Methods of Liming to Accelerate the Reversal of Acidic Deposition Effects in Calcium-Depleted Adirondack Watersheds

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Final Report

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Abstract

Acidic deposition levels in the Adirondack region of New York State at the close of 2020 declined to values estimated for the early 20th century before the emergence of large-scale industrialization. However, chemical recovery of surface waters has lagged as a result of soil calcium (Ca) depletion, and long-term monitoring has not provided a clear indication that soil Ca availability has begun to increase. Recent computer modeling of Adirondack streams suggests that recovery goals may not be met in Ca-depleted watersheds for decades even with aggressive reductions in acidic deposition. The delayed response of soils and surface waters to reduced deposition has prompted consideration of adding Ca in the form of lime to increase the rate at which the effects of acidic deposition are being reversed through decreased pollutant emissions alone. In this regard, an experiment was conducted from 2013–2019 to evaluate the utility of liming to accelerate recovery in Adirondack stream watersheds that drain directly into Honnedaga Lake, near Forestport, NY. Results of this research were combined with relevant research results from prior liming studies to develop a methodology for the use of liming to accelerate reversal of acidic deposition effects tailored for the Adirondack region of New York State. This document presents (1) a rationale for liming in the context of the current acidic deposition regime, (2) an assessment of benefits and risks to be considered with various application methods, (3) a suggested method for selecting watersheds where liming would be most beneficial, and (4) an explanation of the primary factors that need to be addressed to successfully implement watershed liming.

Keywords

calcium depletion, acidic deposition, recovery from acidification, watershed liming, stream liming, aluminum toxicity

Acknowledgments

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Acronyms and Abbreviations

Al	aluminum
Al_i	inorganic aluminum
ANC	acid-neutralizing capacity
Ca	calcium
$CaCO_3$	lime in the form of calcium carbonate
Ca^{2+}	dissolved calcium
$cmol_c\ kg^{-1}$	centimole positive charge per kilogram
DOC	dissolved organic carbon
GIS	geographic information system
GPS	global positioning system
ha	hectare
Hg	mercury
K	potassium
kg	kilogram
MeHg	methylmercury
Mg	magnesium
Na	sodium
NADP	National Atmospheric Deposition Program
pH	$logarithm_{10}$ of hydrogen concentration in units of moles per liter, multiplied by -1
$\mu eq\ L^{-1}$	microequivalents per liter
$\mu mol\ L^{-1}$	micromoles per liter

Executive Summary

By the close of 2020, acidic deposition levels in the Adirondack region of New York State had declined to values estimated for the early 20th century before the emergence of large-scale industrialization. However, chemical recovery of surface waters has lagged as a result of soil calcium (Ca) depletion, and long-term monitoring has indicated only minor increases in soil Ca availability. Recent computer modeling of Adirondack streams suggests that recovery goals may not be met in Ca-depleted watersheds for decades even with aggressive reductions in acidic deposition.

The delayed response of soils and surface waters to reduced deposition has prompted consideration of adding Ca in the form of lime to increase the rate at which the effects of acidic deposition are being reversed through decreased pollutant emissions alone. In this regard, an experiment was conducted from 2013–2019 to evaluate the utility of liming to accelerate recovery in Adirondack stream watersheds that drain directly into Honnedaga Lake, near Forestport, NY. Results of this research have been combined with relevant research results from prior liming studies to develop a methodology for the use of liming to accelerate reversal of acidic deposition effects tailored for the Adirondack region of New York State.

Findings indicate that liming should be considered on the basis of individual watershed characteristics. Positive responses to liming are most likely if the watershed has been depleted of soil Ca, has surface waters with toxic aluminum (Al_i) concentrations, and contains little or no wetland areas. The dosage of added Ca should be sufficient to substantially reduce the toxicity of surface waters without causing large changes in soil pH. Addition of lime should be done in the fall immediately after leaf-drop and all well-drained areas of the watershed should be treated to maximize benefits to both the terrestrial and aquatic ecosystems. Results of this study thus far have evaluated only the first five years following treatment and therefore do not provide information on the longevity of the treatment.

1 Introduction

The harmful effects of acidic deposition became widely recognized in the United States and elsewhere by the 1970s through growing reports of fish extinctions (Schofield, 1976). However, modeling of historic acidic deposition rates (Shao et al., 2020), reconstructions of past and current watershed mass balances (Likens et al., 1996), plot studies (Siccama et al., 1982), and an analysis of tree ring chemistry patterns (Shortle et al., 2017) all suggest that the depletion of soil calcium (Ca), a critical factor in the mobilization of toxic aluminum (Al) was likely occurring by the 1960s, and possibly earlier based on European soil data (Lawrence et al., 2005). As more effects were identified, concern grew that extensive damage would occur before acidic deposition levels could be sufficiently reduced through legislation and technological advances (Driscoll et al., 1996).

To mitigate acidic deposition effects, early studies focused largely on the direct application of lime to surface waters to neutralize acid inputs. Direct liming of lakes in the Adirondack region of New York State caused pronounced and abrupt changes in acid-base chemistry that helped to reduce fish mortality, but was largely ineffective at sustaining acid neutralization under high levels of acidic deposition (Gloss et al., 1988). Lime additions to streams were complicated by the rapid fluctuations in chemistry that occur with changes in flow and the decrease of acid neutralization downstream of where lime was added. As with direct lake liming, stream liming reduced toxicity to fish, but even with expensive automated dosing systems, stable water chemistry was difficult to achieve under high levels of acidic deposition (Zurbuch et al., 1996). On the basis of these results, Woods Lake watershed, located in the western Adirondack region of New York State, was experimentally limed in 1989. In the two to three years following treatment, substantial acid buffering in the lake occurred and chemical changes were found to be more gradual than observed in direct liming additions to surface water (Driscoll et al., 1996). However, watershed liming did not preclude large fluctuations in stream chemistry nor prevent episodic acidification (Newton et al., 1996). The increased acid buffering of lakes and streams achieved in the experiment was attributed largely to the dissolution of lime applied to saturated soils and wetland areas (Driscoll et al., 1996).

The 1990 Clean Air Act Amendments, ongoing declines of acidic deposition, and evidence of chemical recovery in surface waters, all contributed to a waning interest in widespread liming for mitigation. However, some stream (McClurg et al., 2007) and lake liming efforts in the United States continued with a shift in goals from mitigation of acidification to restoration of fish populations (Adirondack Almanac Archive, 2014). This shift was evident in the Adirondack region, where the number of lakes with acid neutralizing capacity (ANC) less than 0.0 microequivalents per liter (μeq L^{-1}) decreased from 15.5% in 1991–1994 to 8.3% in 2006–2007 (Waller et al., 2012). However, similar to surface-water recovery in other regions, ANC increases were limited by decreases in dissolved Ca (Ca^{2+}) concentrations (Lawrence et al., 2016a).

