diversity and resilience. Reduced water quality affects both the ecological health of a lake and its cultural value. Consequently, developing a lake restoration plan to reduce phosphorus levels is a high prioity for many lake managers. Some restoration plans may also address toxins or control of exotic species.

The first step in creating a restoration plan is to determine goals for water quality. The depth of acceptable water clarity needs to be established followed by calculations for the necessary reduction in phosphorus and chlorophyll levels to achieve that depth. Testing can then be performed to determine if reductions are achievable and if the water clarity goal is feasible. Plan implementation can commence after feasible goals are set.

Implementation of a restoration plan requires two phases each addressing a different source of phosphorus in lakes. The first phase needs to be watershed-wide to deal with phosphorus loading from external sources. External reduction is generally attained by treating incoming water to remove phosphorus or by diverting run-off away from the lake. The second phase cannot be implemented until after the first phase has been completed. In the second phase, methods are employed to reduce the recycling of phosphorus from sediment into the water column, known as internal loading. Internal loading can be dealt with through biomanipulation, dredging, lake draw-downs, aeration and chemical treatments. Chemical precipitation and inactivation as a method to reduce internal phosphorous loading has been extensively studied and applied. Precipitation removes inorganic phosphorus from the water column while inactivation traps phosphorus in the sediment and prevents its release.

When determining an appropriate method to control internal phosphorus loading, it is important to appreciate the complexity of a lake's biology and chemistry. Information gained from preliminary laboratory and observational work can help determine the best chemical regiment to use for treatment. Proper chemical selection as well as dosing and timing of treatment will greatly improve a plan's potential for success and reduce the possibility of harming lake biota.

This paper will first look at eutrophication, the build up of nutrients in lakes, and how human activities and land use accelerate this process causing lake degradation. Next, the role of internal phosphorus loading as part of the eutrophication process will be examined. Finally, methods of chemical treatments to reduce internal loading will be explored. The main chemical treatment discussed will be alum (aluminum sulfate) as it is the most commonly applied compound in lake restorations. Case studies will be used to illustrate the efficacy of alum treatment, its advantages, disadvantages and limitations.

Eutrophication

One common cause of decline in water quality is eutrophication. As lakes age they naturally undergo eutrophication or a gradual accumulation of nutrients along with a slow decrease in lake depth due to deposits of silt and decaying organic matter. Due to this natural eutrophication process, lakes have finite lifespans varying from a few years to millions of years (Harper 1992). Human activities that alter hydrology and increase nutrient loads into lakes typically speed up this process. This acceleration of lake enrichment is referred to as cultural eutrophication.

Human changes in land use have greatly increased the amounts of nutrients and silt that enter lakes. Most increases in nutrient flow can be linked to urban run-off through storm water systems, effluent from sewage treatment plants, failing septic systems, agricultural run-off, deforestation, and nutrient-rich waste waters from industries such as food processing (Perry and Vanderklein 1996). Cultural eutrophication has increased rapidly since the 1930s with greater reliance placed on artificial fertilizers and sewage treatment plants. An unforeseen result of treating sewage for public and environmental health is the breaking down of organic wastes that then provide a readily accessible supply of nutrients for algae (Harper 1992).

Internal Loading of Phosphorus

In order to effectively plan for lowering in-lake phosphorus, a lake's nutrient budget must be characterized. A nutrient budget identifies the amount of nutrients entering a lake verses the amount removed by flushing, seepage and plant uptake. Lakes can have different ratios of phosphorus being added to the water column by external sources verses internal cycling and sediment release. When assessing the extent of external loading, sources in the entire watershed must be carefully scrutinized. Non-point source loading due to such things as deforestation may not be immediately evident. There have been many cases of reduction in external phosphorus loading that have not brought about expected in-lake phosphorus reductions (Phillips et al. 1994). In these cases, the phosphorus budget was not sufficiently characterized to foresee that internal loading needed to be reduced as well as external loading.

Internal loading of phosphorus through recycling and sediment release is a complicated process with many factors affecting its magnitude. Lake morphometry affects water retention times, how long water resides in a lake, and so determines how long phosphorus can accumulate before it is flushed out (Ryding and Rast 1992). Lake depths determine the size of anoxic zones above sediments, providing conditions for phosphorus release. Deeper lakes have larger anoxic zones. Shallow lakes have a zone of maximum algal productivity directly over sediment. Both anoxic zones and shallow areas over sediment are sources of recycled phosphorus. Internal loading is generally larger in lakes with extensive littoral and wetland areas or with close proximity between epilimnion and anoxic sediments (Cooke et al. 1993).

