Nippo Lake, Barrington, NH Aluminum Compound Treatment Report, 2021



Photo courtesy of Nippo Lake Association



June 2022

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Forward

"Nippo Lake was notable in southeastern New Hampshire in having crystalline waters like those of Winnipesaukee and Squam rather than sharing the brownish tint of many of the water bodies in Rockingham and Strafford counties. It was a pleasure to gaze deeply into the limpid waters in those days, watching for a big snapping turtle moving slowly across the bottom, knowing we were indeed in a 'New Hampshire everlasting and unfallen' as imagined by Thoreau."

These mid-20th century reflections of a New Hampshire historian, James Garvin, were typical of the impressions of those who knew the unspoiled beauty of Nippo Lake and its quintessentially New Hampshire watershed. That changed on a late spring day in 2010 when, for the first time in its ancient history, Nippo Lake experienced a cyanobacteria bloom. The stunningly unexpected fouling of the lake's waters marked the first in what would become an annual recurrence over the decade to follow. Ironically, the apparent tragedy of the lake's reversal of fortunes was a true "watershed event" in its history, signaling an urgent call to lake residents and New Hampshire's scientific community to link arms in a years-long campaign for its restoration.

In the 12 years since that first cyanobacteria bloom, a dedicated team of Nippo Lake Association volunteers, outside experts and New Hampshire Department of Environmental Services professionals worked tirelessly, contributing their time, treasure and inspiration to studying, planning, managing and implementing myriad aspects of the lake watershed's recovery.

In 2021, on the foundation of this essential work, Nippo Lake was the site of an historic in-lake treatment that promises to break the grip of chronic cyanobacteria blooms, and with continued diligent stewardship of its watershed, restore Nippo Lake to its former place, "everlasting and unfallen", among New Hampshire's treasured natural resources. With heartfelt thanks to all those whose love of our lake, hard work and scientific inspiration made this possible— and with the hope that it may help guide the restoration of other lakes— what follows is the formal report of the treatment of Nippo Lake.

Kevin M. Fitzgerald, Nippo Lake Association.

Executive Summary

Nippo Lake is a 35-hectare mesotrophic lake in Barrington, New Hampshire. From 2010 through 2019 the lake experienced regular cyanobacteria blooms during the summer season. Blooms typically lasted two or more weeks, significantly interfering with the recreational use of the lake. An analysis of historic water quality data documented a significant increasing trend in total phosphorus concentrations, the nutrient that typically limits the growth of photosynthetic organisms in lakes, like algae and cyanobacteria. Additionally, the lack of oxygen in the bottom depths (hypolimnion) of the lake promoted the release of phosphorus from bottom sediments and accounted for 34% of the estimated total phosphorus load and resulted in peak hypolimnetic total phosphorus concentrations in 2016 of 180 ug/L.

A watershed-based plan completed by the Nippo Lake Association in 2019 established a target total phosphorus concentration of 7.2 ug/L in the upper depths (epilimnion) of Nippo Lake. In order to achieve this goal, it was determined that the hypolimnetic total phosphorus load would need to be reduced by 80-90% (10 - 12 kg/yr.) since fall and spring mixing of the epilimnion and hypolimnion supply nutrients to the entire lake.

The use of aluminum compounds (alum) to bind phosphorus in the bottom sediments was identified as the most cost-effective and safe lake management strategy with the highest likelihood of success. To increase the effective longevity of an alum treatment several projects were completed to reduce external nutrient loads to the extent possible (~5 kg/yr.).

The use of aluminum compounds was permitted by the New Hampshire Department of Environmental Services as a "demonstration project" through the issuance of a state surface water discharge permit. The permit carried limits on the types and amounts of chemicals allowed for use as well as limits to certain water quality parameters, conditions specifying required safety measures and monitoring. The permit specified that 65% of the lake area (23 hectares) would be treated over a period of approximately one month. A public hearing and comment period was held to receive input on the project.

The treatments were completed on nine separate days from May 25 through June 17, 2021, and included adding 85,353 liters of aluminum sulfate and 45,092 liters of sodium aluminate to Nippo Lake in all areas deeper than 4.6 meters. The ratio of aluminum sulfate to sodium aluminate was 1.9:1 and the total dose of aluminum was 52 grams / square meter. The treatment resulted in a white flocculant that settled on the bottom of the lake and served as the binder for phosphorus released from the sediment.

On days during which treatments occurred, continuous mean pH readings with the treatment zone ranged from 6.76 to 7.08. Acid soluble aluminum concentrations during treatments were above chronic water quality criteria but below acute water quality criteria. Turbidity remained low (<1 NTU) during treatments. After treatments were completed, pH continued to decline at deep water sites, but this was believed to be largely influenced by the significant rainfalls amounts (36 cm) that fell in the local area in July through August. Acid soluble aluminum concentrations were approximately five times lower the week after treatments ended and continued to decline to background levels from August through October.

Several "response indicators" were monitored to assess the lake conditions for the first four months (July – October 2021) after treatments were completed. In particular, total phosphorus concentrations decreased from approximately 20 to 5 ug/L to in the surface waters. More importantly, in the hypolimnion, total phosphorus concentrations remained relatively constant at around 20 ug/L but significantly lower than peak concentrations observed in 2016. Overall, it was estimated that the 2021 reduction the hypolimnetic phosphorus load, due mainly to bottom sediment phosphorus release, ranged from 70% to 90%.

To date, the project highlights the potential for the use of aluminum compounds to reduce internal phosphorus loads in New Hampshire lakes. Our experience on Nippo Lake documented significantly lower total phosphorus concentrations in the hypolimnion in the months following treatment as compared with historic data. We documented challenges associated with the use of these chemicals in meeting aluminum water quality criteria as the chronic criteria were exceeded during treatment. However, strict adherence to the permitted chemical ratio, creation of multiple treatment zones, extended treatment period (~1 month), and careful real-time monitoring of pH minimized noticeable impacts to aquatic life. Additional monitoring in 2022 will provide a full season assessment of lake conditions 1-year after treatment and likely a better understanding of what can be expected in future years.

1.0 Introduction

Nippo Lake experienced cyanobacteria blooms in 8 of the 10 years between 2010 and 2019 (**Figure 1**). The quantity of algae and cyanobacteria in Nippo Lake was related to the concentration of the nutrient in shortest supply, phosphorus (P). In 2019, a watershed-based plan was completed that identified the sources of phosphorus loading. The total phosphorus nutrient load to Nippo Lake was estimated to be approximately 38 kg/yr. Of that, 13 kg/yr (34%) was identified as coming from internal sources, namely bottom sediments. The remainder of the phosphorus load was identified as coming from watershed sources (44%), waterfowl (5%) or atmospheric deposition (10%). Data from 2016 documented epilimnetic phosphorus concentrations from 10-16 ug/L and a mean hypolimnetic concentration of 95 ug/L with a maximum in October 2016 of 180 ug/L (**Figure 2**). Some of the deep-water phosphorus is



Figure 1. Nippo Lake cyanobacteria bloom, fall 2015. Photo courtesy of the Nippo Lake Association (NLA).

undoubtedly mixed into surface waters over the course of the summer seasons as the epilimnion increases in depth and the remainder is mixed during fall turnover. In Nippo Lake, only about 40% of the volume of water is exchanged every year so a substantial portion of the mixed sediment-derived phosphorus is available in the following growing season to fuel cyanobacteria. It is also likely that cyanobacteria growth was supported near the thermocline during stratification where cyanobacteria "harvested" phosphorus from the hypolimnion and migrated upwards in the water column by using buoyancy regulating gas vacuoles resulting in lake-wide blooms. These three mechanisms likely account for a majority of the internal load. All three are expected to be substantially reduced by this project. Ultimately, the goal of watershed and internal load phosphorus control is to achieve a summer epilimnetic total phosphorus concentration below the mesotrophic threshold of 8 ug/L and greatly reduce the frequency and severity of cyanobacteria blooms.

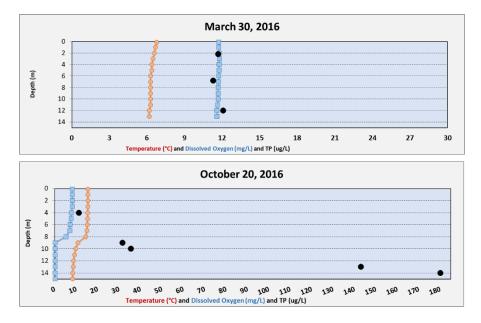


Figure 2. Temperature, dissolved oxygen and total phosphorus from 0 to 16 meters in Nippo Lake, March 30 and October 20, 2016.

The watershed-based plan developed for Nippo Lake included an annual epilimnetic nutrient concentration target of 7.2 ug/L and restoration strategies necessary to achieve these targets. While several of these strategies included reductions in external sources, the internal load represented the largest singular source of phosphorus to the lake. Further, based on the loading analysis, a significant reduction in the internal load was required to meet the in-lake nutrient concentration target. Last, because the internal load is focused during the growing season, it is disproportionately more important in fostering cyanobacteria blooms than sources that are more evenly spread over the year.

To identify the strategy that would best address the internal phosphorus load in Nippo Lake, an alternatives analysis was completed (**Appendix A** - Nippo Lake Treatment Plan). The best option to reduce the internal phosphorus load was identified as the addition of aluminum compounds which bind phosphorus to the bottom sediments even under anoxic conditions. Aluminum treatment was chosen over other internal nutrient management options such as aeration, oxygenation, or dredging to achieve the necessary reduction of internally recycled phosphorus derived from bottom sediments. Aluminum compound treatments target the release of nutrients from bottom sediments, are specific in dosing and the target area of application and require a short-term application phase (days) to achieve a long-term benefit (years). The aluminum compound treatment for Nippo Lake was executed in summer of 2021. The details of the treatment and outcomes are explained below.

To ensure the durability and long-term success of the aluminum treatment, several projects to reduce external phosphorus loads from stormwater and residential sources were addressed in years prior to implementation of an in-lake management action. It would be inefficient to sequester sediment nutrients if there were still unaddressed watershed sources of phosphorus still contributing to the lake.

The goal of the aluminum treatment was to reduce the hypolimnetic total phosphorus load by 80%-90% (10 - 12 kg/yr). By reducing the phosphorus load, the risk of cyanobacteria blooms in Nippo Lake is expected to be minimized for a period of 10-20 years, provided additional external nutrient sources

continue to be controlled. The aluminum compound treatment serves as a demonstration project and was designed to improve the overall condition of Nippo Lake by reducing the frequency and extent of cyanobacteria blooms and, in turn, the length of time that the waterbody is a potential risk to human, pet, and livestock health, as well as increasing the length of time it is suitable for recreation.

The Nippo Lake Association (NLA) served as the oversight and financial entity responsible for hiring the professional expertise necessary to plan, execute and pay for the treatment. DK Water Resource Consulting, LLC (Don Kretchmer, Principal) served as the lead consultant to the NLA with assistance from Water Resource Services, LLC (Kenneth Wagner, Principal). Solitude Lake Management was hired to complete the aluminum treatment. The New Hampshire Department of Environmental Services (NHDES) served as the permitting agency, the lead for identification and remediation of watershed phosphorus sources, and in completing water quality monitoring throughout the duration of the project. The details below describe the plan for the treatment, the permitting process and subsequent requirements, and the outcome of the treatment through the end of the 2021 growing season. The treatment of Nippo Lake with aluminum compounds was the second of its kind in New Hampshire since a project on Kezar Lake in Sutton in 1984 (NHDES 2005).