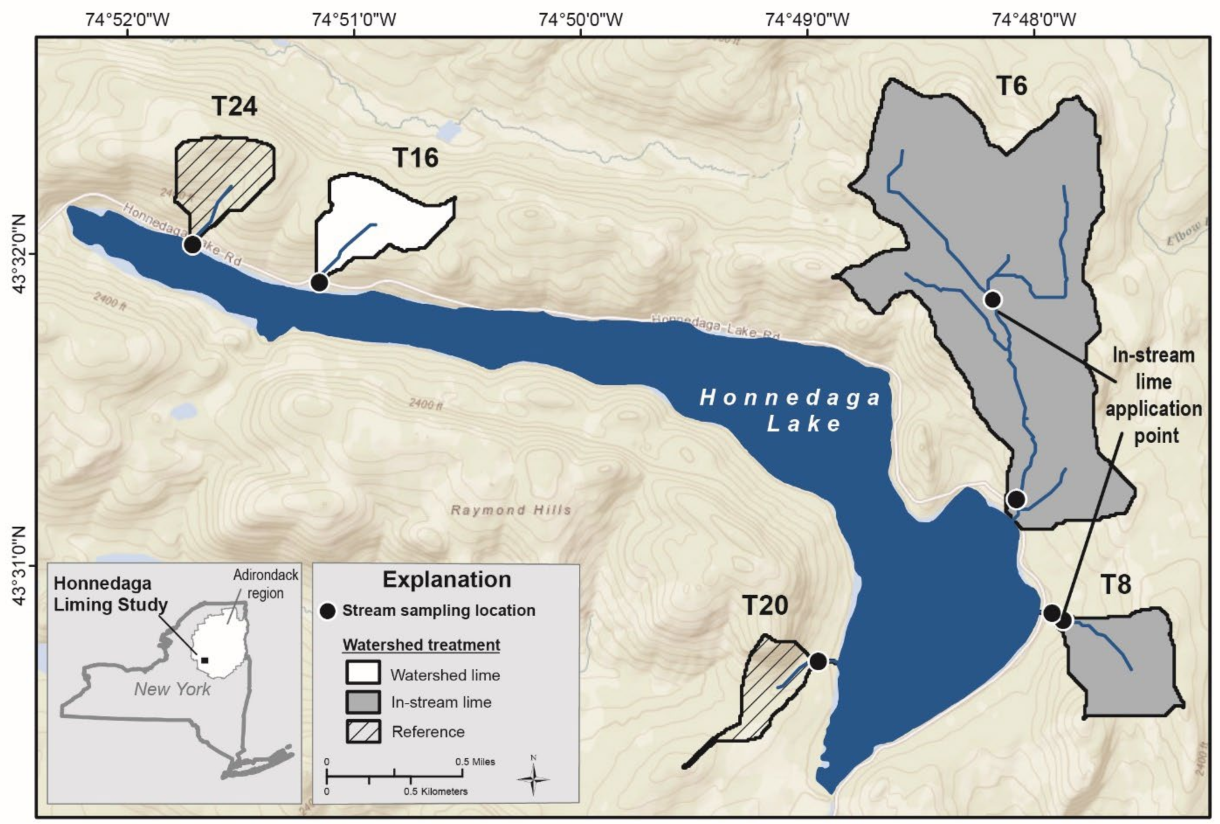
Most recently, acidic deposition levels in the Adirondack region have declined to values estimated for the early 20th century before large-scale industrialization occurred (Shao et al., 2020). However, soil monitoring at multiple sites throughout the northeastern United States and eastern Canada, including the Adirondack region, provided no indications that soil Ca availability had begun to increase by 2014 (Lawrence et al., 2015a). Furthermore, recent biogeochemical modeling of Adirondack streams in severely Ca-depleted watersheds suggested that recovery targets may not be met with additional acidic deposition reductions for decades, and in some cases may never be achievable (Shao et al., 2020). The delayed response to decreasing acidic deposition results from (1) the slow process of weathering that is necessary to release Ca from rocks, and (2) acid-forming compounds that accumulated in the organic matter of forest soils during the era of high acidic deposition levels are now being released gradually through natural decomposition (Lawrence et al., 2020b).

The hindering effect of soil Ca depletion on chemical recovery from acidification led to a recent Adirondack experiment to determine the effectiveness of adding Ca in the form of lime to increase the rate at which acidic deposition effects were being reversed by decreased pollutant emissions alone. This experiment was conducted from 2013–2019 in Adirondack watersheds that drain directly into Honnedaga Lake near Forestport, NY and is referred to hereafter as the Honnedaga Liming Study (Figure 1). Soils in the experimental watersheds are shallow and rocky (Figure 2) with Ca availability that ranged from extremely low to moderate. Lime was applied aerially (Figure 3) to the entire area of one watershed (T16; Figure 1), which had Ca-depleted soils and a chronically acidified stream. A nearby watershed (T24; Figure 1) with similar soils and stream chemistry served as an untreated reference. The forests of these watersheds were comprised largely of beech infected with beech bark disease, and red maple, a forest type common in the region (Lawrence et al., 2018a).

In the Honnedaga Liming Study, lime was also directly added multiple times to a stream with relatively high levels of organic acidity (T6; Figure 1), and to a stream more prone to episodic acidification (T8; Figure 1). Stream liming was included in this experiment to determine if this treatment would be useful to augment watershed liming in helping to reduce episodic acidification during high flows by having lime on the channel bottom prior to flow increases. A watershed with a moderately buffered stream (T20; Figure 1) served as a reference for the direct stream liming, as described in Josephson et al. (2019). Included in the assessment of liming as a method for accelerating recovery from acidification was a study of liming effects on mercury (Hg) transport from soil to streams and Hg cycling within stream ecosystems (Millard et al., 2020). The results of that liming experiment are summarized in this document with details provided elsewhere (George et al., 2018; Homan et al., 2016; Josephson et al., 2019; Lawrence et al., 2021b; Millard et al., 2018; Millard et al., 2020). The objective of this report was to combine results of the Honnedaga Liming Study with relevant scientific literature to identify the benefits, limitations, and risks of liming for the purpose of accelerating reversal of acidic deposition effects on Adirondack ecosystems. Steps for implementing a liming program tailored to the Adirondack region are included. Method details and additional results are available elsewhere (George et al., 2018; Josephson et al., 2019; Lawrence et al., 2021b).

Figure 1. Locations of Study Watersheds in the Honnedaga Liming Study

Circles indicate the primary stream sampling locations, with the exceptions of the lime application points indicated in watersheds T6 and T8.



Base from ESRI World Topographic Map; NAD 1983 UTM Zone18N
Stream network from USGS StreamStats 2017

Figure 2. Evidence of the Shallow, Rocky Soils of Research Watersheds in the Honnedaga Liming Study

Photo by G. Lawrence, 2010.

Figure 3. Helicopter Liming of Watershed T16 in the Honnedaga Liming Study

Photo by D. Josephson, 2013.