Other factors influencing phosphorus cycling include climate, surrounding landforms, bottom feeding fish and invertebrates, and numbers of predator fish verses planktivorous fish. Climate determines lake temperature which influences aquatic plant grow as well as dictate seasonal lake stratification and mixing. Surrounding landforms affect wind mixing of water and temperature fluctuations. Bottom-feeding fish excrete both phosphorus and nitrogen into the water column. A study in one southern Minnesota lake showed that for that specific lake, half the annual phosphorus load came from fish excretion (Rydin and Rast 1992). Bioturbation, burrowing activities of organisms in the sediment, can cause exchange of materials between the sediment and water column including phosphorus. Few predator fish allow for a larger population of planktivorous fish which eat zooplankton (Shapiro 1996). When zooplankton abundance decreases, phytoplankton abundance increases. Van Liere et al. (1992) looked at some additional means of phosphorus cycling in shallow lakes in the Netherlands. They found that submerged plants did not play a big role in recycling but zooplankton did. Zooplankton excrete phosphorus that is added to the pool of available phosphorus for phytoplankton which in turn boosts lake productivity.

In some lakes aquatic plants play a role in phosphorus cycling. Carpenter (1982) found a positive feedback loop in shallow lakes vegetated by macrophytes that concentrate biomass near the water surface and have high biomass turnover rates. A positive feedback loop stimulates one process to proceed after another process has occurred. In these lakes, phosphorus released from sediment enhances lake productivity which increases accumulation rates of sediment which increases the surface area to be colonized by macrophytes which increases internal loading. Internal loading is due to the decay of macrophytes which acts as a transport mechanism to move phosphorus from the sediment to the water column. Under some conditions macrophyte management may need to be addressed as part of a phosphorus reduction plan (Morency and Edwards 1985). Macrophytes can also interfere with the ability of chemical treatments to precipitate phosphorus and provide a uniform covering over sediment (Welch and Schrieve 1994). In addition, macrophytes are thought to protect the sediment surface from wind mixing in shallow lakes and hold the precipitated phosphorus in place.

Lake chemistry can vary from lake to lake and can be a confounding factor fore predicting how best to reduce phosphorus levels. Iron and sulfate both play a role in phosphorus release from sediments. Iron binds phosphorus. Release occurs when, under the right conditions, sulphate is reduced and the resulting sulphide removes iron (FeII) from the pore water resulting in lower iron concentrations in the sediment and more phosphorus release (Phillips et al. 1994).

Chemical Treatment Background and Theory

After a lake’s phosphorus budget has been characterized, watershed-wide reduction in external phosphorus loading has been completed, and mechanisms of internal phosphorus loading have been determined, in-lake phosphorus treatment options can be examined. Chemical treatment of phosphorus to reduce internal loading is frequently a component of lake restoration plans. The use of chemical precipitation to remove phosphorus from water has a long history. The first chemical precipitation treatment of lakes was performed in Sweden during the late 1960s and early 1970s. The first lake treated in the U.S. was Horseshoe Lake in Wisconsin in 1970. Aluminum, iron, and calcium salts have been used for centuries to treat drinking water (Cooke et al. 1993). These same salts are the primary chemicals to precipitate phosphorus.

There has been some confusion between chemical precipitation and algicides. Algicides such as copper sulfate are toxic to algae, can impact non-target organisms and are short acting, perhaps only days in duration. Chemical precipitants are generally considered non-toxic and long acting. They reduce algal growth by limiting available phosphorus. Chemical reactions work in two ways to lower a lake's phosphorus content (Cooke et al. 1993). First, precipitation removes phosphorus from the water column by forming insoluble compounds that settle to the lake bottom. The phosphorus is then inactivated by further chemical reactions that prevent its release from the sediment.

Iron can be used for precipitation because of its key role in the cycling of phosphorus. During periods of thermal stratification hypolimnetic anoxic conditions can occur. Anaerobic bacterial metabolism in sediments reduces iron (Cooke et al. 1993). In the reduced state, iron becomes soluble and iron-bound phosphorus moves up into the water column. In oxidized conditions from pH 5 to 7 $Fe(OH)_3$ sorbs phosphorus from the water column and retains it in sediment. Aeration or artificial circulation may have to be used in conjunction with iron treatment to prevent anoxic conditions and subsequent reduction of iron (Cooke et al. 1993). Iron is not commonly used for lake treatment but could be considered for lakes with low pH when aluminum salts cannot be used.