2.0 Nippo Lake characteristics and historic water quality

Nippo Lake in Barrington, New Hampshire is a 35-hectare (ha) waterbody with a mean depth of 6 meters (m), a maximum depth of 16m, and a flushing rate of 0.43 times per year (**Figure 3**). It was classified as mesotrophic by NHDES in 1982 and 2004. Landcover in the watershed is approximately 65% forested, 27% water or wetland, and 8% developed or open space. Overall, the lake is best described as relatively deep for its size with a small contributing watershed area (174ha; watershed area:lake area ratio = 5.0). The surrounding land use is almost exclusively diffuse residential development with low density two-way paved local and state roads. The immediate shorefront is moderately developed with 40-50 seasonal and full-time residences. There is no public access to Nippo Lake; however, there is an area where lake residents have egress for their boats, but it is not an official public access site. The lack of a formal public access point made the coordination and execution of this project much easier.

Historically, Nippo Lake was monitored through the University of New Hampshire's (UNH) Lay Lakes Monitoring Program (LLMP). In 2015, NHDES summarized the water quality data available to date. The summary documented an increasing trend in the period from 1982 to 2015 in total phosphorus concentrations in the epilimnion that ranged from 5 to 13 ug/L annually, a stable trend in chlorophyll *a* that ranged from 1.2 to 6 ug/L, and a decreasing trend in water clarity as measured by Secchi disc transparency, with a range of 3.5 to 6m. Historic hypolimnetic phosphorus concentrations have ranged from 22 to 109 ug/L with an average of approximately 50 ug/L. Monthly sampling in 2016 by the LLMP and NHDES provided a detailed account of hypolimnetic phosphorus accumulation with concentrations starting at 15 ug/L in March/April and reaching concentrations of 180 ug/L in October (**Figure 2**).

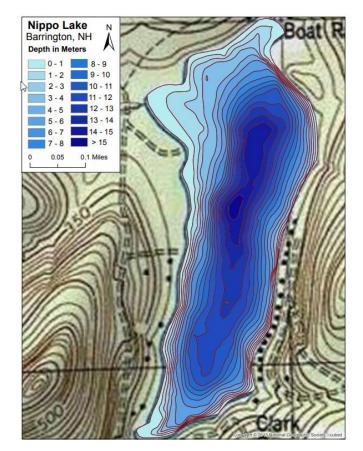


Figure 3. Nippo Lake, Barrington depth contours (1m increments).

3.0 Watershed Planning and Control of External Sources of Phosphorus

In 2016, the NLA, an all-volunteer group working to protect Nippo Lake, began efforts to identify sources of phosphorus loading to the lake. Initially, a phosphorus source assessment was conducted in the north end of the watershed where high priority sites for phosphorus control were identified. With the north watershed assessment completed and local capacity established, the NLA was awarded a 2017 NHDES Watershed Assistance Grant to further develop the watershed management plan, construct best management practices to reduce external phosphorus loading sources from priority locations in the north watershed, and conduct additional assessments for to identify external sources of phosphorus elsewhere in the watershed.

The resulting *Nippo Lake Watershed Management Plan* (December 2019) incorporates output from septic system surveys, lake loading response modeling, internal loading estimates, pollution source assessments for residential properties, roads and other sources, outreach planning, and more. The watershed management plan set a water quality goal of decreasing phosphorus loading to reduce the probability of cyanobacteria blooms. The watershed management plan recognizes that to meet water quality goals for the lake, phosphorus reductions are needed from external as well as internal sources.

To attain the plan's water quality goal, NLA began efforts to implement best management practices (BMPs) to control external sources of phosphorus loading to the lake as described in the watershed plan. To date, implementation activities to control external phosphorus sources have been conducted on

gravel roads and residential properties (**Table 1**). Additionally, the NLA was awarded two Watershed Assistance Grants to address internal phosphorus loading from benthic sediments.

Ongoing efforts by the NLA to control external sources of phosphorus continue as watershed residents implement residential stormwater practices and repairs to another gravel road using funding provided through a 2022 Watershed Assistance Grant. The NLA has also increased land protection efforts in the watershed to prevent future sources of phosphorus loading. Federal funding from NHDES to control external and internal sources of phosphorus loading is matched by financial and in-kind contributions from the NLA.

Management Category	Location	Management Description	Year of Installation	Estimated Phosphorus Reduction (kg/yr)
Roads	Golf Course Way	Road paving and drainage BMPs	2019	3.81
Roads	Flower Drive	Drainage BMP for gravel road	2020	0.60
Residential	Golf Course Way, Sarah Lane, Nippo Court, Flower Drive	Raingardens, water diversion, infiltration trenches, shoreline buffer enhancements	2018 -2021	0.45
Septic Systems	Nippo Court	Septic system upgrade	2018	0.45
-			Total external phosphorus load reduction	5.31

 Table 1. Nippo Lake external phosphorus load reductions through 2021.

The Nippo Lake Association is committed to implementation of the restoration and protection efforts to achieve and maintain the desired water quality for the lake. Implementation of the watershed management plan requires continued collaboration among watershed residents, landowners, town commissions, state and federal agencies, UNH, nonprofit land conservation organizations, and other partners.

4.0 Aluminum Compound Treatment Plan

The treatment of Nippo Lake with aluminum compounds for the purposes of bottom sediment phosphorus inactivation included three primary components; a determination of the area where treatment will occur, the dose or amount of aluminum per area that is needed to successfully bind the chemically available phosphorus in the bottom sediment, and the ratio of aluminum compounds to be added. For Nippo Lake, the treatment area, dose rate and ratio were based on previous water quality data, bottom sediment sample collection and analysis, and prior knowledge of effective aluminum compound ratios used in previous treatment of surface waters in New England. The total area targeted for treatment was 22.7 hectares (ha) (65% of total lake area) and included all portions of the lake greater than 4.6 meters (m) (15 feet) deep (**Figure 4**). This included approximately 15ha where the lake depth was greater than 8m (26 feet) and regularly experienced anoxia (dissolved oxygen <1 mg/L) throughout the summer and fall. An additional 7.4ha was included in the treatment plan at depths from 4.6m to 8m to maximize treatment effectiveness in those areas where temporary anoxia may occur in the sediments and to reduce nutrient contributions from loosely bound and labile sediment phosphorus sources.



Figure 4. Nippo Lake target aluminum compound treatment area.

The dose rate (mass of aluminum per area, grams of Al/m²) was based largely on sediment core samples collected in 2018. Results of the sediment sample effort were detailed in a sediment analysis memorandum. A total dose rate of 54 g aluminum/m² was recommended for treatment based on the sum of the mass of all forms of sediment phosphorus (loosely-bound, iron-bound, and labile organic phosphorus) per dry weight of sediment with a target sediment treatment depth of 10cm and a ratio of 10-parts aluminum (treatment) to 1-part phosphorus (sediment). Typical lake treatment application rates for inactivation of sediment phosphorus range from 10-150 g aluminum/m² (Wagner 2004). The determination of the dose rate based on sediment composition was a key step in the planning of the aluminum compound treatment. The dose rate determined for Nippo Lake required consultation with professionals experienced in the use of aluminum compounds to control internal phosphorus loads.

The two aluminum compounds chosen for use in the treatment were aluminum sulfate $[Al_2(SO_4)_3]$ and sodium aluminate (NaAlO₂). A ratio of 1.8 parts of aluminum sulfate to sodium aluminate were planned for application. The ratio was chosen based primarily on prior projects in New England of similar nature. Aluminum sulfate is an acidic compound. Sodium aluminate is added as a buffering agent to reduce the likelihood of pH conditions in the receiving waters falling below 6.0 units. The pH in Nippo Lake typically averaged around 6.5 in the epilimnion. Additionally, Nippo Lake has low alkalinity in the range of 5.5 – 6.0 mg/L. Thus, Nippo Lake, like many New England surface waters, is highly susceptible pH reductions. Therefore, minimizing the risk of low pH conditions was an important consideration in planning this treatment since aluminum is toxic to aquatic organisms, especially under acidic conditions.

The treatment plan also included specifics on the period over which the compounds were to be added to the lake and the specific locations within the lake where treatment was planned to occur on a given day. In total, the treatment was scheduled to take place on 9 separate days in May/June 2021 that occurred over the course of approximately 4-weeks. The treatment was broken up into three specific treatment periods. A 1-day pilot application followed by two weeks with no treatment and then two (Phase 1 and Phase 2), four consecutive day treatments with phases separated by two days of no treatment.

The plan also called for the lake to be sectioned into five distinct zones of approximately 4-5 ha each, with aluminum compounds to be added to a specified non-adjacent zone on each day of treatment. The extended period of treatment along with the partitioning of lake zones for treatment was planned to minimize the risk of impacts to aquatic organisms due to aluminum toxicity and to accommodate the logistics of chemical delivery (see **Figure 5** for treatment zones).

5.0 Permit for treatment

The authorization for the addition of aluminum compounds to Nippo Lake presented a unique circumstance for NHDES as the only prior use of aluminum dosing was in 1984 in Kezar Lake in Sutton, NH, and there was no clear history on the permitting process that was used for that project. For Nippo Lake, NHDES explored several permitting avenues including consideration of a federal discharge permit (National Pollution Discharge Elimination System, NPDES) through the US Environmental Protection Agency (EPA), a pesticide use permit from the NH Department of Agriculture, Markets and Food (NH Dept. of Ag., RSA 430), and a wetlands dredge and fill permit through NHDES (RSA 482-A). However, none of these options were ideal for a variety of reasons including the temporary nature of the "discharge," neither sodium aluminate or aluminum sulfate are considered pesticides, and the fact that the end result of the treatment was not consistent with the purpose of regulating wetland alteration activities.

Ultimately, the treatment of Nippo Lake was permitted as a "demonstration project" under RSA 485-A:13 (water discharge permits), which gives NHDES the authority to issue permits for the release of certain substances into state waters with the inclusion of limitations relative to water quality criteria, as well as monitoring and reporting requirements. The permit was developed in accordance with administrative rule Env-Wq 300 (Surface Water Protection). In general, the permitting process had several components that included a permit application, an application review by NHDES Biology Section, a public hearing including a comment period, consideration and response to public comments, and the issuance of the final permit. The final permit detailed the treatment plan as described above, specified receiving water limits, included a requirement for an operations and management plan, and monitoring and reporting requirements. As this was the "one-of-a-kind" permit issued by NHDES, it did not represent a perfect process nor is it the exact process that will be used if future treatments of similar nature are requested or recommended.

5.1 Receiving water limits

The state surface water discharge permit (Permit No. Nippo Lake – 001) included limits on the ratio of aluminum compounds to be added and the maximum daily and total dose of aluminum to be added (**Table 2**). Further, the permit included limits for which aluminum (acid soluble) concentrations, turbidity measures and pH levels in Nippo Lake must remain within (**Table 3**).

N/A	Limit of Application	Limit of Application	Limit of Application
Chemical Additive	Approximate Ratio of Application	Maximum Daily Dose (grams of aluminum / m ²)	Permit Dose Maximum (grams of aluminum / m ²)
Aluminum Sulfate , Al ₂ (SO ₄) ₃ ; ~4.4% aluminum by volume	1.8 parts aluminum sulfate : 1 part sodium aluminate by volume	27	54
Sodium Aluminate, NaAlO ₂ ; ~10.2% aluminum by volume	1.8 parts aluminum sulfate : 1 part sodium aluminate by volume	27	54
рН	-	None such that the receiving water limits are exceeded.	None such that the receiving water limits are exceeded.