2 Benefits of Liming under Low Levels of Acidic Deposition

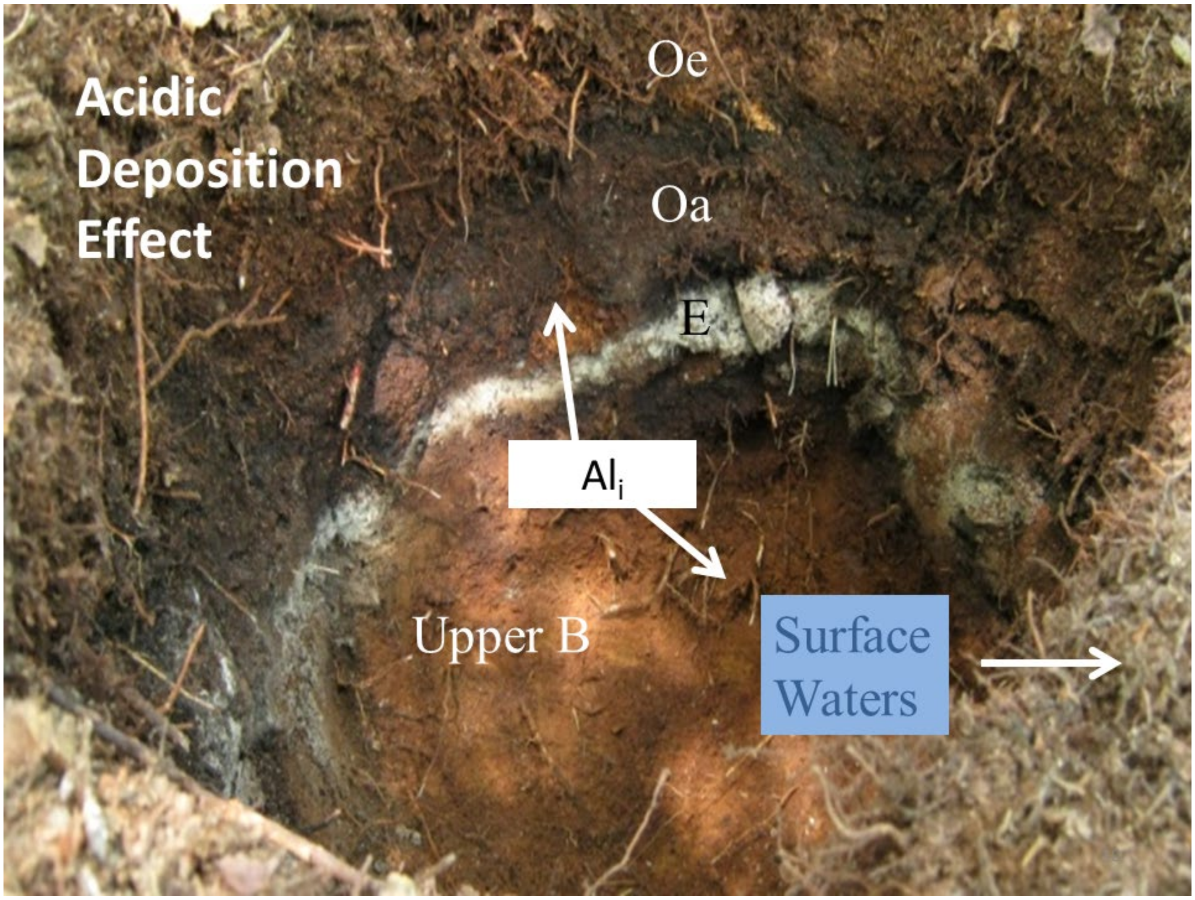
2.1 Soil Benefits

The primary effects of acidic deposition on Adirondack soils are the depletion of soil Ca and mobilization of inorganic forms of aluminum (Al_i) that are toxic to terrestrial and aquatic biota. These soil changes, which have occurred largely within the rooting zone of the upper soil profile (Figure 4), have altered the composition and structure of Adirondack forests by impairing the health and regeneration of sugar maple (*Acer saccharum* Marsh) and several other Adirondack tree species (Lawrence et al., 2018a). Increasing Ca availability through watershed liming can provide a relatively rapid and effective method to reduce or prevent the mobilization of Al_i in soils. Mobilization of Al_i in the soil occurs at a threshold determined by (1) Ca availability and (2) concentrations of strong acids in soil solution that have been derived from acidic deposition (Lawrence et al., 2007). Increasing Ca availability to a level above this threshold by adding lime will prevent the harmful effects of Al mobilization, and increase the biotic utilization of Ca, which is a key nutrient for many terrestrial plants and animals (Beier et al., 2012). Increased Ca availability from addition of the Ca-rich mineral wollastonite was found to strengthen the resistance of red spruce from a region wide winter injury event in a New Hampshire watershed (Hawley et al., 2006). Increasing Ca availability may also increase plant diversity of the Adirondack forest understory (Zarfos et al., 2019) and potentially promote diversity of soil macrofauna (Beier et al., 2012). The importance of small scale spatial variations in the biogeochemical landscape was identified in a study of acidic deposition effects in the Appalachian Trail corridor (Lawrence et al., 2015b).

Figure 4. A Typical Adirondack Soil Profile

Shows the organic Oe horizon (just below the layer of undecomposed leaves on the soil surface), the organic Oa horizon underlying the Oe, the mineral leached E horizon (light color) beneath the Oa, and the upper B horizon beneath the E. Acidic deposition mobilizes Al in harmful inorganic forms (indicated as Al_i) in the upper B horizon, that can be transported upward and downward within the soil profile as well as from soil into surface waters.

Photo by G. Lawrence.



Soil chemistry was measured in the treatment and reference watersheds just prior to lime application on October 1–4, 2013. A dose of 4,516 kilograms (kg) of lime ($CaCO_3$) per ha (hectare; approximately 2 tons per acre) was applied by helicopter over the full 30-ha treated watershed. This dose was approximately double that used in the Woods Lake watershed liming in 1989, when acidic deposition levels were approximately double those in 2013 (http://nadp.slh.wisc.edu/ntn/; based on NADP site NY20, accessed December 1, 2020). These measurements showed that soil chemistry in the reference and treated watersheds were statistically indistinguishable before liming and that soil chemistry in the reference watershed did not change throughout the 2013–2018 study period, except for a small decrease in pH in one of the soil organic layers (Oe horizon, Table 1).

Liming increased exchangeable Ca concentrations in the organic Oe horizon (Table 1). This form of Ca is available to roots and can be readily transported into stream water. Liming also increased soil pH and base saturation (the sum of exchangeable Ca, magnesium (Mg), sodium (Na), and potassium (K) concentrations expressed as a percentage of total exchangeable cation concentrations) and resulted in a substantial decrease in exchangeable Al in the Oe horizon. In the Oa horizon, large increases were also observed in the limed watershed for exchangeable Ca and base saturation and a small increase was observed for pH. Liming effects extended into the upper B horizon where exchangeable Ca concentrations increased by more than a factor of three and base saturation more than doubled.