Quaak et al. (1993) indicate that iron can be used for in-lake phosphorus fixation in shallow lakes where sediments are frequently resuspended by wind-wave action, boating, and bottom dwelling organisms. They injected iron (III) into bottom sediment while stirring with water-jets to create intensive mixing. This method successfully reduced total phosphorus, chlorophyll and suspended solids. Long-term results are uncertain since their trial lake still has a large external loading problem. Further studies are recommended to determine the usefulness of this technique.

Calcium carbonate sorbs phosphorus at high pH, greater than 9.0, and forms insoluble compounds. Trials have been done with calcium compounds to precipitate lake phosphorus (Murphy et al.1988). Slaked lime ($Ca(OH)_2$) was added to the surface of a lake in Canada to precipitate phosphorus. Considerable precipitation was formed but all of the precipitate dissolved in the hypolimnion so success was short term. The use of lime has many benefits; it is inexpensive, easy to apply, and does not form toxic compounds. One use for calcium may be in conjunction with aluminum to treat shallow areas of lakes (Cooke et al. 1993). Further research is needed to outline conditions suitable for lime treatment.

Sediment oxidation in shallow lakes is another method using calcium for phosphorus inactivation. Liquid calcium nitrate ($Ca(NO_3)$) is injected into sediment to stimulate denitrification and oxidation of organic matter (Ripl 1976). Oxidation of organic matter will enhance greater binding of phosphorus with ferric hydroxide complexes. Willenbring et al. (1983) used liquid calcium nitrate in laboratory treatments of lake sediment from Long Lake in New Brighton, Minnesota. They determined that a dose of 149 grams nitrogen/square meter would oxidize sediments to a depth of at least 10 centimeters and prevent the release of phosphorus. If oxidation depths do not exceed 10 centimeters, unoxidized sediments could become exposed in the future releasing phosphorus to the water column. Cooke et al. (1993) recommended oxidizing the top 15 to 20 centimeters of sediment. Ferric chloride may need to be added if there is not enough iron present in the sediment. The addition of lime can raise pH, if necessary, to encourage microbial denitrification. Sediment oxidation holds promise for long-term reduction of phosphorus loading but more lakes need to be treated and analyzed. Reported lake treatments have not been successful due to underdosing and failure to adequately reduce external loading.

The most commonly used chemical treatment is alum (aluminum sulfate). When alum is added to lakes with a neutral pH of 6 to 8, a milky solution is produced that quickly forms an insoluble colloidal aluminum hydroxide floc that binds inorganic phosphorus. The floc grows in size and precipitates out of the water column resulting in more visibly clear water within hours. The floc rests on top of the lake's sediment and continues to sorb and retain phosphorus, even under reducing conditions. This floc blanket over sediment prevents phosphorus from being released and added to the internal lake load. The precipitation process also removes organic and inorganic particulate that clears water and reduces available nutrients to algae. Since alum is so frequently used, the remainder of this paper will focus on alum methods, case studies and limitations.

Alum Treatment Methods

The first alum (aluminum sulfate) treatment to reduce in-lake phosphorus was done by applying dry alum to ice on a Swedish lake in 1968 (Cooke et al. 1993). Subsequent alum applications have been done by mixing the aluminum salts in a slurry and applying it to open water. In 1970 a slurry of alum was applied to Horseshoe Lake in Wisconsin (Peterson et al. 1973). The slurry was applied 30 centimeters below the surface through a three meter wide, four centimeter in diameter, perforated pipe manifold. The manifold was pulled behind a small propeller driven boat. The propeller and a 10-15 mph wind provided adequate mixing and dispersion of the chemicals. A dose of 18mg aluminum per liter was used.

Many current applications of alum are being done with modified weed harvesters. The modification utilizes an application manifold, similar to the one used on Horseshoe Lake, but with a hydraulically operated front conveyer that can adjust the manifold from the surface to a hypolimnion as much as two meters below the surface (Connor and Smith 1986). Hypolimnetic application delivers the maximum dose to the anoxic area that is the primary source of internal phosphorus. This method is more costly, slower, and requires more specialized equipment than surface application and cannot be used for oxic littoral sediments (Cooke et al. 1993).

