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A review of the effectiveness and feasibility of alternate liming techniques to mitigate for acid rain effects in Nova Scotia

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ABSTRACT

This paper is a summary of some techniques that may be applicable to liming acid rivers in Nova Scotia. It emphasises the preservation of Atlantic salmon habitat. Acid rain has eliminated salmon entirely from some rivers in Nova Scotia and reduced salmon production in many others. Controls on atmospheric emissions have produced less improvement than expected in the pH of Nova Scotia's rivers. As an interim measure, alkaline substances (lime) can be added to acidified surface waters to raise their pH. Liming is costly and is justified only where equivalent benefits to fish can be produced. A pH of 5.3 is necessary to protect salmon in Nova Scotia during the incubation period through to first feeding. Salmon parr and smolt require a minimum pH of 5.0. Liming adds calcium to natural waters and this may confer additional benefits on fish besides raising the pH of their habitat. Concentrations of toxic metals do not increase in limed waters. Limestone is usually the best neutralising substance owing to its relatively low cost and safety. Other substances are available but these are more expensive and may be dangerous to personnel handling them and to the fish.

Once liming has been carried out, the higher pH can only be maintained by frequent or continual liming. This paper describes several methods of applying lime to lakes and rivers. The factors that affect efficiency of dissolution are discussed. Methods are described for calculating the required lime dose for each method of liming. The relative costs, efficiency and reliability of different liming techniques are compared.

RÉSUMÉ

Ce document présente un sommaire de certaines techniques pouvant servir au chaulage des rivières acides en Nouvelle-Écosse, et met l'accent sur la conservation de l'habitat du saumon atlantique. Les pluies acides ont exterminé le saumon de certaines rivières de la Nouvelle-Écosse et en ont réduit la production dans de nombreuses autres. La hausse du pH des rivières en question consécutive à la lutte contre les émissions atmosphériques a été moindre que prévu. Comme mesure provisoire, il est possible de relever le pH des eaux de surface acides en leur ajoutant des substances alcalines (chaux). Le chaulage est dispendieux et seulement justifié si l'on peut en tirer des avantages équivalents pour les poissons. Le saumon de la Nouvelle-Écosse est protégé de la période d'incubation des œufs jusqu'à la première alimentation lorsque le pH atteint 5,3. Les tacons et les smolts exigent un pH d'au moins 5,0. Le chaulage ajoute du calcium aux eaux naturelles, ce qui peut avoir, pour les poissons, des bienfaits additionnels à la hausse du pH de leur habitat. Les concentrations de métaux toxiques n'augmentent pas dans les eaux calcaires. Grâce à son coût relativement faible et à son innocuité, le calcaire constitue habituellement la meilleure substance de neutralisation. D'autres substances peuvent être utilisées, mais elles sont plus coûteuses et peuvent être nocives pour le personnel qui les manipule, ainsi que pour les poissons.

Une fois le chaulage effectué, la seule manière de maintenir le pH à un niveau élevé consiste à ajouter souvent ou continuellement de la chaux. Plusieurs méthodes d'application de la chaux aux lacs et rivières sont décrites dans ce document. On y discute des facteurs qui influent sur la vitesse à laquelle la chaux se dissout, en plus d'y décrire comment calculer la dose de chaux nécessaire pour chacune des méthodes de chaulage, et d'y comparer les coûts, le rendement et la fiabilité relatives de ces différentes techniques

INTRODUCTION

This paper is a summary of the experience gained to date in the mitigation of the effects of acid rain on fish populations by adding lime to their natural habitats. It was prepared in response to concerns regarding the impact of acid rain on the Atlantic salmon in southwestern Nova Scotia. Some of the information summarised here was gained in Nova Scotia but considerably more extensive experiments have been conducted in other areas and especially in the eastern United States and in Scandinavia.

Acid rain has had major adverse effects on freshwater fish populations in North America and Western Europe. As the increased acidity of rain in those continents is caused by man-made pollution of the atmosphere, the most effective remedy is to reduce the amounts of pollutants at their sources. Acidic pollutants may be transported great distances and across international boundaries. The problem of acid rain can not be solved locally by purely local action; international co-operation is necessary. Furthermore, the control of acid emissions into the atmosphere is costly. The reduction of acid emissions into the atmosphere is proceeding somewhat slowly while some fish populations remain in danger. Liming of natural waters may provide an interim way of preserving fish populations until a more permanent solution has been found to the problem of acid rain.