Table 1. Concentrations of Exchangeable Ca and Al

Centimole positive charge per kg [$\text{cmol}_c\ \text{kg}^{-1}$]), measurements of Base sat. (base saturation as percent), and pH of Oe, Oa and upper B horizon samples from reference and treated watersheds in 2013 (pretreatment) and 2018 (five years after treatment).

All values represent the mean of 15 samples collected in each watershed in the respective sampling year; sig. represents significance levels for differences between sampling years at individual watersheds abbreviated as a: $P < 0.01$; b: $0.01 < P < 0.05$; and a dash: $P > 0.10$.

	Reference			**Treated**		
	2013	**2018**	**sig.**	**2013**	**2018**	**sig.**
	------------- Oe ---------------					
Ca	14.2	12.4	-	11.4	87.6	a
Al	2.5	2.0	-	2.1	0.14	a
Base sat.	65.6	63.2	-	61.5	99.0	a
pH	3.34	3.17	b	3.07	5.25	a
	------------- Oa ---------------					
Ca	3.8	3.9	-	3.9	15	a
Al	11.5	11.3	-	7	6.4	-
Base sat.	17.1	21.2	-	24.5	57.2	a
pH	3.08	2.91	-	2.96	3.23	b
	------------- Upper B --------------					
Ca	0.3	0.28	-	0.25	0.74	a
Al	6.2	5.8	-	5.1	5.7	-
Base sat.	5.8	6.8	-	5.4	11.6	a
pH	3.54	3.47	-	3.51	3.57	-

Within five years, the lime addition had a positive effect on Ca availability that extended down into the upper B horizon. The input of lime neutralized soil acidity in addition to increasing exchangeable Ca concentrations, which resulted in large decreases in soil acidity that were primarily restricted to the Oe horizon. Nevertheless, even with the 2-pH unit increase in the Oe horizon, the soil pH remained mildly acidic with a value of 5.25 in 2018. The decrease in exchangeable Al in the Oe horizon, although substantial, was similar to or less than Al decreases in the Oe horizon measured in a recent study of other untreated Adirondack watersheds (Lawrence et al., 2021b). However, no change in Oe horizon Al was observed in the Honnedaga reference watershed over the study period, and the decrease in Al seen in the Oe horizon of the limed watershed occurred in about one-third the time of that observed for the Adirondack watersheds in the Lawrence et al. (2021b) study.

The minimal changes in exchangeable Al and pH in the Oa and upper B horizons indicated that the liming did not cause a strong acid-neutralization response below the Oe horizon within five years, even though the treatment did increase Ca availability down into the upper B horizon. The increase in exchangeable Ca concentrations in the upper B horizon is of particular significance because this change was likely to have reduced the mobilization of potentially toxic Al, which occurs in this horizon unless Ca availability is sufficient to neutralize strong acids derived from acidic deposition (Lawrence et al., 2007). The downward transport and storage of Ca in the B horizon represented a reversal of the Ca depletion that occurred when acidic deposition caused high rates of cation leaching that depleted available Ca in the B horizon. This recent accumulation of available Ca in the mineral soil was possible because of the low rates of Ca leaching that have resulted from the large decreases in acidic deposition during the five post-treatment years.

The increase in Ca availability in the upper soil profile is likely to relieve ecosystem Ca limitation to some degree. Terrestrial Ca limitation in Ca-depleted soils has been suggested by forest liming experiments that showed liming markedly increased diameter growth for sugar maple and yellow birch (Halman et al., 2015; Long et al., 2011; Ouimet et al., 2017), two of the most abundant tree species in the Adirondack region (Widman et al., 2012). Studies relating sugar maple condition to soil Ca availability also show that low soil-Ca availability is associated with reduced canopy vigor, poor regeneration, poor foliar Ca nutrition, and lowered stress resistance (Bailey et al., 2004; Hallett et al., 2006; Lawrence et al., 2018a).

2.2 Surface Water Benefits

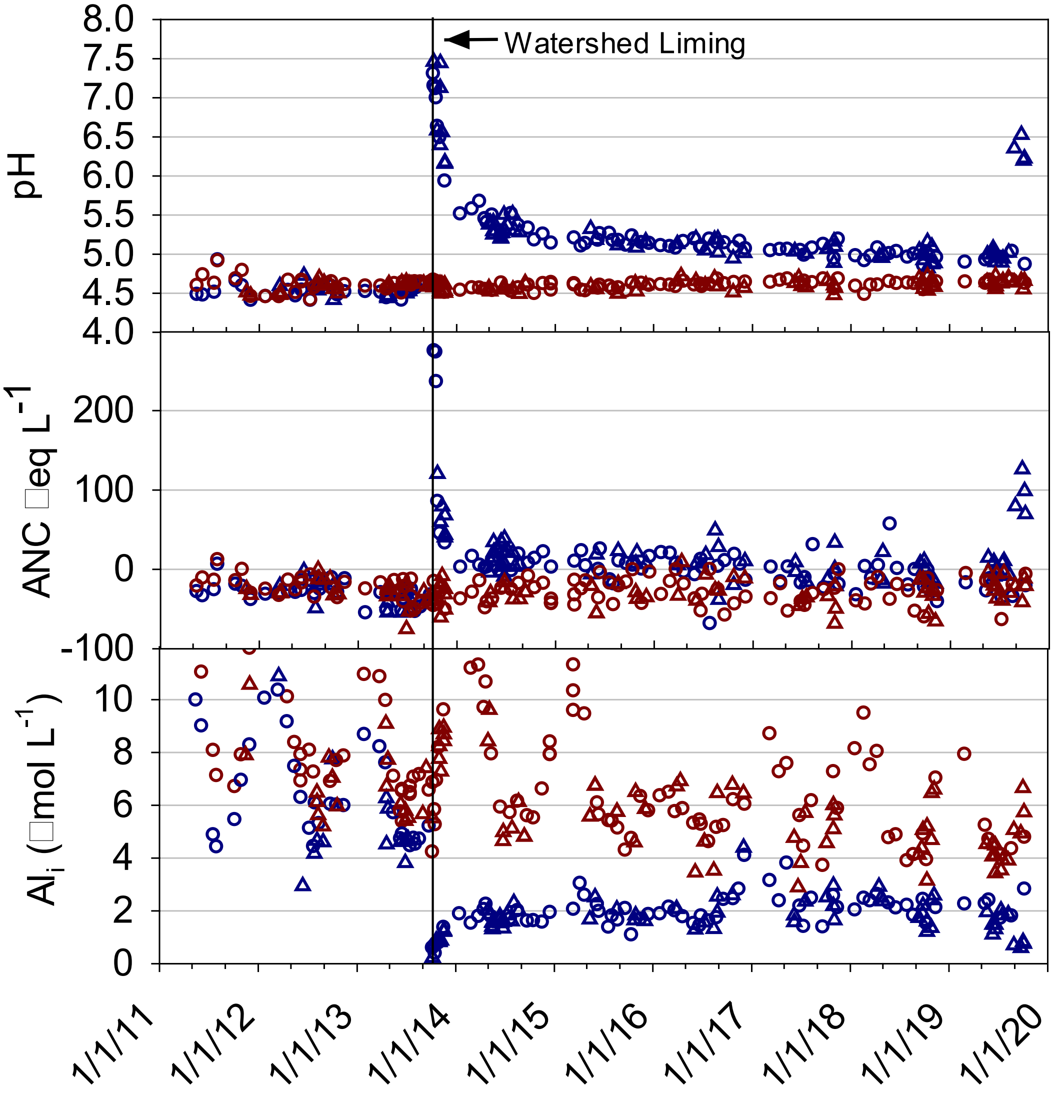
The depletion of soil Ca in Adirondack soils has meant that Al_i can continue to be mobilized in the “soil and transported into surface waters despite the current low levels of acidic deposition. Concentrations of Al_i harmful to fish (greater than 1.0 micromole per liter [μmol L^{-1}]) remain common in lakes and streams in the Adirondack region (Baldigo et al., 2019a; Baldigo et al., 2020). Low concentrations of Ca^{2+} in surface waters have also reduced production and diversity of zooplankton that play a key role in aquatic food webs (Jeziorski and Smol, 2017; Leach et al., 2019). By increasing soil Ca availability, liming could promote higher leaching rates of Ca in watersheds where tight recycling of Ca by terrestrial vegetation limits transport of Ca into streams and lakes. Improved Ca availability in soil is therefore a likely prerequisite for alleviating Al_i toxicity and nutritional limitation in aquatic ecosystems.