A restoration project on Kezar Lake in New Hampshire used the above modified weed harvester method with one exception (EPA 1995). After an extensive diagnostic/feasibility study was completed, a restoration plan was developed that included copper sulfate treatment one week prior to alum application to kill phytoplankton. As the phytoplankton decomposed phosphorus was released into the water column. It was assumed that the phosphorus tied up in the phytoplankton would continue to recycle in the lake if it was not released prior to alum treatment. This additional phosphorus source was then inactivated in the sediment with the alum treatment.

Jacoby et al. (1982) indicated that it is necessary to use a dose of alum exceeding what is merely necessary to remove phosphorus from the water column. A thick enough floc needs to be produced to sufficiently cover the sediment and prevent long term release of phosphorus. To determine proper dosing, laboratory experiments must be performed using water and sediment from the lake in question. Increments of alum are added to water samples until the desired amount of phosphorus is removed and floc is produced (Cooke et al. 1993). The maximum dose of alum is one that produces less than 50ug of dissolved aluminum per liter of lake water. Water pH needs to be between 6.0 and 8.0 for maximum precipitation and prevention of the formation of toxic soluble aluminum compounds.

Case Studies of Alum Treated Lakes

The Wisconsin Department of Natural Resources and the University of Wisconsin joined together in the late 1960s to establish the Inland Lake Renewal and Management Demonstration Project. They developed a number of criteria to choose a demonstration site for lake restorations. Their criteria included: nutrient concentrations high enough to be able to detect changes, availability of background data on physical and chemical characteristics of the lake, large enough surface area to be typical but small enough to permit treatment and monitoring within their budget, deep enough for persistent thermocline and well- defined hypolimnion, public access, sufficient water retention time to prevent water exchange from masking consequences of treatment, and minimal external loading. They saw immediate positive results after the first alum treatment to Horseshoe Lake in Wisconsin; the depth to the zone of depleted oxygen went from 4.6 meters to 6.1 meters, water clarity improved and there was an absence of algal bloom. Twelve years after treatment there was more eutrophication than immediately after treatment but less than before treatment even though the lake is still receiving external phosphorus loads (Garrison and Knauer 1983).

The first shallow lake treated with alum in the U.S. was Pickerel Lake in Wisconsin in 1973. This is a naturally eutrophic, polymictic lake with no inlets or outlets. Polymictic lakes have frequent or continuous mixing of their water with no thermal stratification. Treatment was unsuccessful due to frequent wind- wave induced mixing that redistributed the floc layer to the center of the lake (Garrison and Knauer 1983). Alum treatment in shallow lakes is frequently unsuccessful due to resuspension and dispersion of bottom material by wind induced waves (Quaak et al. 1993).

Snake Lake in Wisconsin, a softwater dimictic lake, was treated with alum by surface application in 1972. Dimictic lakes are deep enough to experience thermal stratification in the summer and mix twice a year in spring and fall. Snake Lake had been a lake of high water quality until sewage effluent started to be discharged into the lake in 1942 (Garrison and Knauer 1983). The effluent was diverted starting in 1964 but algal blooms and low oxygen conditions continued for several years due to phosphorus release from contaminated sediment. Ten years after alum treatment water quality remained the same as it was immediately after treatment. Although water quality has been improved, the lake remains eutrophic due to continuing storm water input.

In 1986, Eau Galle Reservoir in Wisconsin was treated by hypolimnetic injection of alum. Phosphorus treatment was attempted since internal loading during stratified periods was three to six times greater than external loading (James et al. 1991). Internal loading dropped substantially the first year after treatment. Exceptionally high external loads increased the epilimnetic phosphorus to levels that negated the reduced internal loading by the second year. High external loading is one example of the difficulties presented in managing reservoirs. Reservoirs have continuous and unpredictable changes in inflow, outflow, precipitation, hydraulic residence time, flushing and external loading from the watershed. These factors frequently make the control of internal loading a moot point.