Acidification affects all juvenile stages of Atlantic salmon *Salmo salar* from incubation to seaward migration of smolts. It has eliminated salmon entirely from some rivers in Nova Scotia and reduced salmon production in many others. Watt (1986) estimated that 3,500,000 m^2 of salmon habitat are totally lost ($pH<4.7$) and that acidification has reduced production in a further 3,800,000 m^2 of salmon habitat. He estimated the loss in production owing to acidification in Nova Scotia to be 22,500 salmon annually. The main acidifying compound in Nova Scotian salmon rivers is sulphuric acid (H_2SO_4).

Although the emissions of acidifying pollutants have been curtailed in the past decade, the increase in pH of surface waters and decline in toxicity in Nova Scotia has been less than expected (Watt *et al*. 2000). This means that until even more stringent controls have been imposed on emissions, acid rain will continue to reduce salmon production in Nova Scotia.

Freshwater habitat can be protected from acidification by adding alkaline substances (lime) to the water. The lime reacts with acids to produce either neutral products or much weaker acids. The usual neutralising substance is limestone. Limestone consists mostly of calcium, and magnesium carbonate with some impurities. Other compounds are available but these are generally more expensive and sometimes dangerous to use. Methods of mitigation other than liming, fish culture, for example, are not discussed here.

This paper concerns mitigation by liming in salmon habitat only. Mitigation of acidification in Nova Scotia has been carried out mainly to protect salmonid fish and especially Atlantic salmon. However, other species may benefit from liming. The brook trout *Salvelinus fontinalis*, which is the other native salmonid fish in Nova Scotia, survives in waters that are now too acidic to support Atlantic salmon. Measures that mitigate acidification sufficiently to protect salmon will also protect trout.

Liming has been conducted on a very large scale in Sweden, Norway, the United States and, within Canada, Ontario. Considerable experience has been gained in these areas with many methods of liming including some that have not been seriously tested in Nova Scotia. Usually, lime has been spread onto the surface of a lake or else introduced into a stream from stationary "lime dosers" of various patterns. Experiments have also been made with liming of dry land or wetlands. Various liming products have been used but ground limestone or dolomite has been found most satisfactory (Henrikson and Brodin 1995).

The first point to consider in any liming program is the pH that must be attained to produce the desired result, in this case protection of salmon. A second important consideration is the feasibility of the proposed project; some watercourses may be so acidic that it would be prohibitively expensive to raise the pH sufficiently to protect fish during all stages of their life history. The third consideration is whether there are any fish to protect in the waters that are to be limed. A fourth is whether the pH is low enough to justify any mitigation. These obvious points are raised here only because liming of particular watercourses has, on occasion, been promoted passionately but without consideration of these matters.

EFFECTS OF LIMING

Tolerance of Atlantic salmon towards pH and metals

Information from other areas, particularly Sweden and Norway, on the pH tolerances of Atlantic salmon can not be applied uncritically to Nova Scotia. Measurements of the lethal thresholds of Atlantic salmon in Nova Scotia are considerably lower than European measurements. Conspicuous fish-kills are not a feature of high flow events in Nova Scotia as they have been in Scandinavia (Hesthagen 1989). The reason for the discrepancy is that the toxic effects of dissolved aluminum amplify those of acid in European waters. Aluminum concentrations are correlated with concentrations of hydrogen ions (H^+) in European waters and it is hard to separate the effects of the two in the field.

Aluminum toxicity generally is not considered to be a factor in the majority of acidified Nova Scotian rivers. Aluminum concentrations are high in many watercourses in Nova Scotia but organic compounds bind most of the aluminum into non-toxic forms (Lacroix 1989, Lacroix & Townsend, 1987). The pH, which must be attained to protect fish, is lower in Nova Scotia than in some other areas and the task of mitigation is consequently easier.

There may be some exceptions to this rule. The apparent lethal pH threshold for salmon parr in water from Sandy Lake, Nova Scotia was pH 5.7 (Hutcheson and Strong 1982) which is considerably higher than most values recorded in Nova Scotia. Aluminum collected on the gills of these fish (Hutcheson and Odense 1984). The concentration of dissolved organic compounds can be determined from the colour of natural waters. The water colour in Sandy Lake, N.S. was only 5.5 Hazen units (White *et al*. 1984). This value indicates an unusually low concentration of dissolved organic substances for Nova Scotia where the water colour usually exceeds 30 units (Watt *et al*. 1996). If liming is being considered for any waters in Nova Scotia, their colour should be measured so as to determine whether aluminum is likely to be present in toxic form.

The following discussion of pH tolerance limits applies to salmon in Nova Scotia and not to any other area.