Table 2. Surface water quality permit limits for aluminum compound additions to Nippo Lake, Barrington, NH.

N/A Receiving Water Limitation		Receiving Water Limitation	Receiving Water Limitation
Receiving Water Characteristics	Daily Event Maximum	Weekly Average	End of Permit Term
Acid Soluble Aluminum (ASA), ug/L	750	87	Pre-aluminum compound application ambient concentration
Turbidity	10 NTUs above conditions prior to treatment	10 NTUs above conditions prior to treatment	10 NTUs above conditions prior to treatment
рН	6.5 - 8.0 Standard Units	6.5 - 8.0 Standard Units	6.5 - 8.0 Standard Units

 Table 3. Limit of receiving water criteria in Nippo Lake Barrington, NH.

5.2 Operations and Management Plan

Prior to the treatment of Nippo Lake, the contractor applying the aluminum compounds, Solitude Lake Management, was required to submit an operations and management plan to NHDES for review that documented the logistics for chemical delivery, transfer, and application. The plan also detailed methods for minimizing and containing potential chemical spillage, emergency contacts, and details for cleaning up the site after the treatment was complete.

5.3 Treatment-related water quality monitoring

To track real-time pH levels within the active treatment zone on each day of treatment, a calibrated field instrument was used to collect continuous pH measurements. The instrument was towed behind a boat at a depth of 1-2m, with the sonde held horizontally in the water column inside a plastic housing to prevent trailing of the sonde and subsequent variation in depth of measurements. Care was taken not to obstruct the sensors of the probe. The boat used to collect continuous pH measures maintained a distance of approximately 50-75m behind the boat that applied the aluminum compounds to ensure that chemicals had been mixed into the water column by the prop-wash of the application boat prior to pH measurement. Additionally, the staff operating the pH boat watched for distressed aquatic organisms within and outside the treatment zone at all times. Prior to each day of treatment, the lake perimeter was surveyed by boat to look for evidence of stressed or dead aquatic organisms.

Ten additional supplementary monitoring locations were established around the perimeter of the lake at evenly spaced intervals (**Figure 5**). Supplemental monitoring locations were checked for pH using a calibrated field instrument during the pilot treatment and on each day of treatment for phase 1 and 2 during pre-treatment, mid-treatment, and post-treatment monitoring events. All pH measures were taken at approximately 0.5m of depth by submerging the instrument's probe into the water and waiting for it to stabilize.

Fixed station, deep site water quality monitoring was required to be completed before (baseline monitoring), during (application monitoring), and after the application was completed (post-application

monitoring). For this, three deep water sites (NIPBARD, NIPALUMS, NIPALUMN) were established (**Figure 6**). Baseline monitoring was completed on a single date in May 2021 at the deep-water sites two weeks prior to adding aluminum compounds and included a temperature/dissolved oxygen profile (1m increments for all profiles), raw water samples collected at the mid-point of the epilimnion, metalimnion, and hypolimnion, a Secchi disc transparency reading, and a vertical plankton haul of upper two-thirds of the total depth (80-micron mesh net, NIPBARD only). All raw water samples were analyzed for specific conductance, turbidity, pH, alkalinity, hardness, dissolved organic carbon (DOC), acid soluble aluminum (ASA), total aluminum, total phosphorus, and chlorophyll *a*.

Application monitoring was conducted on each day during the pilot treatment, phase 1, and phase 2. On each occasion, monitoring occurred approximately 1-hour before the daily treatment began (pretreatment), after one-half of the day's treatment was completed (mid-treatment), and approximately 1hour after the completion the scheduled treatment (post-treatment). Pre-treatment monitoring included the measurement of dissolved oxygen / temperature by profile and a vertical plankton haul of the upper two-thirds of the total depth at NIPBARD. At all three deep water sites, pH and turbidity samples were collected at the mid-epilimnion, mid-metalimnion, and mid-hypolimnion during pre-

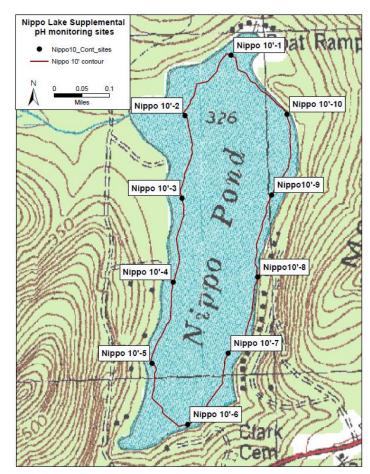


Figure 5. Supplemental pH monitoring locations on Nippo Lake perimeter.

treatment, mid-treatment and post-treatment monitoring. Additionally, for post-treatment monitoring at all deep-water sites pH, turbidity, ASA, and total aluminum samples were collected at the mid-

epilimnion, mid-metalimnion, and mid-hypolimnion. Alkalinity samples were collected the same discrete depth increments at the NIPBARD monitoring location during post-application monitoring. On the day of the pilot treatment, on the last day of the phase 1 and phase 2 treatments, and on each day of post application monitoring, samples were collected for DOC, hardness, total phosphorus and chlorophyll *a* at all three deep site monitoring locations and at each discrete depth interval as described above.

Post-application monitoring occurred at NIPBARD on a weekly basis for the first four weeks after the application had been completed and then monthly for the next three months. Ultimately, this resulted in two monitoring events in June, two in July, and one in each of August, September, and October. Three additional monthly post-application monitoring events are scheduled to be completed from May – July 2022.

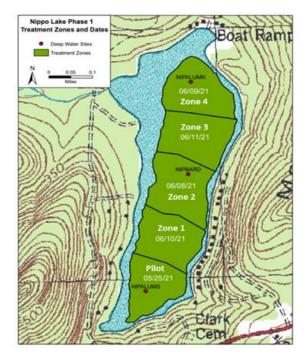
With the exception of continuous pH measures in the treatment zone and pH measures at supplementary monitoring locations, all water quality parameters were processed either in the NHDES Jody Connor Limnology Center or the NH Department of Health and Humans Services public health water lab.

6.0 Results

6.1 Aluminum Compound Treatment Plan Execution

Aluminum compounds were added to Nippo Lake on May 25 (pilot), daily from June 8 - 11 (phase 1), and daily from June 14 – 17 (Phase 2) (**Figure 6**). The pilot area of treatment was 4ha (10ac). For phase one, all treatment zones were 4.7ha (11.5ac). Phase two treatments zones were 5.6ha (14ac). Solitude Lake Management produced maps depicting the tracks of the vessel on each day of treatment (**Figure 7**) based on data from an onboard GPS unit. In general, a crisscross pattern was used within each zone

Figure 6. Nippo Lake, Barrington aluminum compound pilot, phase one, and phase 2 treatment zones, dates of treatment, and deep-water monitoring sites, May-June 2021.



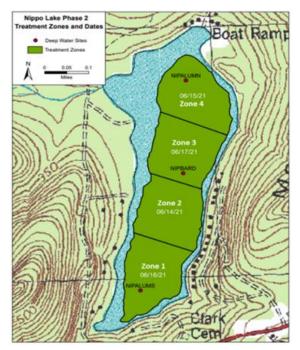




Figure 7. Vessel paths of chemical application within each zone (sector) of Nippo Lake, May-June 2021.

on the treatment day, whereby the chemicals were applied to the entire zone in one direction then applied in the same area in paths rotated by approximately 90 degrees. An onboard flow meter kept track of the volume of each chemical applied, which allowed for a check on the ratio of aluminum compounds (**Table 4**). Overall, 85,353 L of aluminum sulfate and 45,092 L of sodium aluminate were added to the target treatment zone. The average ratio of aluminum sulfate to sodium aluminate was 1.893 (range 1.825 – 1.929). This ratio was slightly higher than the target ratio as real-time monitoring early in the pilot treatment suggested that the pH behind the treatment barge was slightly elevated over ambient pH, but still well within the permitted range. After adjustments were made, pH was kept within a few tenths of ambient and well within the acceptable range (see **Table 6** and **Figure 10** for real-time treatment area pH and ambient pH measures, respectively).

	Application		Aluminum	Sodium	Ratio of Aluminum
Phase	Date	Zone (Sector)	Sulfate	Aluminate	Sulfate to Sodium
	Dute		Volume (L)	Volume (L)	Aluminate
Pilot	5/25/2021	Pilot	7,590	4,054	1.872
Phase 1	6/8/2021	Zone 2	8,275	4,338	1.929
Phase 1	6/9/2021	Zone 4	8,937	4,667	1.908
Phase 1	6/10/2021	Zone 1	8,828	4,577	1.902
Phase 1	6/11/2021	Zone 3	9,070	4,770	1.915
Phase 2	6/14/2021	Zone 2	10,444	5,572	1.977
Phase 2	6/15/2021	Zone 4	10,508	5,758	1.874
Phase 2	6/16/2021	Zone 1	10,815	5,470	1.850
Phase 2	6/17/2021	Zone 3	10,887	5,886	1.825
Tatal		A 11	05 252	45.002	1.893
Total	-	All	85,353	45,092	(average)

Table 4. Volumes and ratios of aluminum sulfate and sodium aluminate added on each day of treatment.

A modification to the application pattern was made in response to the observation of some young-ofthe-year (YOY) fish mortality on 6/11/2021. YOY fish of an unknown species (0.5-0.75 inches long) had been observed in the immediate treatment area on 6/9/2021 and 6/10/2021; the vast majority (thousands) appeared to be acting and swimming normally. On those days approximately 10 stressed YOY fish were observed. It is unknown if observed stress was due to propwash or the treatment chemicals. However, in response to an observation of additional YOY fish stress on 6/11/2021 (approximately 50 individuals), a change in treatment pattern was undertaken whereby consecutive parallel passes of the treatment barge were not adjacent to one another, giving YOY fish a zone of escape on both sides of the treatment path. The non-treated lanes were then filled in later in the treatment day once the flock had settled. Once this change was implemented, observed YOY fish stress was essentially eliminated. The only additional day that YOY fish stress was observed was on 6/15/2021 (<5 fish observed). Thousands of apparently healthy YOY fish were observed throughout the treatment period. Additionally, a few dead adult and juvenile fish of an unknown species (<10) were discovered nearshore on 6/10/2021 after extremely hot weather. Due to their location well out of the treatment zone, it was believed that high water temperatures were the cause of death. Several more dead fish were observed on 6/11/2021 under similar circumstances to 6/10/2021. The weather was much cooler during the second week of treatment and no nearshore fish mortality was observed during that period.

The daily dose (grams Al / m^2 of lake area treated) was determined based on the density (g/L) of each chemical added multiplied by the percentage of aluminum in each compound multiplied by total volume added on a given day, and then divided by the respective area of the lake that was treated. All chemicals for the project were supplied by Holland Company, Adams, Massachusetts. As required by permit, a sodium aluminate solution (4.4% aluminum) with an estimated density of 1,330 g/L (58.7 g Al/L) and an aluminum sulfate solution (10.2% Al) with an estimated density of 1,450 g/L (151.1 g Al/L) were supplied for the entirety of the project. Daily dosages of aluminum averaged 26.1 g/m² and ranged from 24.3 to 27.3 g/m² (**Table 4**). As planned, by combining the pilot, phase 1, and phase 2 treatment areas, the aluminum compounds were added to the entire treatment area twice. In total, this resulted in a total aluminum dose of 52.1 g/m² which fell slightly below the limit of the permit (54 g/m²) (**Table 5**).