Pros, Cons, and Limitations of Alum Treatment

There are many advantages to using alum as a phosphorus treatment in lakes. Alum treatment is relatively simple to apply over large areas and can be a cost-effective treatment. In properly buffered lakes of pH 6 to 8 aluminum salts will form insoluble $Al(OH)_3$ (aluminum hydroxide) which has a high capacity to adsorb large amounts of inorganic phosphorus (Cooke et al.1993). $Al(OH)_3$ has been shown to have low to zero toxicity to lake biota. Some particulate organic particles such as detritus, can coagulate and become trapped in the $Al(OH)_3$ floc helping to improve water clarity. Floc layers over lake sediments have been proven to have long-term effectiveness in preventing the release of phosphorus from sediment.

The use of alum has the potential to cause negative impacts under some conditions. If alum is added to poorly buffered lakes pH can fall below 4.0 allowing soluble Al^{3+} to dominate rather than insoluble $Al(OH)_3$. Soluble aluminum can be toxic to lake biota making it critical to be able to accurately predict a potential drop in pH and compensate for it, during application, by adding buffers to the alum slurry. Sodium aluminate is often used in conjunction with alum for this purpose. If lake pH rises above 8, aluminum will become soluble which can allow release of phosphorus as well as increase potentially toxic aluminum ions. Lake Morey in Vermont and some other soft water lakes exhibited problems with soluble aluminum after alum application that temporarily decreased density and species richness of benthic invertebrates (Smeltzer 1990). The decrease was an unexpected finding since sodium aluminate was added with the alum to maintain pH. Soft water lakes would be good candidates for further trials with calcium. Cooke et al. (1993) maintain that in nearly all alum treated lakes species diversity and benthic invertebrate populations have been altered to some extent. One unintended consequence of increased water clarity and better light penetration can be increased macrophyte growth, which in some cases can be undesirable.

There are several limitations to the use of alum treatment. Al(OH)3 is not very effective at removing dissolved organic phosphorus which will continue to cycle in the water column. If the floc layer formed by insoluble aluminum hydroxide ($Al(OH_3$) is too thin, sediment dwelling organisms can mix the floc layer with underlaying sediment layers reducing its effectiveness (Ryding and Rast 1989). One solution may be to apply a layer of fly ash over the sediment that would become cement-like and hold the floc in place. This solution has a potential two-fold negative impact. First, what detrimental affect will this have on bottom dwellers and lake ecology? Second, will fly ash introduce toxic substances?

Alum treatment is not able to compensate for continued external loading of phosphorus or other external inputs that can alter in-lake phosphorus cycling. Atmospheric sulfur from the burning of fossil fuel and fertilizer use have greatly increased surface water sulphate concentrations. Studies by Caraco et al. (1989, 1993) have shown that sulphate concentrations in surface water play an important role in phosphorus release from sediments.

Conclusion

Evaluating lakes for treatment success is an important part of any restoration plan. Lakes that have been alum treated should experience reduced spring and fall algal blooms when destratification mixes the hypolimnia with a lake's upper waters (Cooke et al. 1993). Long-term evaluation of alum treated lakes has shown that several conditions need to be met in order for treatment to be successful (Garrison and Knauer 1983). Watershed-wide issues that affect water quality must be addressed and external loading reduced before in-lake treatments are done. Lakes chosen for alum treatment should not be prone to frequent mixing that can redistribute a floc layer. The floc layer should not be denser than the underlying sediment or it will not remain on top of the sediment to prevent phosphorus release. Finally, the treated lake should not have a high sedimentation rate or the floc layer can become buried.

Cooke et al. (1993) noted several additional conditions for alum effectiveness. Since aluminum toxicity can be a problem in softwater lakes alum should not be used for their treatment. Treatment with iron or calcium salts may be a better option, providing aerobic conditions can be maintained. Acidic lakes and lakes with pH higher than 9.0 should not be treated with alum, also due to potential aluminum toxicity.

With so many factors involved in determining if alum treatment will be successful or not it is difficult to generalize recommendations for its use. For the most part, it appears that precipitation and inactivation with alum treatments are beneficial. In many cases where treatment was unsuccessful the causes of failure have been identified and recommendations for improvement noted. For some lakes success is determined by an increase in water clarity, reduced algal blooms and positive feedback from lake users. In other situations, success is determined by demonstrating improvement in a lake's trophic state including a reduction in phosphorus release from sediment and lowered phosphorus concentrations in the photic zone (Cooke et al. 1993).

Phosphorus reduction, as part of a lake restoration plan, needs to be a highly customized process. Much preliminary work needs to be done before developing a restoration plan in order to insure success and not bring about unintended harmful effects.

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