Before liming, the streams in the treatment and reference watersheds were chronically acidic, with pH values that ranged mostly between 4.4 and 4.7 and ANC values that were mostly less than 0.0 μeq L^{-1} (Figure 5). Values of pH in the reference watershed increased slightly in the five post-treatment years but remained less than 4.7. Concentrations of Al_i were greater than 4.0 μmol L^{-1} and therefore lethal to brook trout (Baldigo et al., 2020) in the streams of both watersheds prior to watershed liming, and lethal concentrations continued through the experiment in most samples in the reference watershed, although maximum Al_i concentrations declined over the five year post-treatment period, presumably in response to decreasing acidic deposition levels (Figure 5).

Immediately upon liming, the pH of the stream in the treated watershed spiked then gradually declined to below 5.0 by the fourth year after treatment. However, through the five years of post-treatment, the pH in the stream of the treated watershed remained at least 0.2 pH units above that measured in the stream of the reference watershed (Figure 5). Values of ANC in the stream of the treated watershed also gradually declined after an initial spike but remained above an ANC of 0.0 μeq L^{-1} for most samples throughout the study period. In the stream of the reference watershed, ANC remained less than 0.0 μeq L^{-1} in all samples and averaged -29 μeq L^{-1} during the experiment. Liming caused a precipitous decrease in Al_i concentrations in the stream of the treated watershed from an approximate range of 4 to 12 μmol L^{-1} to an approximate range of 1 to 3 μmol L^{-1}. Although Al_i concentrations remained harmful throughout the five years after treatment, the probability that resident brook trout would die during a typical 10-day exposure was reduced from 100% to 20% (Baldigo et al., 2020), and should permit small populations of mature brook trout to exist in some parts of the stream in the treated watershed (Baldigo et al., 2019a; Baldigo et al., 2019b). Concentrations of Al_i would likely need to decrease further to enable all life stages of brook trout to occupy the stream of the treated watershed throughout the year.

Figure 5. Chemical Measurements of Stream Water in the Limed and Reference Watersheds

Blue symbols indicate stream values in the treated watershed; red symbols indicate stream values in the reference watershed. Circles represent samples collected monthly; triangles represent samples collected with an automatic sampler during rapid increases in flow. ANC is acid-neutralizing capacity; Al_i is inorganic aluminum.