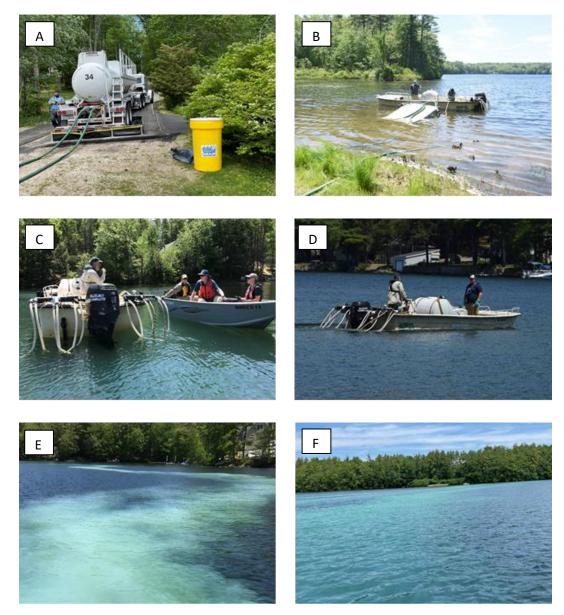
Phase	Application	Zone	Area Treated	Mass (kg) of Al from	Mass (kg) of Al from	Aluminum
	Date	(Sector)	(ha)	Aluminum Sulfate	Sodium Aluminate	Dose (g Al / m ²)
Pilot	5/25/2021	Pilot	4.0	445	613	26.4
Phase 1	6/8/2021	Zone 2	4.7	485	656	24.3
Phase 1	6/9/2021	Zone 4	4.7	524	705	26.2
Phase 1	6/10/2021	Zone 1	4.7	518	692	25.7
Phase 1	6/11/2021	Zone 3	4.7	532	721	26.7
Phase 2	6/14/2021	Zone 2	5.6	613	842	26.0
Phase 2	6/15/2021	Zone 4	5.6	616	870	26.5
Phase 2	6/16/2021	Zone 1	5.6	634	827	26.1
Phase 2	6/17/2021	Zone 3	5.6	639	890	27.3
Total	-	All	22.7*	5,006	6,814	52.1**

Table 5. Mass and dose of aluminum added on each day of treatment.

* The 22.7 ha treatment area was covered approximately twice; once as the sum of the areas from the pilot and phase 1 (22.8 ha) and once as the sum of the areas from phase 2 (22.4 ha). ** The total dose (52.1 g Al / m²) is the sum of the total mass of aluminum sulfate (5,006 kg) and sodium aluminate (6,814 kg) divided by the treatment area (22.7 ha).

The operations and management plan described the process by which chemicals would be delivered, transferred, and applied, as well as the protective measures employed to contain risks associated with spillage or leakage. A tanker truck equipped with divided tanks, one for aluminum sulfate and one for sodium aluminate, was used to transport the chemicals to the lake daily (**Figure 8**). Rigid flexible hoses

Figure 8. Aluminum compound treatment photographs. A. Chemical delivery truck, spillage/leakage containment apron, emergency boom in yellow container, rigid flexible chemical transfer hose; B. transfer of aluminum compounds to application vessel via hoses; C. application vessel and chemical delivery hoses (left) and monitoring support vessel (right); D. application vessel with chemical holding tanks applying aluminum compounds; E. trail of milky-white precipitate (floc) immediately after application; F. Well-mixed aluminum compound floc ~10 or more minutes after application.



were used to transfer the chemicals from the truck directly to separate chemical-specific tanks aboard the treatment vessel. Protective measures included a containment apron around the area at end of the tanker, emergency shut off valves, chemical neutralizing agents, and absorbent booms. On each day of treatment, the vessel applying the chemicals made between 12 to 16 loading trips to transfer chemicals from the tanker truck to the treatment vessel. Two staff people operated the treatment vessel on each day of application. In the case of Nippo Lake, there was no formal boat launch and shallow water depth at the makeshift launch and staging area necessitated the use of a smaller than normal vessel for application. During application, aluminum compounds were discharged from the stern of the boat from multiple trailing, flexible hoses to a depth of approximately 1-2m. The compounds immediately mixed in the propwash behind the treatment boat while reacting to form a milky-white precipitate (floc) of aluminum hydroxide (AL(OH)₃). The floc remained visible for several hours after treatment but quickly started to sink to the bottom of the lake. Within several hours an aluminum hydroxide floc was visible on the lake bottom (**Figure 9**).

Figure 9. Underwater photo showing accumulation of aluminum hydroxide floc on bottom of Nippo Lake following the addition of aluminum compounds. Photo courtesy D. Kretchmer.



6.3 In-treatment Continuous and Supplemental pH monitoring

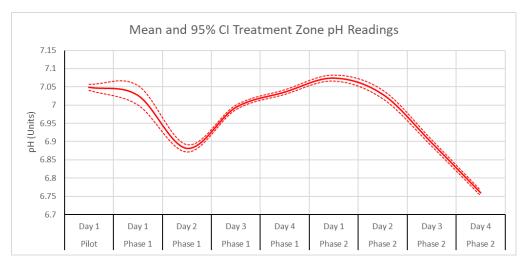
Continuous field measures of pH within the treatment zone successfully tracked the immediate effect the aluminum compounds had on the acidity of Nippo Lake. On each day treatment day between 1,196 and 3,798 field measures of pH were documented within the respective treatment zone (**Table 6**). The daily average of the continuous pH readings within the treatment zone ranged from 6.76 to 7.07. The greatest number of field pH measures less than 6.5 was 431 (12%) on 6/17 on the last day of treatment.

Phase	Day	Date	Zone	n	Min	Max	Mean	Upper	Lower	Count pH	Count pH	Count pH
								95% CL	95% CL	<6.5	<6.0	>8.0
Pilot	1	05/25/21	Pilot	3,237	6.27	7.86	7.05	7.06	7.04	2	0	0
Phase 1	1	06/08/21	Zone 2	1,196	6.28	9.30	7.03	7.05	7.00	39	0	69
Phase 1	2	06/09/21	Zone 4	3,249	5.05	8.00	6.88	6.89	6.87	137	47	0
Phase 1	3	06/10/21	Zone 1	2,703	6.38	7.68	6.99	7.00	6.99	10	0	0
Phase 1	4	06/11/21	Zone 3	2,797	6.21	7.65	7.04	7.04	7.03	28	0	0
Phase 2	1	06/14/21	Zone 2	3,215	5.35	7.70	7.07	7.08	7.07	74	17	0
Phase 2	2	06/15/21	Zone 4	2,784	6.09	7.80	7.03	7.04	7.02	80	0	0
Phase 2	3	06/16/21	Zone 1	3,798	5.55	7.72	6.90	6.91	6.89	239	41	0
Phase 2	4	06/17/21	Zone 3	3,538	5.76	7.22	6.76	6.77	6.75	431	51	0

Table 6. Daily summaries of continuous field pH measures taken within the respective treatment zones during the time of aluminum compound application.

For the rest of the treatment days, field pH measures below than 6.5 were typically less than 3% of all the measures collected on the respective day. The only day when field pH measures exceeded 8.0 was on 6/8. Overall, the mean daily pH within treatment zones from the continuous measures was approximately 7.0. There was little variability in field pH readings within or among treatment days with most readings ranging from 6.75 - 7.1, however, there was a slight, yet gradual decline in daily treatment zone field pH measures from approximately 7.10 to 6.75 during phase 2 (**Figure 10**).

Figure 10. Mean (solid line) and 95% confidence intervals (dashed lines) of continuous pH measures collected during each respective treatment day during the time of aluminum compound application (see table 6 for dates corresponding to the days of the pilot, phase 1, and phase 2 treatments).



Field measures were collected at the supplemental monitoring locations before (pre-treatment), during (mid-treatment), and after (post-treatment) each day when aluminum compounds were added and indicated that mean pH measures ranged from 6.52 to 7.02. Throughout the nine days of treatment,

mean pH measures remained relatively constant with pre-treatment pH readings lower than midtreatment or post-treatment readings on all days (**Figure 11**).

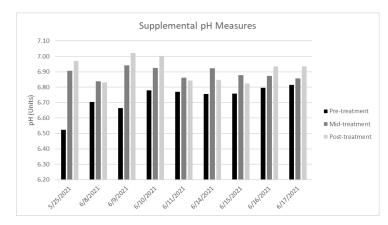


Figure 11. Mean field pH measures taken at supplemental nearshore pH measurement sites before (Pre-treatment), during (Mid-treatment), and after (Post-treatment) on each day aluminum compounds were added.

6.4 Fixed station, deep water site monitoring

Temperature and dissolved oxygen profiles before (baseline), during (pilot, phase 1, and phase 2) and after (post-application) documented that Nippo Lake was well-stratified from May through October (**Figure 12**). Throughout this period the thermocline was positioned from 7 to 10m. As expected, dissolved oxygen dropped to zero or near zero during all dates except 5/4. The depth of anoxia typically occurred around 10m and extended to the bottom (16m at the deepest point). Documentation of stratification dynamics including depth of anoxia throughout and after the treatment was important for estimating phosphorus loading and mass estimates.

6.5 Permit Limit Water Quality Criteria Parameters

Discrete water samples were collected at deep sites (NIPBARD, NIPALUMS, and NIPALUMN) during baseline, pilot, phase 1 and 2, and post-application monitoring events to determine if the water quality criteria permit limits were met.

6.5.1 pH

Mean pH readings from all sites and depths (mid-epilimnion, mid-metalimnion, mid-hypolimnion) on a given day ranged from 5.66 to 6.59 (**Figure 13**). Samples collected on 5/4, prior to treatment (baseline), indicated the mean ambient pH was 6.39. Throughout the treatments (pilot, phase 1 and 2) mean pH remained relatively stable and similar to pre-aluminum treatment, ranging from 6.43 to 6.52. After the applications was completed, from 6/23/2021 to 8/25/2021, mean pH decreased gradually from 6.23 to 5.66. Mean pH then increased in September and was slightly above pre-application and state water quality criteria levels in October. The decrease in pH measured after the treatment was completed through the end of August was believed to be strongly influenced by excessive rainfall in July and August (~36cm, as measured in Barrington, NH). Typical rainfall amounts for New Hampshire in the months of July and August are 17cm combined. Mean rainfall pH in New Hampshire as measured by NHDES from 2000 – 2013 was 4.43 (Nelson et al. 2015) and 5.21 for summer 2021 (W. Henderson, personal communication).

Figure 12. Temperature and dissolved oxygen depth profiles in Nippo Lake, Barrington from May through October 2021. Baseline included profiles from three stations (NIPBARD, NIPALUMS, NIPALUMN). Pilot profile was from NIPBARD only. Phase 1 and phase 2 included profiles collected on each day of treatment, from NIPBARD only. Similarly, all profiles measured from July to October 2021 are for NIPBARD only.

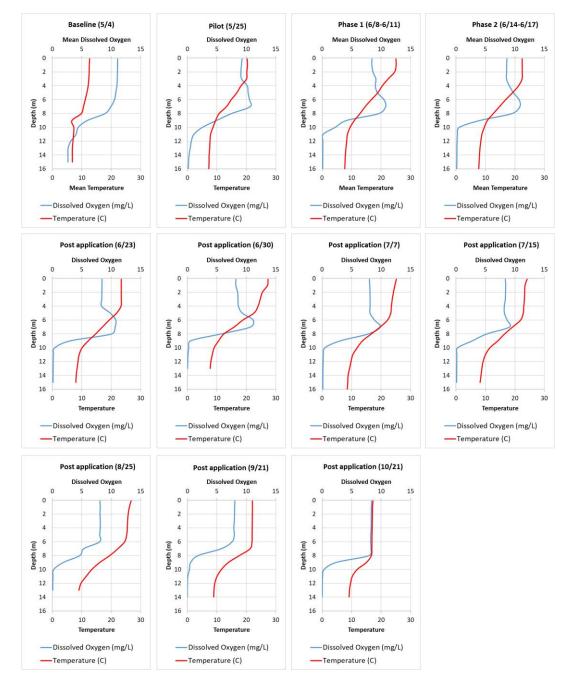
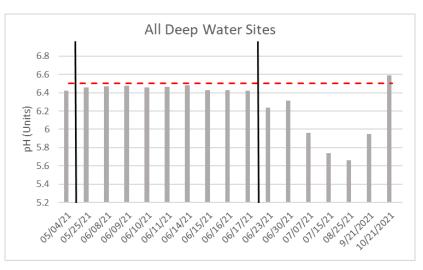


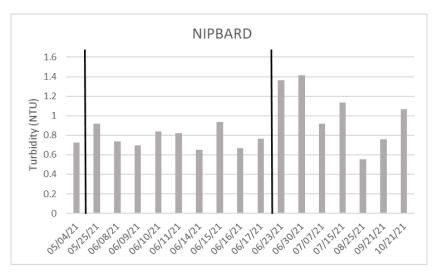
Figure 13. Mean pH readings from deep water monitoring sites (NIPBARD, NIPALUMS, NIPALUMN). Narrow vertical bars indicated the start and end of aluminum compound treatment. Horizontal dashed line is NH state pH water quality criteria (6.5 units).



6.5.2 Turbidity

Turbidity was tracked over time in a manner identical to deep spot pH with values averaged among sites and depths (**Figure 14**). Baseline monitoring mean turbidity was 0.73 NTU. During the application period (5/25-6/17), mean turbidity ranged from 0.65 to 0.93 NTU. In the two weeks following the treatment (6/23 & 6/30) mean turbidity was slightly higher at around 1.4 NTU. In all other post-application monitoring events mean turbidity ranged from 0.55 to 1.13 NTU. At no time did turbidity levels during or after the application period reach 10 NTUs above background (Permitted limit and NHDES water quality criteria).

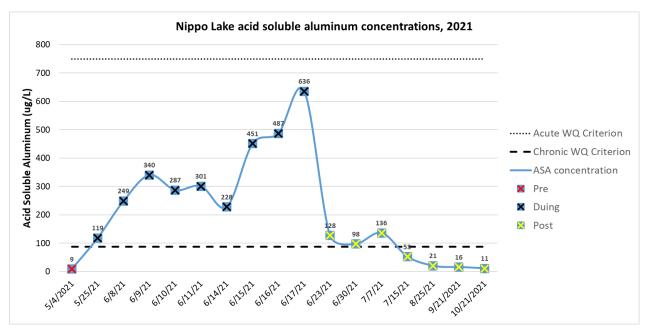
Figure 14. Mean turbidity readings from deep water monitoring sites (NIPBARD, NIPALUMS, NIPALUMN). Vertical bars indicated the start and end of aluminum compound treatment.



6.5.3 Aluminum

Discrete water samples collected at deep sites and processed for ASA indicated that baseline concentrations were 9.4 ug/L in Nippo Lake (**Figure 15**). ASA concentrations increased to 119 ug/L during the pilot treatment, then to a mean of 340 ug/L on day two of phase 1, and finally to a maximum mean of 636 ug/L on day four of phase 2. Post-application concentrations declined quickly from a mean of 128 ug/L to 53 ug/L in the first four weeks following the application period (6/23/2021-7/15/2021), and then gradually from a mean of 21 ug/L to 11 ug/L on subsequent post-application monitoring events (8/25/2021-10/21/2021). State chronic water quality criteria for aluminum (87 ug/L) were exceeded during all days when treatments occurred and on the three weekly monitoring events after the treatment was complete (6/23/2021, 6/30/2021, 7/7/2021). On all subsequent post-application monitoring to 10/21/2021, ASA concentrations were below chronic state water quality criteria. At no time did ASA concentrations exceed acute state water quality criteria for aluminum (750 ug/L).

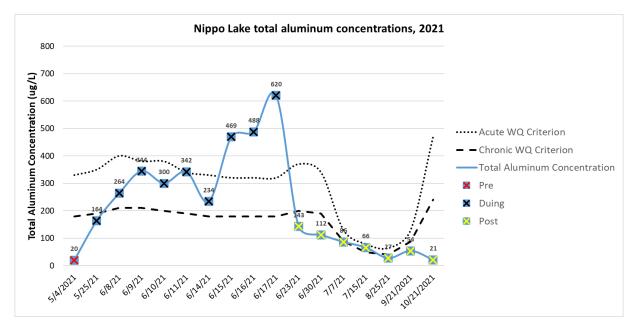
Figure 15. Mean acid soluble aluminum (ASA) concentrations from discrete water samples collected at deep water sample locations (NIPBARD, NIPALUMS, NIPALUMN) for baseline ("Pre"), application ("During"), and post-application ("Post") monitoring events. Current state ASA chronic and acute water quality criteria included as dashed lines.



In the near future, it is possible that NHDES will be adopting new water quality criteria for aluminum as proposed by EPA. These criteria will be based on total aluminum concentrations and change depending on the water's pH, hardness, and DOC. Discrete water samples collected at deep sites and processed for total aluminum indicated that baseline concentrations averaged 20 ug/L (**Figure 16**). Total mean aluminum concentrations increased to 164 ug/L during the pilot treatment, to a maximum mean of 344 ug/L on day two of phase 1, and to a maximum mean of 620 ug/L on day four of phase 2. Post-application concentrations declined quickly in the first four weeks following treatment (6/23-7/15) to mean of 143 ug/L on 6/23 to 66 ug/L on 7/15. Then gradually from a mean of 27 ug/L to 21 ug/L on subsequent post-application monitoring events (8/25-10/21). Under the proposed total aluminum

criteria, the chronic criteria would have been exceeded on all treatment days except for 5/25. The acute criteria would have been exceeded during none of the phase 1 treatment days and three of the four treatment days of phase 2 (6/15-17). During the post-application monitoring events, the proposed chronic criteria would have been exceeded on 7/7 and 7/15. For the post-application monitoring events on 8/25, 9/21 and 10/21 neither the proposed chronic nor acute total aluminum criteria would have been exceeded.

Figure 16. Mean total aluminum concentrations from discrete water samples collected at deep water sample locations (NIPBARD, NIPALUMS, NIPALUMS) for baseline ("Pre"), application ("During"), and post-application ("Post") monitoring events. Proposed total aluminum water quality criteria shown as dashed lines.

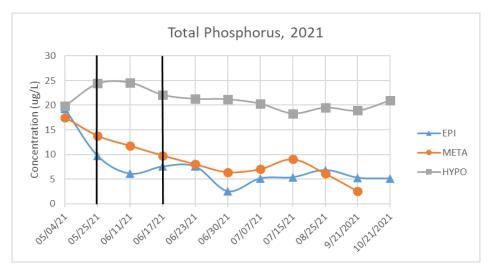


6.6 Aluminum Treatment Response Water Quality Indicators

6.6.1 Total Phosphorus

Mean total phosphorus concentrations on 5/4 during the baseline monitoring event were similar in the epilimnion, metalimnion and hypolimnion and ranged from 17.5 to 19.8 ug/L (**Figure 17**). After each treatment (pilot, phase 1, and phase two), total phosphorus in the epilimnion and metalimnion decreased and ranged from 6.1 ug/L to 9.7 ug/L and 8.01 ug/L to 11.73 ug/L, respectively. Hypolimnetic total phosphorus concentrations during treatments increased slightly from baseline levels and ranged from 22.1 to 24.6 ug/L. Epilimnetic and metalimnetic post-application total phosphorus levels remained low, averaging 5.4 ug/L and 6.5 ug/L, respectively. Hypolimnetic total phosphorus in 2016 averaged 95 ug/L. For context, hypolimnetic total phosphorus concentrations in 2016 averaged 95 ug/L and peaked at 180 ug/L in October 2016. Additionally, historic data from 1982 - 2015 indicates that mean total phosphorus concentrations were 9, 13, and 53 ug/L in the epilimnion, metalimnion and hypolimnion, respectively.

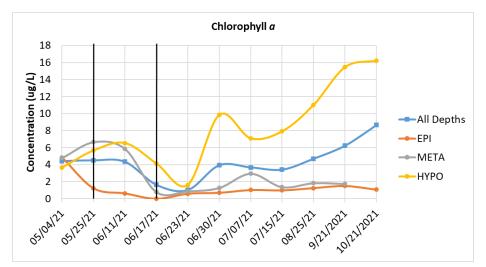
Figure 17. Total phosphorus concentrations for deep spot sites (NIPBARD, NIPALUMS, and NIPALUMN) from May – October, 2021 before, during, and after aluminum compound additions. Vertical bars indicate start and end of treatment.



6.6.2 Chlorophyll-a

The mean chlorophyll *a* concentration during the baseline monitoring event on 5/4/2021 was 4.42 ug/L (**Figure 18**). Mean concentrations from all depths during the treatment period (5/25-6/17) decreased from 4.5 ug/L to 1.64 ug/L but then gradually increased from 1.03 ug/L to 8.65 ug/L from 6/23 to 10/21. The increase in mean chlorophyll *a* concentration was driven by samples collected from the hypolimnion where concentrations increased from 1.62 ug/L on 6/23 to 16.23 ug/L on 10/21. In contrast, chlorophyll *a* concentrations remained below 2 ug/L during post-application sampling events (6/23-10/21) on most occasions in the epilimnion and metalimnion.

Figure 18. Chlorophyll a concentrations for deep spot sites (NIPBARD, NIPALUMS, and NIPALUMN) from May – October, 2021 before, during, and after aluminum compound additions. Vertical bars indicate start and end of treatment.



6.6.3 Plankton Community Dynamics

The plankton community was identified and enumerated on each date of baseline, pilot, phase 1, phase 2, and post-application monitoring. Samples were collected using an 80 um mesh plankton net, lowered to 2/3 depth of the water column at NIPBARD. Samples were preserved in the field with Lugol's solution and analyzed during winter 2021 using NHDES standard methods.

Phytoplankton samples were collected each morning before a treatment began and once again after the treatment was completed. In general, phytoplankton densities ranged between 3000-9000 cells / L before treatment, and an increase trend in plankton phytoplankton abundance was observed from early to May to early June 2021. Densities gradually declined as the treatment progressed from approximately 9000 cells / L on the first day of phase 1 (6/8) and to less than 1000 cells / L on the last day of phase 2 (6/17) then remained low during post-application monitoring (1000-1500 cells / L). A comparison of pre-treatment ("am") and post-treatment ("pm") sample indicated that there was a decline in cell concentration on most days following dosing likely due to sedimentation of algal cells with alum floc.

Chrysophytes (golden browns) mostly *Dinobryon*, but also *Synura* and *Chrysosphaerella* were generally dominant throughout the monitoring period (**Figure 19**). Dinoflagellates (*Ceratium* and an unidentified encysted dinoflagellate) and cyanobacteria (*Snowella, Anabaena* and *Microcystis*) were present in low abundance throughout the monitoring period. Diatoms were also present on occasion in low abundance and primarily included *Asterionella* and *Navicula*.