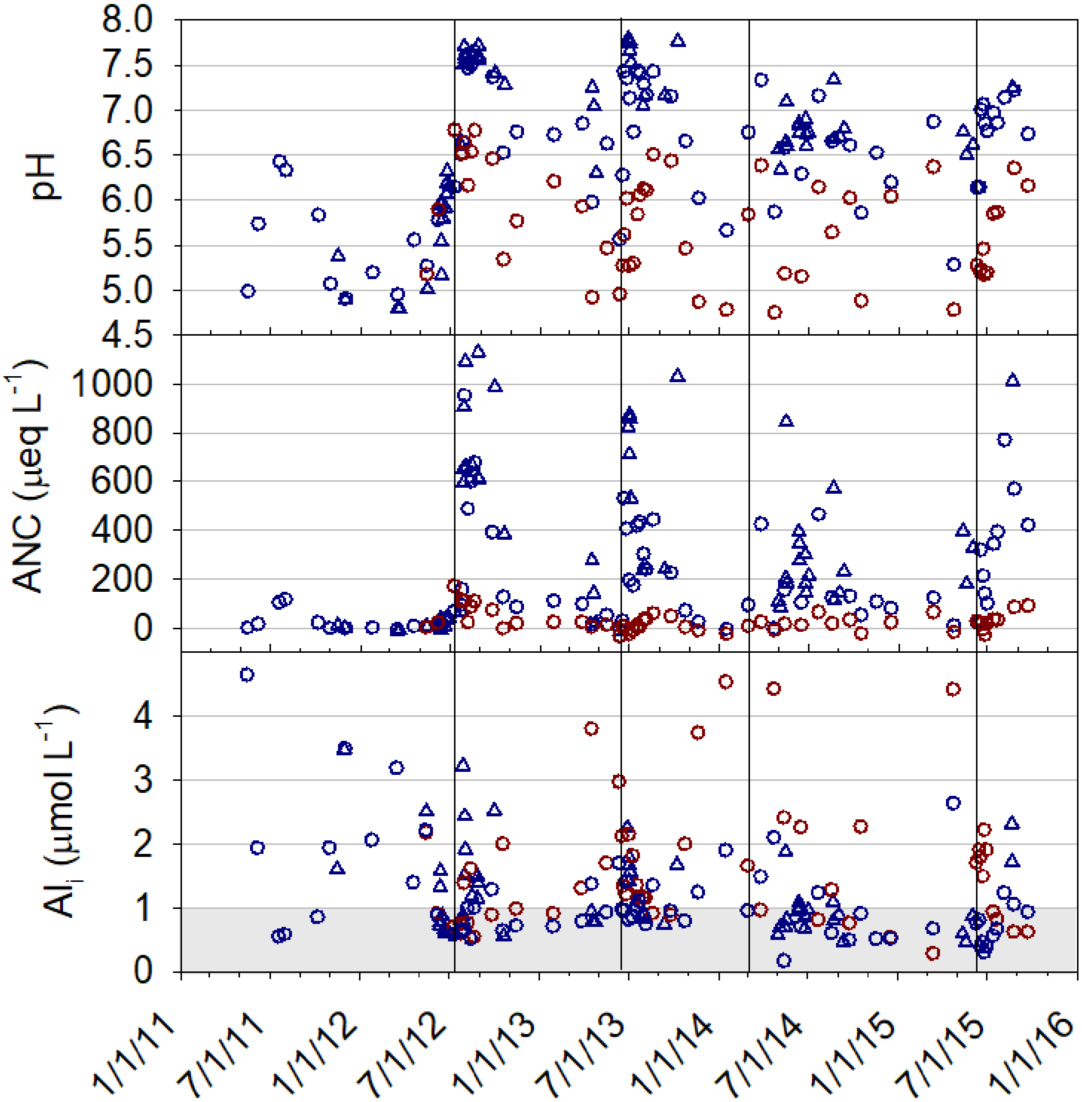


Direst stream liming was accomplished with four treatments of lime added to T8 and T16 at doses of 4,500 kg and 10,000 kg, respectively, between 2012 and 2015. The pelletized lime was poured directly into the streams at single locations as described in Josephson et al. (2019). The response to the first addition to T8 on July 12, 2012, resulted in rapid and pronounced ANC and pH increases relative to measurements upstream of the lime addition (Figure 6). These improvements in water quality were sustained throughout the summer; however, by fall, pH and ANC concentrations began to approach values measured prior to the lime addition. By the spring snowmelt in 2013, pH and ANC levels below the liming were similar to values measured upstream from the lime addition. This pattern was repeated following the second July liming. Following the third addition just prior to spring snowmelt in 2014, pH and ANC concentrations remained higher than values upstream of the lime addition but did not reach the peaks observed after the first two treatments. Concentrations of Al_i downstream of the lime addition generally remained below values measured upstream of the lime addition, and most samples were below 1.0 μmol L^{-1}, a critical threshold for brook trout survival (Josephson et al., 2019). The changes in pH, ANC, and Al_i concentrations following the fourth treatment on June 16, 2015 were comparable to the responses observed during the three prior treatments. The yearly direct lime additions were effective at ameliorating Al_i concentrations but created large fluctuations in water chemistry. The repeated pattern of reacidification between treatments suggested that water chemistry would approach or exceed unhealthy Al_i concentrations if liming was not repeated at least every other year. Results also suggested that direct stream lime additions just prior to snowmelt were likely to have a greater positive effect on water chemistry than summer liming.

Improved water chemistry corresponded with higher densities of that year's young brook trout after direct stream liming began, which suggested that liming resulted in chemical conditions that were adequate for naturally reproducing brook trout populations. Once Al_i concentrations were sufficiently decreased in the episodic stream, suitable habitat for reproduction was provided by ground water inputs and adequate water temperatures (Josephson et al., 2019). However, neither liming technique succeeded in improving the condition of stream insect communities. Changes in physical habitat and chemical instability from the in-stream liming and acidic episodes that were reduced in severity, but not eliminated in either liming method, limited possible benefits of improved water chemistry (George et al., 2018).

Figure 6. Chemical Measurements of Stream Water in the Episodically Acidic Stream

The stream received direct additions of lime on the four dates indicated by the vertical lines in the chart. Red circles represent samples collected upstream of the lime additions; blue symbols represent samples collected approximately 150 meters (m) downstream of the lime additions. Circles represent monthly samples; triangles represent samples collected with an automatic sampler during rapid increases in flow. ANC is acid-neutralizing capacity; Al_i is inorganic aluminum.



3 Limitations and Risks of Liming under Low Levels of Acidic Deposition

3.1 Lake Liming

Application of lime directly onto lake surfaces has been the most common method used for mitigating effects of acidic deposition, but the benefits of this approach were determined to be relatively brief due to the generally short hydrologic residence time of Adirondack lakes that limited acid neutralization by the added lime (Driscoll et al., 1989). Furthermore, the ongoing recovery from reduced levels of acidic deposition appears to be stronger thus far for lakes than for streams (Lawrence et al., 2016a). This may be due to the internal nutrient dynamics of lakes that are associated with primary productivity and to the greater hydraulic residence time of lakes relative to streams (Gerson et al., 2016; Kelly et al., 1987).

Direct liming of lakes under the current acidic deposition regime therefore poses the risk of destabilizing aquatic ecosystems as they adapt to the gradual shifts in water chemistry driven by acidic deposition decreases. These factors suggest that direct liming of lakes under the current conditions is not warranted.

3.2 Stream Liming

The repeated treatments required to maintain the benefits of direct stream liming caused fluctuating chemistry under the current low-acidic deposition levels, as shown in Figure 6. The treatment had a positive effect on the number of current young brook trout in the episodically acidic stream, but other factors including groundwater discharge into the streambed were also necessary to achieve benefits from liming (Josephson et al., 2019). Fish species other than brook trout have been less responsive to stream liming (Lawrence et al. 2016a), and stream insects generally do not show positive responses (Lawrence et al. 2016a), including in the Honnedaga Liming Study (George et al., 2018). Direct stream liming in a watershed with soils that exhibit mobilization of Al_i also poses the hazard of precipitating Al_i within the stream channel as discharging soil waters mix with limed stream water. As Al_i hydrolyzes and precipitates, its toxicity increases until it is fully removed from solution (Poleo et al., 1994). In streams that continue to show minimal recovery, the benefits of direct stream liming can be counterbalanced by the disruption of the stream ecosystem.

3.2.1 Liming Hydrologic Source Areas

Application of lime to hydrologic source areas, those areas within watersheds that produce surface flow during precipitation events, was viewed as a method to remediate acidified surface waters with less fluctuation in chemistry because the acidity is treated before reaching surface waters. However, in the Adirondack region, hydrologic source areas often include organic-rich riparian wetlands with high base-neutralizing capacity and vegetation adapted to naturally acidic conditions . Liming hydrologic source areas also requires large doses of lime and is likely to harm native vegetation . Furthermore, the varying discharge and recharge of hydrologic source areas driven by seasonal rainfall and snowmelt causes variations in the effectiveness of the lime in reducing the acidity of adjacent streams and lakes . Liming of hydrologic source areas is unlikely to have a sustained effect on the chemistry of adjacent surface waters and poses a high risk to wetland plant communities.

3.2.2 Liming Effects on Soil Macroinvertebrates

Large chemical changes in the forest floor (Oe and Oa horizons) resulting from liming may create conditions that are either unfavorable or simply not advantageous for some existing resident animals, but might provide opportunities for other resident species as well as attract new species capable of exploiting the altered chemical environment. This was observed in a plot liming experiment conducted in the western Adirondack region that resulted in decreased breakdown of leaf litter in limed plots that may have been related to a large increase in snail abundance coupled with a large decrease in millipede abundance (McCay et al., 2013).