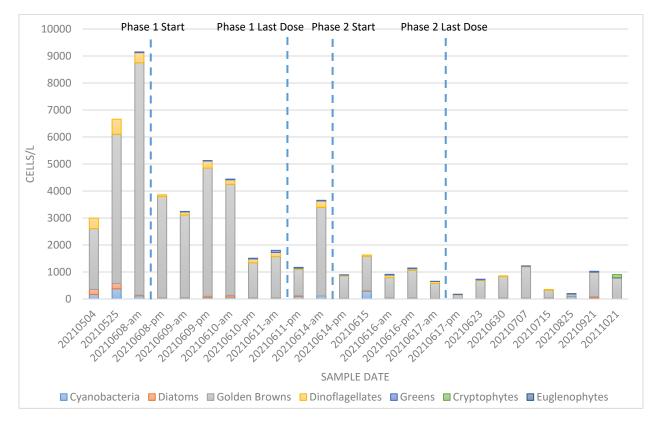
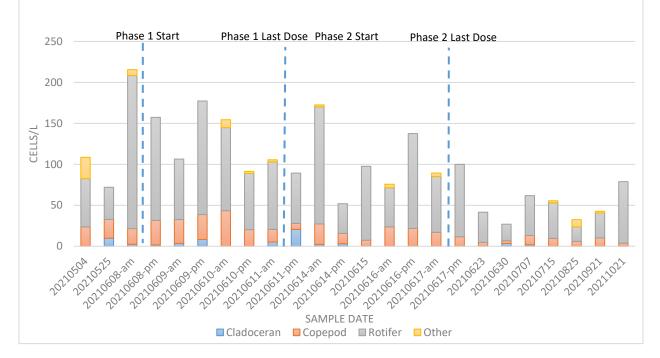


Figure 19. Phytoplankton cell concentration for deep spot (NIPBARD) from May – October, 2021 before, during, and after aluminum compound additions. Dashed vertical lines indicate start and end of treatment.

Like phytoplankton, zooplankton were also collected each morning before treatment began and once again after the day's treatment was completed. All zooplankton sample densities were less than about 225 cells / L and similar to phytoplankton, were lower after the application was completed. Rotifers were the dominant zooplankter throughout the monitoring period, with copepods and other taxa (*Actinophyrs, Chaoborus*) also present in samples (**Figure 20**). While Cladocerans (*Daphnia sp.*) were observed in some whole water samples collected for chemical analysis, especially at NIPALUMN, they were not captured in plankton samples at NIPBARD.

Figure 20. Zooplankton cell concentration for deep spot (NIPBARD) from May – October, 2021 before, during, and after aluminum compound additions. Dashed vertical lines indicate start and end of treatment.



6.6.4 Secchi Transparency

Secchi transparency on 5/4 prior to the start of treatment averaged 5.1m (**Figure 21**). Mean transparency from deep spot monitoring locations during the treatment period (5/25- 6/17) increased slightly and ranged from 5.7 to 6.6m. From 6/23 to 7/13, transparency decreased from 7.3 to 4.5m with some modulation of readings taken in between these dates. From 7/13 to 10/21 transparency estimates more than doubled from 4.5 to 10.2m with September and October estimates averaging 8.6m.

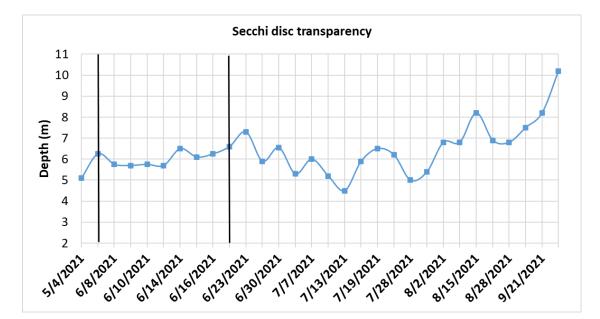


Figure 21. Secchi discs transparency estimates for deep spot sites (NIPBARD, NIPALUMS and NIPALUMN) from May – October 2021 before, during, and after aluminum compound additions. Vertical bars indicate start and end of treatment.

6.7 Reduction in internal loading of phosphorus

The annual internal load of phosphorus is often calculated by subtracting the hypolimnetic concentration at the onset of stratification (typically early June) from the hypolimnetic concentration observed at the peak of stratification (mid-September). Alternatively, in a given year the difference between the epilimnetic and hypolimnetic concentration in mid-September multiplied by the hypolimnetic volume can be used; however, the hypolimnetic concentration at that point in time may also include phosphorus that has settled from the epilimnion over the course of the summer. Because early data were collected prior to treatment and the aluminum treatment resulted in a stripping of the water column phosphorus, it was not possible to calculate the annual internal load data from the onset of stratification this year. Instead, calculating the annual internal load using September data yielded a gross internal load of 4.81 kg/yr. Assuming that 25% of the hypolimnetic phosphorus originated in the epilimnion yields an annual net internal load of 3.6 kg or a reduction of 72% from estimated pretreatment annual internal load estimates (12.9 kg). The magnitude of the internal load estimated for 2021 may not be predictive of long-term conditions as July was one of the wettest on record and external loading of phosphorus to the epilimnion (ultimately settling to the hypolimnion) was likely quite high relative to a typical year. A full year of data post-treatment will likely give a better picture of the true post-treatment internal load and may result in a somewhat different estimate.

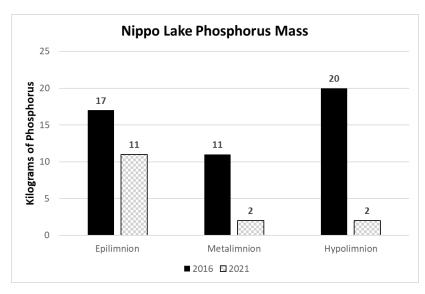
Data was also compared from 2016 and 2021 to estimate the change in phosphorus loads between years. To do this the average total phosphorus concentration and water volume from August – October for the epilimnion, metalimnion, and hypolimnion were determined (**Table 7**). In each year, the concentration and volume averages were then multiplied together to determine the estimated mass in kilograms (kg) of phosphorus contained in each layer. Based on these data, phosphorus loads from all layers were reduced from 2016 as compared to 2021 (**Figure 22**). Epilimnetic and metalimnetic load

reductions were likely a result of the stripping of water column phosphorus-laden particles, like plankton, by the aluminum compounds. In contrast, reductions in the hypolimnetic load by 90% were more reflective of a reduction in the liberation of phosphorus from anoxic sediments, as expected.

Table 7. Average total phosph and 2016.	norus concentration, average	e water volum	ne, and estima	ated phosphorus mass in 2021
	Veen	2010	2024	1

Year =>	2016	2021
EPILIMNION	-	-
Total phosphorus avg. concentration (ug/L)	10	6
Avg. volume (m ³)	1,733,325	1,922,963
Phosphorus mass (kg)	17	11
METALIMNION	-	-
Total phosphorus avg. concentration (ug/L)	21	4
Avg volume (m ³)	522,605	403,735
Phosphorus mass (kg)	11	2
HYPOLIMNION	-	-
Total phosphorus avg. concentration (ug/L)	119	20
Avg volume (m ³)	165,189	94,421
Phosphorus mass (kg)	20	2

Figure 22. Estimated phosphorus load in 2016 and 2021 in the epilimnion, metalimnion, and hypolimnion based on average concentrations and volumes from August through October.



7.0 Discussion

Aluminum compound application to Nippo Lake, Barrington represented the first in-lake management effort of this kind in New Hampshire since 1984 in Kezar Lake, Sutton (NHDES 2005). Because of the long hiatus of this type of treatment in the state, current staff at NHDES had little experience in planning, permitting and overseeing the execution of the project, and there was little historical record other than actual data and reports on the project. As such, the Nippo Lake project was deemed a "demonstration" of the use of aluminum compounds to control internal phosphorus loads in New Hampshire lakes. For this reason, the project was best described as a partnership between NHDES, the NLA, and technical consultants from DK Water Resource Consulting, Water Resource Services and Solitude Lake Management. NHDES served as the lead agency completing the permitting and participating in monitoring activities. NLA provided funding for the project and communicated with the local community. DK Water Resource Consulting, with support from Water Resource Services, were instrumental in much of the technical work used to develop the permit application, develop recommended chemical dosing rates and ratios, provide on-water application oversight, and assist in monitoring activities. Last, Solitude Lake Management served as the lead chemical application and safety contractor.

The final permit was issued by NHDES to the NLA on May 10, 2021, under the authority provided in RSA 485-A:13 and administrative rule Env-Wq-300. The issuance of the final permit included a public comment period (March 19 – May 7, 2021, and public hearing held virtually on April 20, 2021). During the public comment period and after the hearing, NHDES responded to seven inquires all of which dealt with minor details about how the project would be carried out or indicated general support of the project. None of the inquiries were related to concerns moving forward with project.

The execution of the project closely followed the permit. Aluminum compounds were added during a one-day pilot phase (May 25, 2021) and during two subsequent 4-day periods (Phase 1, June 8 – 11, 2021 and Phase 2, June 14-17, 2021). The pilot treatment was used to inform subsequent daily treatments in terms of effectiveness, residual aluminum concentrations and pH, as well as general application and monitoring logistics. On each day of prescribed treatment, a specified zone (pilot, zones 1 - 4) was selected for treatment. On any given treatment day, the zone in which chemicals were applied was non-adjacent to zone from the prior or next scheduled treatment. The purpose of spreading out the application over 9 days was to minimize the potential impact to aquatic organisms and meet the target maximum daily dose. Throughout the project wind speed and direction was light to moderate (0-15 kph) and out of the south. For this reason, treatments were completed in a northern zone followed by the next, non-adjacent zone to the south. This resulted in two to three days between treatments in adjacent zones to minimize the potential of chemicals drifting into the leeward zone that could result in water quality impacts related to pH, aluminum or turbidity.

The pattern by which the chemicals were applied within a treatment zone was initially side-by-side passes in one direction across the entire treatment zone and then using the same pattern in the perpendicular direction within the same zone. However, based on field observations of YOY fish on the first two days of treatment, side-by-side passes were abandoned in favor of more distant chemical application "strips" within the treatment zone leaving a "untreated strip" in between. While it was unclear why the YOY fish were struggling, it was theorized that increasing the space between the

chemical treatment "strips" would allow the YOY fish a better opportunity to avoid the immediate treatment area. After this change was made, few struggling YOY fish were observed.

Daily treatments were dependent on the timing of chemicals arriving at the staging area on the northern end of Nippo Lake. Travel time from Adams, MA (site of chemical production facility) to Barrington, NH was over 3 hours each day. Daily treatments usually began around 10am and lasted to 3pm with monitoring before and after the treatment period each day. Solitude Lake Management and Holland Company instituted the necessary safety protocols and provided records of lading and treatment as required by permit. There were no accidental spills during the application.

Aluminum compound ratios and dosage rates have been documented from several projects in New England in the last 25 years. For 10 ponds treated on Cape Cod, Wagner et al. (2017) reported that the ratio of aluminum sulfate to sodium aluminate was typically 2:1 and that dose rates ranged from 25 g/m² to 100 g/m² but were usually around 50 g/m². For the treatment of Ticklenaked Pond in Vermont in 2014, the Vermont Department of Environmental Conservation prescribed ratio of 2:1 (aluminum sulfate : sodium aluminate) and dose of 60 g/m² for a majority of the treatment area with a smaller area receiving a dose of 105 g/m² (Aquatic Nuisance Control Permit #2014-001, May 2014). For East Pond, Maine in 2018, the Maine Department of Environmental Protection set chemical the ratio to 2:1 and the dose rate ranged from 35-50 g/m² depending on the area of the lake being treated (MEPDES permit #ME0002755, May 2018).

For Nippo Lake, the NHDES permit prescribed a ratio of aluminum sulfate to sodium aluminate of 1.8:1 as recommended by the technical consultants. This is slightly lower that the typical 2:1 ratio used for other similar projects in New England. Nippo Lake has a low pH, around 6.5 historically, but was lower, 6.39 during baseline sampling. Further, Nippo Lake has little to no acid buffering capacity with an average alkalinity of 6.54 mg/L. Thus, it is highly susceptible to acidic inputs. Aluminum sulfate when dissolved in water releases free hydrogen ions contributing to low pH conditions. Sodium aluminate is a basic solution that balances the acidity of aluminum sulfate. A slightly lower ratio of aluminum sulfate to sodium aluminate was permitted for use in Nippo Lake to minimize the potential for pH reductions and aluminum toxicity. The average ratio of aluminum sulfate to sodium aluminate was 1.9, just slightly above the permitted limit. This ratio was intentionally adjusted slightly upward during the early phases of the treatment in response to an observed slight increase in real-time pH as measured behind the treatment vessel.

Based on sediment analysis and a target sediment treatment depth of 10cm, a total dose of 54 g of aluminum / m^2 of treatment area was recommended and was similar to other projects in New England. Generally, an upper limit of 25 g/m²/day is recommended in order to avoid the potential for negative impacts from aluminum (Wagner et al. 2017). For Nippo Lake, a slightly higher maximum daily dose of 27 g/m² was established by permit based on recommendations from the project technical consultants. In turn, this required that the treatment area be treated twice to achieve the permitted target dose of 54 g/m². Dosages of aluminum added to Nippo Lake during each treatment day never exceeded the permit limit and averaged 26.1 g/m² per day in the treatment zones. Each area was treated twice, resulting in 52.1 g/m² over the course of the application.

Water quality monitoring permit requirements placed a high priority on tracking pH. The aluminum compounds used in the project affect the ambient acidity of the waters to which they are added and can

result in increased aluminum toxicity if pH is driven below 6.0 or above 8.0. The acidity of Nippo Lake waters was tracked in three ways, continuous surface measurement in the immediate treatment area, field measures at supplementary monitoring locations around the perimeter of the lake, and at discrete depths at deep water monitoring sites. Continuous surface measurements in the immediate treatment area indicated daily average pH measures remained above NHDES surface water criteria (>6.5) during all treatments ranging from 6.76 to 7.07 with approximately 4% and 0.2% of all continuous measures falling below 6.5 or above 8.0, respectively. Average daily supplemental pH measures were above the 6.5 criterion and ranged from 6.52 to 7.02 indicating that during the treatment, pH around the perimeter of the lake was not impacted. Deep water site baseline average pH measures were lower than continuous and supplemental pH measures and averaged 6.41. The lower pH at deep water sites was a result of the inclusion of hypolimnetic samples which had an average pH of 5.91 (NIPBARD only). Post-application pH monitoring at the deep-water sites averaged across all sample depths documented a substantial decrease in average pH from approximately 6.4 during the treatment to 5.66 on 8/25. This may, in part, have some linkage to addition of aluminum compounds. However, the data from the continuous and supplemental pH measures taken during the treatment are contrary to that conclusion. Rather, declining pH at deep-water monitoring sites in July and August were believed to be primarily a result of frequent rain events. From July into August, New Hampshire experienced an unusually high amount of rainfall with almost 36cm (14 inches) of rain recorded in Barrington, NH. Based on NHDES records of rainfall, the pH is typically around 5.0 (2021 mean = 5.2, personal communication, W. Henderson, 2000 - 2013 median = 4.43, NHDES report R-WD-15-5). Additionally, the naturally low alkalinity of Nippo Lake (6.54 mg/L), likely made the lake highly susceptible increasing acidity due to the large quantity of low pH rainwater. Data from five nearby lakes documented similar declines in average pH from 6.39 in June to 5.97 in August (NHDES volunteer lake assessment program; Ayers Pond, Northwood Lake, Pawtuckaway Lake, Pleasant Lake, Harvey Lake).

Overall, the continuous field measures were most useful in tracking changes in pH in real-time, while the supplemental measures during the treatments were helpful in documenting the lack of changes in pH outside the treatment area. Deep water site pH monitoring, while helpful was confounded by external environmental factors that were not associated with the treatment. Further, it seems that pH impacts, if not observed during the treatment, are unlikely to occur after the aluminum compounds are added, and therefore, are not a critical part of post-application monitoring, if pH during treatment is kept within an acceptable range.

Turbidity was documented to determine if the floc produced by of the addition of aluminum compounds resulted in water quality impacts. As shown in **Figure 7** and **Figure 8**, the treatments resulted in a white precipitate ("floc") of aluminum hydroxide. The floc was concentrated as a trail behind the track of the application vessel initially and quickly mixed downward and outward. After a daily treatment was complete, the treatment area typically appeared aqua-blue. The white precipitate ultimately settled on the bottom sediments and served as the active material to bind sediment phosphorus and reduce internally loading. Over time the aluminum mixes with bottom substrates, likely through physical and chemicals means, and through disturbance of the sediments by benthic dwelling organisms (Welch and Cooke 1999).

State water quality criteria dictate the turbidity cannot increase more than 10 NTUs over background. Baseline monitoring at deep water sites indicated the average turbidity was 0.73 NTU. During the entire application period, average turbidity ranged from 0.65 to 0.93 NTU and never exceeded 2.5 NTU. While turbidity was well within the permit limits, monitoring for this parameter is important given the distinct visual effect of the treatments and the direct linkage to state water quality criteria. It is also important to note that while turbidity remained low during the treatments, the observation of YOY fish struggling during the initial treatment days, may have been a result of floc material on gill filaments. While not quantified, the alteration of treatment paths to reduce floc density in the immediate treatment zone seemed to reduce the negative effect on larval fish by allowing escape from the active plume.

Aluminum toxicity represents the primary threat to aquatic life when treating lakes with aluminum compounds. As noted above, aluminum is most toxic to aquatic organisms when the water's pH is below 6.0 or above 8.0 (Gensemer and Playle 1999). Current New Hampshire chronic and acute water quality criteria use the acid soluble fraction (ASA) of aluminum rather than total concentration of aluminum (NH water quality criteria: chronic = 87 ug/L; acute = 750 ug/L). Baseline average ASA concentrations from deep spots were less than 10 ug/L. Treatments resulted in an immediate increase in concentrations above the chronic state water quality criteria which continued throughout the application, peaking at 636 ug/L on the last day of treatment (6/17). The concentrations dropped from the maximum on 6/17 to 128 ug/L in one week yet continued to be in exceedance of the chronic criteria until 7/15. At no time were the current acute water quality criteria for aluminum exceeded.

Currently, the EPA is requesting that states adopt new aluminum water quality criteria that are based on the total aluminum concentration. These criteria are variable and depend on the water's pH, hardness and DOC. To demonstrate how this would apply to Nippo Lake, the proposed criteria were computed based on EPA's MS excel calculator and compared against total aluminum concentrations from samples collected at deep spots. The proposed chronic criteria were exceeded less frequently (9 of 17 sample events) as compared current chronic criteria (12 of 17 sample events). On most treatment days the proposed chronic criteria were greater than 180 ug/L versus 87 ug/L under the current water quality criteria. However, after the application was complete on 7/15 and 8/25, the proposed chronic criteria were lower at 49 ug/L and 40 ug/L, respectively, which coincided with the dates with the lowest deep spot average pH readings. The proposed acute aluminum criteria were substantially lower than the current criteria (750 ug/L) on all days when water monitoring was completed and ranged from 64 ug/L to 400 ug/L. On three days during the application of aluminum compounds the proposed acute criteria were exceeded (6/15, 6/16, 6/17). The proposed acute criteria declined from 6/23 through 8/25 but then increased in September and October which coincided with an increase in the pH of Nippo Lake water.

Based on the experience at Nippo Lake, it seems unlikely that aluminum compound treatments could avoid exceedances of the current state chronic aluminum water quality criteria or either the EPAproposed chronic or acute criteria. When treatments were occurring, current chronic criteria were exceeded on all days and seven of eight days for the proposed criteria. Acute criteria exceedances were less frequent, but nevertheless did occur on three of eight treatment days under the proposed criteria. Wagner et al. (2017) indicates the upper total aluminum concentration to be avoided for impacts to aquatic life is 5,000 ug/L during aluminum compound treatments provided that pH is kept within the target range. Presumably, this high concentration assumes the toxic fraction of aluminum compounds is minimized and is best suited for waters with higher pH, DOC and buffering capacity (hardness). Aluminum is a complex element that assumes many forms under specific environmental conditions and in the presence of variety of ions (Cooke et al. 2005, Gensemer and Playle 1999). It is well known that the acidity of the water has a major impact on aluminum toxicity and that harmful impacts can be minimized if pH is maintained between 6.0 and 8.0. Throughout the application phases, continuous pH monitoring data in the treatment zones, field measures at supplemental sites, and at deep water monitoring sites documented lake-wide pH. During the application phases, in-situ continuous measures indicated that the pH remained well within this range, minimizing the formation of aluminum compounds that are toxic to aquatic life. From July through September the average pH of Nippo Lake was low (<6.0) based on deep-site monitoring site sample results, however at this time, the vast majority of the aluminum compound floc had already settled to the bottom of the lake and had likely become partially integrated with the bottom sediments, presumably lessening the availability of aluminum to the overlying waters.

In addition to pH, DOC and hardness impact aluminum toxicity and, for this reason, have been incorporated into the aluminum criteria proposed by EPA. For this project 56 DOC samples were collected as part of the monitoring efforts and averaged 2.55 mg/L (range 1.6-3.2 mg/L). A total of 56 hardness measures averaged 13.2 mg/L (range 12.3-17 mg/L). Thus, while both of these parameters were relatively low, they were stable before, during, and after the application and probably did not contribute to increased aluminum toxicity as a result of the aluminum compound application.

Nevertheless, aluminum toxicity in lakes with relatively low pH, minimal buffering capacity (hardness), and low DOC is an important factor to consider when planning aluminum compound treatments in lakes to control internal phosphorus loading. As described above, the Nippo Lake treatment was planned to include use of sodium aluminate to buffer the aluminum sulfate and an extended application timeframe was followed with rest periods in between treatments (pilot, phase 1, and phase 2). Additionally, the waterbody was broken up into zones and non-adjacent zones were treated on consecutive days. These basic project execution logistics were helpful in minimizing the risk of potential impacts to aquatic organisms. One consideration for future aluminum compound treatments in waters like Nippo Lake, that are lower in pH, DOC, and buffering capacity, might be to separate the treatments into more distant phases (such as spring and fall) in an attempt to avoid aluminum water quality criteria exceedances. However, based on our experience at Nippo Lake, it is still likely that exceedances of the chronic criteria may occur if consecutive days of treatment occur. Lastly, projects of this nature must accept some inherent risk and even, perhaps, assume a short-term (weeks) environmental impact, such as the temporary exceedance of aluminum water quality criteria, as they are designed to produce long term benefits (i.e. significant reduction in internal phosphorus loading for 10 – 20 years).

In the period following the application when Nippo Lake was monitored in 2021, four response variables were monitored that documented marked improvements over baseline and historic data. First, total phosphorus was historically <10 ug/L in the epilimnion but had increased to over 12 ug/L in more recent years (2010-2015; **Appendix B**). On May 4, 2021 during baseline monitoring, total phosphorus concentrations were approximately 19 ug/L. Epilimnetic concentrations decreased to between 5 - 10 ug/L during the application and remained near 5 ug/L for the remainder of the post-treatment sample events in 2021. The pattern of metalimnetic phosphorus concentrations in 2021 mimicked epilimnetic concentrations but were slightly higher.