Changes in soil pH from liming could also drastically alter organic carbon and nutrient dynamics of forest soils by enabling the invasion of earthworms (Bohlen et al., 2004). A combined field and laboratory study run in conjunction with the Honnedaga Liming Study found no earthworms in Adirondack soils with pH less than 3.6 (Homan et al., 2016). Mean pH of soil samples collected in the untreated watershed, and before Ca addition in the limed watershed was less than 3.6 in Oe, Oa and upper B horizons, and no earthworms were observed in any soil sampling. However, in a microcosm experiment that increased Honnedaga soil pH from 3.1 to 3.7, survival of Lumbricus terrestris, an earthworm of European origin commonly used for fishing, increased from less than 20% to nearly 80%. The risk of altering litter processing in the forest floor from changes in the soil fauna community should be included in considerations of watershed liming.

3.3 Potential for Mercury Release from Liming

Much of the Hg in forest soils is bound to organic matter, which when mobilized, facilitates the transport of Hg into surface waters. Because dissolved organic carbon (DOC) concentrations are increasing in surface waters recovering from acidic deposition (Driscoll et al., 2016; Lawrence and Roy, 2021), concern has arisen that recovery from acidic deposition may increase Hg concentrations in aquatic food webs. Likewise, increases in DOC concentrations from liming could increase transport of Hg from soils to surface waters, which could enhance the bioavailability of Hg. Of particular concern is methylmercury (MeHg) concentrations. Methylmercury is produced microbially from the processing of ionic Hg by sulfate or iron reducing bacteria, generally in wetlands or sediments. Methylmercury is the form that poses the greatest health risk to aquatic organisms and humans due to its ability to bioconcentrate, bioaccumulate, and biomagnify in food chains (Munthe et al., 2007).

In the Honnedaga Liming Study, direct stream liming resulted in increases in DOC and total Hg in stream water in both the high DOC stream and the stream prone to episodic acidification, but the effect lasted for only a few days after each treatment (Millard et al., 2020). Watershed liming, however, caused a multi-year increase in concentrations and loads of total Hg in stream water, although concentrations of MeHg in water and macroinvertebrates did not increase during the three-year monitoring period after liming (Millard et al., 2020). The well drained soils that are dominant throughout most of the treated watershed limit the opportunity for Hg methylation. However, during the study period, increased MeHg in stream water of the treated watershed followed an extended summer drought. Wet-dry cycles have been shown to enhance production of MeHg (Chen et al., 2012). Future changes in this type of climatic event may affect production and transport of MeHg from wetland soils to streams.

Because wetlands have the capacity to produce high DOC concentrations and provide conditions suitable for Hg methylation, the transport of MeHg from wetlands to surface waters can enhance the bioavailability of Hg (Driscoll et al., 2007). Application of lime to wetlands would likely increase the solubility of organic matter, thereby increasing the transport of MeHg into streams and lakes. Liming of wetlands should therefore be avoided for this reason, as well as to prevent damage to plant communities.

4 How to Implement Liming to Accelerate Reversal of Acidic Deposition Effects

4.1 Liming as a Management Tool

The remarkable progress in reducing acidic deposition levels has shifted the primary management goal from ecosystem protection to restoration of conditions more similar to those that existed prior to the impacts. Because the rate of chemical recovery of lakes and streams is now primarily limited by the rate at which soil Ca availability can increase in terrestrial ecosystems, liming of entire watersheds offers a method to accelerate the overall recovery process by utilizing the natural biogeochemical linkage between soils and surface waters. The desired outcome of substantially lowered emissions continues to be the reversal of acidic deposition effects through natural recovery. However, this goal is not only limited by long-term effects on soils but is also confounded by the sparse information available on environmental conditions prior to the advent of acidic deposition. Furthermore, the growing influence of climate change on forest and aquatic ecosystems suggests that a full return to prior conditions may not be sustainable and perhaps not possible. These factors complicate the establishment of recovery targets. Therefore, reversal of known harmful effects may be a more achievable goal than restoration or recovery to pre-acid rain conditions. Nevertheless, Adirondack ecosystems that have been affected by acidic deposition remain highly valued, so historical information should be used where possible to serve as a guide to remediate adverse ecological effects of acidic deposition (Higgs et al., 2014). In specific circumstances, liming might help to achieve this goal, with the recognition that outcomes are likely to differ from pre-industrial conditions.

With the current low inputs of acidic deposition, liming could be used to neutralize soil acidity well beyond conditions prior to acidic deposition, but this would create a large disruption to the ecosystem and therefore should be avoided. Liming may instead be used to help restore damaged ecosystems to a biogeochemical state that more closely reflects the natural state of Adirondack soils, which are acidic but lack harmful forms of Al. Complete neutralization of natural soil acidity would require large doses of lime and would likely be highly destabilizing to the forest ecosystem. The goal of liming should be to increase Ca availability, prevent Al mobilization and improve nutrient balances that will promote the stability of interconnected terrestrial and aquatic ecosystems. The best way to achieve this goal is through application of a moderate dose of lime to the entirety of a well-defined watershed. At the current stage of recovery, benefits of direct lime applications to lakes, streams, and hydrologic source areas are limited and pose risks that could alter ecosystems in unintended ways or impede natural recovery.

4.2 Criteria for Identifying Adirondack Watersheds where Liming would be Beneficial

Due to the geochemical variations of the Adirondack landscape and watershed sensitivity to acidic deposition, watershed recovery status varies widely across the region. The results of repeated surveys of headwater streams (Lawrence and Roy, 2021) indicate that many Adirondack watersheds now show few if any effects of acidic deposition, and others are demonstrating a strong capacity for reversing previous adverse effects under the current low-deposition levels. However, watersheds that are showing minimal capacity for reversing effects are also common, particularly in the western Adirondack region (Shao et al., 2020). For this reason, application of liming to accelerate reversal of acidic deposition effects should be considered on the basis of individual watershed characteristics. Positive responses to liming are most likely if the watershed meets the following criteria:

1. Base saturation of the upper B horizon is less than 15%.
2. Stream water Al_i concentrations exceed 1.0 µmol L^{-1} during summer or 3.0 µmol L^{-1} during spring high-flow.
3. Soils are generally well drained throughout the watershed— riparian and other areas with poorly drained soils should comprise a small fraction of the watershed area.
4. The watershed possesses high-ecological value that has been significantly degraded by acidic deposition.
5. Accelerating the reversal of chemical changes in soils and surface waters caused by acidic deposition would potentially enhance the ecological value of the watershed.
6. The size and location of the watershed is practical for liming without excessive costs.