Most importantly, hypolimnetic total phosphorus concentrations began at 20 ug/L in 2021, lower than most historic annual summer medians (range 15 – 109 ug/L). However, unlike hypolimnetic samples in 2016 when total phosphorus concentrations averaged 95 ug/L and peaked at 180 ug/L, 2021 concentrations were comparable to baseline measures, remining around 20 ug/L through October 2021. The low late season hypolimnetic concentrations in 2021 occurred despite continued anoxic condition below 10m, indicating that the aluminum was effective in controlling the release of sediment-bound phosphorus. In fact, based on results from 2021, the estimated annual internal load was 3.6 kg/yr, a 72% reduction from pre-treatment annual internal load estimates. Further, when the hypolimnetic phosphorus load from 2016 was compared to 2021 using average hypolimnetic total phosphorus concentrations and water volumes from July through October, a reduction from 20 to 2 kg (90%) of phosphorus was documented. Additional monitoring for 2022 will allow for another estimate in internal load reduction following the aluminum compound treatment.

Despite recent cyanobacteria blooms, the historic chlorophyll *a* concentrations in Nippo Lake have remained relatively stable since the mid-1980s ranging from 2 to 4 ug/L. On 5/4, 2021, during the baseline monitoring of deep spots, the lake-wide average chlorophyll *a* concentration was 4.42 ug/L. An immediate decrease was observed during the treatment period when the average concentration declined to 1.64 ug/L, a second indicator that the floc associated with the addition of aluminum compounds incorporated some of the pelagic algal community. Chlorophyll *a* concentrations remained below 2 ug/L and 4 ug/L after the treatment in epilimnetic and metalimnetic samples, respectively probably reflecting the limited availability of phosphorus in these upper layers. However, chlorophyll *a* concentrations in the hypolimnion increased dramatically following the application reaching a maximum of over 16 ug/L on 10/21. The rapid and dramatic increase in chlorophyll *a* in the hypolimnion may have been a result of a concentrated layer of phytoplankton that were able to absorb light at lower depths than in the past due to increased water clarity. It is also likely that while hypolimnetic phosphorus concentrations following the application were substantially lower than historic levels, they remained much higher than concurrent concentrations in the epilimnion or metalimnion (20 vs. 5 ug/L) and mobile phytoplankton were selectively descending to the lower depths to utilize the higher nutrient waters.

Reductions in phytoplankton populations are expected following aluminum treatment and have been documented in other studies (Holz and Hoagland 1999, Dawah et al. 2015). In Nippo Lake, 2021 plankton community dynamics were relatively stable and indicative of a mesotrophic lake. Phytoplankton densities were between 3000– 9000 cells/L when the project began, and the community was primarily comprised of golden algae (*Dinobryon*). Zooplankton densities were about 100-250 cells/L during the baseline monitoring and just before the pilot treatment began. Rotifers and copepods made up >90% of the zooplankton community. Densities of phytoplankton and zooplankton decreased throughout the treatment phases supporting the hypothesis that the aluminum compounds had a temporary impact on the plankton community by binding the organisms into the floc. After the treatments were completed, plankton densities remained below ambient conditions through October.

Water clarity following the treatment of Nippo Lake with aluminum compounds exceeded historic Secchi disc transparency readings which tended to vary between 3m and 5m as an annual average. Prior to beginning the application, the Secchi disc transparency was 5.1m (5/4) but more than doubled from 4.5m to 10.2m from mid-July to October. As noted above, increased water clarity was likely of result of, the binding capacity of the aluminum compounds that probably stripped some of the existing plankton

and particulate matter from the water column during treatment. Second, the reduction in the release, and consequently availability, of phosphorus from the bottom sediments likely reduced the growth of all types of phytoplankton. As a response indicator, the improvement in Secchi disc transparency is an important public communication tool. The dramatic and obvious increase in water clarity experienced at Nippo Lake in the months following treatment was satisfying to the lake's recreational users and NLA who provided financial support for the project.

The application of aluminum compounds to Nippo Lake provided an opportunity to test the effectiveness of a well-known in-lake management technique used to control the internal loading of phosphorus from bottom sediments. As noted, however, the use of aluminum in this fashion in New Hampshire has been limited to date with only Kezar Lake, Sutton receiving a similar type of treatment in 1985. Here, data from the Nippo Lake in the year immediately following the chemical application demonstrate a significant reduction the phosphorus load contained in the lake, especially the contribution of hypolimnetic phosphorus from bottom sediments, as expected. In turn, favorable immediate improvements were observed, namely in water clarity (Secchi disc transparency). Ultimately, one of the important goals of the treatment was to reduce the incidence of cyanobacteria blooms. While cyanobacteria were observed infrequently in plankton samples from 2021, future monitoring in 2022 and beyond are necessary to determine if this goal is met.

Critical to the long-term success of the use of aluminum compounds as an in-lake nutrient management technique is the prior control of external nutrient sources (Marsden 1989). At Nippo Lake, the NLA worked with NHDES, consultants, and engineers to identify and control external sources before considering the use of aluminum. Through this process improved stormwater drainage and pavement was applied to a dirt road to correct excessive erosion and several landowner stormwater controls were implemented to reduce runoff. Except for a few additional minor external sources, there were no other unnatural nutrient sources to control. Going forward, NHDES will require that similar efforts be used to managed external sources, to the extent possible, prior to considering permit applications for the use of aluminum compounds.

An important item that made the Nippo Lake project less complicated logistically was the lack of a public access facility. This simplified the application process allowing Solitude Lake Management and the monitoring crews to complete the planned work without interference. Prior to, and during the application, the NLA informed lakeside residents of the upcoming treatment via email and posted signs around the lake so that no recreational use of the lake took place during or immediately after the treatment. Additionally, water withdrawals were eliminated while treatments were ongoing and for 24 hours following completion. Future treatments on waterbodies with public access points and a significant number of withdrawals could be more challenging and will require a broader outreach campaign to inform the public that all or part of a waterbody receiving treatment will be off limits while treatments are ongoing and for a short period after.

The application of aluminum compounds to Nippo Lake required significant advanced planning. The planning took place over a period of approximately 1.5 years. One key component of the planning was sediment sample collection and analysis to determine the amount of aluminum that was required to bind the various fractions of sediment phosphorus. Additionally, a significant amount of communication was undertaken by the NLA to inform and receive input from lakeside residents and the local

community. From a regulatory standpoint, since this was a unique project that was outside the norm of NHDES' current permitting process, several permitting options were considered, none of which were ideally suited. Ultimately, the use of state surface water discharge permit was identified as the best option. It provided a means to direct amounts and ratios of aluminum sulfate and sodium aluminate to be added, the limits of critical water quality parameters, and special conditions relative to the application plan, reporting, safety, and water quality monitoring. It was, however, not perfect, and future projects of this nature in New Hampshire would benefit from changes to the state's current water quality laws and administrative rules. In particular, consideration should be given to changes that allow for short-term exceedance of water quality criteria, especially pH and aluminum, where it can be shown that impacts to aquatic life will be minimized, and long-term improvements of water quality parameters can be expected.

Based on our experience at Nippo Lake, maintaining a pH between 6.5 and 8.0 can be achieved by managing the ratio of aluminum sulfate to sodium aluminate based on real-time continuous pH monitoring and injection rate monitoring of the chemical compounds. By maintaining a pH in the target range, the potentially toxic effects of aluminum additions will be largely avoided. For this project, the only impacts observed were the observations of low numbers of struggling YOY fish. The impact was reduced by altering the pattern of the treatment vessel.

As previously noted, the use of aluminum compounds to reduce the liberation of phosphorus from bottom sediments under anoxic conditions can be effective for 10 – 20 years or more, depending on a variety of parameters and characteristics of treated waterbodies (Huser 2012, Huser et al. 2016, Welch and Cooke 1999). In NHDES' previous experience using this lake management technique at Kezar Lake in Sutton, the benefits have exceeded that timeframe by more than a decade (NHDES 2005, NHDES VLAP data). The application period on Nippo Lake took place over nine days of actual treatment spread out over approximately 3.5 weeks. Exceedances of water quality criteria for aluminum lasted for approximately one month. It seems that the potential benefit of this treatment, namely the reduction cyanobacteria blooms and resulting public benefit of increased recreational opportunities, far outweigh the short-term exceedance to numeric chronic water quality criteria. However, future projects of this nature, should continue to respect the water quality criteria that are in place and plan treatments accordingly using strategies such a treatment zones, application phases that extend over period of days to weeks, and monitoring programs that inform the frequency of water quality criteria exceedances and make visual observations of potential impacts to aquatic life. While greater costs are incurred in applicator fees for longer-duration treatment periods, the tradeoff is increased safety margins during treatment for the protection of aquatic life.

In summary, the application of aluminum compounds to Nippo Lake provided a unique opportunity to make much needed improvements to water quality while simultaneously addressing the need by NHDES to consider the use of a well-known and researched in-lake management technique. The short-term results (<1 year) seem to indicate the treatment was successful, in large part, because of partnerships between the NLA, technical consultants and NHDES. There are several other lakes in New Hampshire that regularly experience cyanobacteria blooms and are known to have high levels of internal phosphorus loading. Therefore, it is likely that additional projects that recommend the use of aluminum compounds as a restoration tool will be proposed in the coming years. Thus, the experience gained from Nippo Lake will be useful in making these future projects successful.

References

Cooke, G.D., E.B. Welch, S.A. Peterson and Stanley A. Nichols. 2005. Restoration and management of lakes and reservoirs. Taylor and Francis. 591 pages.

Dawah, A, Soliman A, Abomohra, A, Battah, M, Anees, D. 2015. Influence of alum on cyanobacterial blooms and water quality of earthen fish ponds. Environmental science and pollution research. 22:21, 2-13.

Gensemer, R.W. and R.C. Playle. 1999. The bioavailability and toxicity of aluminum in aquatic environments. Critical reviews in environmental science and technology, 29:4, 315-450.

Holz, J.C. and K.D. Hoagland. 1999. Effects of phosphorus reduction on water quality: Comparison of alum-treated and untreated portions of a hypereutrophic lake. Lake and Reservoir Management 12(1):70-82.

Huser, BJ. 2012. Variability in phosphorus binding by aluminum in alum treated lakes explained by lake morphology and aluminum dose. Water Research 46:4697–4704.

Huser, B.J., S. Egemose, H. Harper, M. Hupfer, H. Jensen, K.M. Pilgrim, K. Reitzel, E. Rydin and M. Futter. 2016. Longevity and effectiveness of aluminum addition to reduce sediment phosphorus release and restore lake water quality. Water Research 97:122-132.

NHDES. 2005. A Study of the effectiveness, longevity, and ecological impacts of hypolimnetic aluminum injection in Kezar Lake, North Sutton, New Hampshire.

Wagner, K.J. 2004. The practical guide to lake management in Massachusetts. Prepared for the Massachusetts Executive Office of Environmental Affairs. 160 pages.

Wagner, K.J., D. Meringolo, D.F. Mitchell, E. Moran and S. Smith. 2017. Aluminum treatments to control internal phosphorus loading in lakes on Cape Cod, Massachusetts. Lake and Reservoir Management 33:171-186.

Welch, E.B. and G.D. Cooke. 1999. Effectiveness and longevity of phosphorus inactivation with alum. Journal of Lake and Reservoir Management 15:5-27.