Criteria 1 and 2 relate to the current chemical condition of the watershed based on soil chemistry and stream chemistry. Approximately 25% of the Adirondack Park is estimated to have soils with base saturation levels less than 15% (Lawrence et al., 2018b). In 2017–2018 approximately 10% of the streams within the western Adirondack region exceeded the summer and snowmelt Al_i concentrations suggested in criterion 2 (Lawrence et al., 2021a). Approximately 10% of the streams in the eastern and central Adirondacks also exceed the Al_i concentration during snowmelt, but less than 5% exceed the summer Al_i concentration (Lawrence et al., 2021a). Soil samples should be collected and analyzed for base saturation from the upper B horizon at a minimum of five locations throughout a candidate watershed to address spatial variability that is typically high in forest soils. Information on the collection of soil samples is available in Lawrence et al. (2016b). Methods for chemical analysis of soils are available in Lawrence et al. (2020a). The collection of stream samples should be done in early to mid-April to obtain samples during spring snowmelt. Summer samples can be collected in July–August.

At least four stream samples should be collected during each season to be chemically analyzed for Al_i following the method described in Lawrence et al. (2021a). Measurements such as pH and ANC of surface waters are also used extensively for assessing acidic deposition effects, but these measurements are less informative than Al_i for assessing Ca depletion of the upper soil profile, which is now the primary limitation on acidification recovery. Soils often have naturally acidic pH values regardless of acidic deposition levels, and although ANC relates well to Al_i in stream water, the relationship can be appreciably altered by concentrations of DOC, which vary widely in Adirondack waters and are increasing due to recovery(Lawrence et al., 2021a).

If either criterion 1 or 2 is met, the watershed is responding weakly to decreases in acidic deposition. A base saturation value of less than 15% in the upper B horizon is an indication that Ca availability is low and that mobilization of harmful Al forms is likely to be occurring (Lawrence et al., 2018a). Each of these factors can result in deleterious effects to terrestrial ecosystems such as impaired growth and regeneration of trees with a high Ca demand, including sugar maple. Exceedance of the Al_i concentrations in criterion 2 indicates that aquatic ecosystems are being harmed. Mobilization of Al_i in the soil does not necessarily imply that streams will also have harmful concentrations of Al_i. This stream response depends on geomorphological aspects of the watershed. For this reason, data on both soils and surface waters should be collected to evaluate candidate watersheds for liming.

Addressing criterion 3 requires that both imagery and ground surveys be used to assess landscape variability within a watershed. Lidar imagery, if available, could be used to help identify poorly drained areas that should not be limed to avoid causing damage to wetland plant communities and increases in Hg mobilization. Adirondack wetland mapping is also available (Roy et al., 1997), but ground surveys should be done to ensure that wetlands are minimal within the drainage to be limed. Ground surveys are also useful in identifying whether the watershed possesses unique features, plant communities, or other forms of biota. This information is needed to evaluate the ecological value or uniqueness of ecosystems to determine if they meet criteria 4 and 5. Specifically, the presence of a unique species or community that has been harmed and is recovering slowly as a result of soil Ca depletion and Al_i mobilization is a potentially important factor in this decision-making process. Several studies have shown that various indicators of biological recovery can lag well behind chemical recovery (e.g. Baldigo et al., 2020; Keller et al., 2019). The likelihood that the desired biological recovery will be accelerated by increasing Ca availability and eliminating or reducing Al_i mobilization should also be assessed before considering lime additions.

Lastly, the practicality of liming the watershed must be taken into account (criterion 6). Accessibility is necessary to allow ground transportation to deliver the lime to a landing near the watershed to enable short helicopter flights. The area of the watershed will determine how many flights will be needed to dispense the lime. The airtime of the helicopter may be the greatest expense associated with watershed liming.

The 30-ha watershed limed in the Honnedaga Liming Study provides an excellent example of a watershed that meets these six criteria. The soils were severely Ca depleted, with a mean base saturation of 5.6% in the upper B horizon. The Al_i concentrations in the stream of the reference watershed in 2017–2018 exceeded 4 µmol L^{-1} in most samples throughout the year. Because the stream discharges directly into the lake, potential habitat for spawning could be provided for the genetically unique brook trout inhabiting the lake (Josephson et al., 2014). The lake was considered chemically unsuitable for fish survival in the 1980s due to acidic deposition effects, but a reproducing population of brook trout survived in a handful of small tributaries that were sufficiently acid buffered. Chemical recovery of the lake has enabled fish to repopulate, but density remains low and reproduction may be limited because many of the tributaries remain chemically unsuitable for spawning. Improving the extent of spawning habitat through watershed liming could help to support this unique population of brook trout in the lake watershed. The watershed treatment was also likely to have spurred terrestrial recovery by improving soil chemistry. Helicopter liming was feasible for this watershed because lime could be trucked to a landing area within about 2 miles of the watershed.

4.3 Steps to Accomplish Watershed Liming

Once a watershed has been selected based on the six criteria above, these steps should be followed to implement the lime addition:

1. Locate a source of pelletized, high-Ca lime (approximately 30% Ca by mass). Because Ca is generally in greater biological demand than Mg in Adirondack soils, a high fraction of Mg is unnecessary. The supplier can provide information on the chemical composition, which can vary considerably depending on the quarry. The lime should be pelletized for aerial application.
2. The dose should not exceed 1,400 kg Ca ha^{-1}, which was the dose used in the Honnedaga Liming Study. This dose provided substantially improved Ca availability in soils and a large reduction in the toxicity of stream water. A larger dose would have resulted in a greater reduction in stream toxicity, but also increased the pH of the upper soil beyond the desired level. This dose balanced the benefit of increased Ca availability while minimizing unwanted changes in the natural forest ecosystem and the cost of application. It should be kept in mind that Adirondack watersheds that have experienced severe Ca depletion were naturally low-Ca ecosystems before the onset of acidic deposition.

3. A geographic information system (GIS) shape file of the watershed area to be limed needs to be provided to the helicopter operator. With global positioning system (GPS) technology, helicopter liming can be done with high precision. The area that received lime can be verified by placing pans to catch the lime throughout the watershed, and just inside and outside the watershed boundary.
4. Addition of lime should be done in the fall, preferably after most of the leaves have fallen from deciduous trees. However, weather conditions in the Adirondack region beyond the first or second week of October could limit when the helicopter can fly. The liming of the Honnedaga watershed required the helicopter to be on site four consecutive days (October 1–4).

5 Concluding Remarks

The rationale and procedures for liming to accelerate reversal of acidic deposition effects presented in this document are based largely on the Honnedaga Liming Study, which was specifically designed for this purpose. However, information from liming experiments conducted for mitigation purposes was also utilized. Results of the Honnedaga Liming Study demonstrated that watershed application of lime can be of value in aiding recovery from acidic deposition effects, but this experiment did not address the effectiveness of the treatment beyond the five-year study period. Additional post treatment monitoring would be needed to determine how long and at what rate the initial treatment will continue to accelerate recovery beyond what is occurring under ambient conditions. This information would help to provide a more complete understanding of the long-term value of watershed liming in supporting Adirondack uses such as forestry and recreational fishing, as well as helping to maintain the ecological integrity of the region.

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