Incubation to swim-up

The pH of interstitial water in spawning gravel must be at least 5.0 or higher during the fry stage until swim-up (Lacroix 1985). Lacroix (1989) gives the following equation describing the survival of salmon embryos in waters of different pHs:

$$\%\ \text{Survival} = -356.82 + 86.89\text{pH} \quad \text{-----------------------------------}(1)$$

Solutions to equation 1 suggest that the incipient lethal level is about pH 5.3. and total mortality would occur at pH 4.1.

Low pHs also inhibit hatching enzyme activity below pH 5.5 and hatching mortalities are severe below pH 4.5 (Peterson *et al*. 1980). Salmon fry are especially sensitive to low pH at swim-up. The median lethal pH for swim-up fry is 5.3 (Farmer *et al*. 1980) and the incipient lethal pH is about 5.4 (T. Goff and G. Farmer[1], personal communication).

Parr

The toxic pH threshold for salmon parr is at or slightly below 5.0. The overwintering survival of young parr in the wild was below average where the winter pH fell below 5.0 (Lacroix 1989). The lethal limit for caged parr lay between 4.7 and 4.8 (Lacroix and Townsend 1987). Plasma sodium (Na^+) and chlorine (Cl^-) were still declining in the surviving fish when the experiments were terminated and so longer-term experiments might have yielded a somewhat higher lethal threshold.

Smolts

Exposure to low pHs prevents the development of salt tolerance in Atlantic salmon smolts. Smolts held at pH of 4.6 died when placed in salt water whereas mortality was negligible among smolts held at pH 5.0 or higher when similarly challenged (Farmer *et al*. 1989). Johnston *et al*. (1984) reported that seawater tolerance and osmoregulation in fresh water were impaired in smolts held at pH 4.9 and pH 4.7. Seawater-adapted salmon smolts exposed to acid water lose seawater tolerance (Staurnes *et al*. 1996).

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[1] T. Goff and G. Farmer, Department of Fisheries and Oceans

Table 1. Minimum pH objectives for liming projects to protect Atlantic salmon in Nova Scotia.

Stage	Tolerance
Incubation to hatching	5.3
Swim-up	5.3
Parr	5.0
Smolts	5.0

The effects of calcium on pH tolerance

Liming of acid waters with calcium compounds may confer an additional benefit apart from any effects liming may have on the water's pH. Many neutralising substances are compounds of calcium and so liming of waters will also increase their calcium concentrations. Fish gills are less permeable to Na^+ and Cl^- in the presence of Ca^{++} ions (Hunn 1985) and so the loss of ions is reduced in acid water when the concentration of calcium is high. Calcium enables rainbow trout *Oncorhynchus mykiss* and brown trout *Salmo trutta* to survive longer and withstand lower pHs (Brown 1983, McDonald 1983, McDonald *et al*. 1980, McDonald *et al*. 1983). The threshold for benefits of Calcium lay between 0.5 and 1 mg/L. (Brown 1983). Calcium has been shown to counteract the effects of low pH in acidified Ontario lakes so that the number of fish species surviving at any pH is higher where calcium concentrations are also high (Brown 1982).

Effects of liming on dissolved metals

Metal ions are often toxic to salmon. Limestone may contain considerable amounts of aluminum, iron or other metals. However, the solubility of metals is lower at high pH than in acid waters. Concentrations of dissolved metals usually fall after the pH has been adjusted by liming (Scheider and Brydges 1984). The concentrations of Aluminum, Manganese Copper and Zinc fell significantly in Sandy Lake, Nova Scotia following liming although iron increased unaccountably(White *et al*. 1984). The decrease in dissolved aluminum took place despite the fact that considerable aluminum was introduced into the lake with the limestone. The creation of limestone gravel spawning beds in Nova Scotia had no effect on the aluminum content of the treated water (Mayhew, 1989).

COMPARISON OF DIFFERENT NEUTRALISING SUBSTANCES

A wide variety of compounds are available for liming. They differ in theoretical neutralising capacities, solubility in water, ease and safety of handling and cost (Table 2).

Table 2. Partial list of compounds available for acid rain mitigation.

Neutralising Substance	Chemical formula	Neutralising value per unit weight ($CaCO_3$ = 100%)
Calcium carbonate (limestone)	$CaCO_3$	100%
Dolomitic limestone	$CaCO_3.MgCO_3$	100-138%
Calcium hydroxide (slaked lime)	$Ca(OH)_2$	135%
Sodium hydroxide (caustic soda)	NaOH	136%
Sodium carbonate	Na_2CO_3	94%
Calcium oxide (quicklime)	CaO	179%

Limestone

Limestone is the most commonly used compound. It can be obtained in crushed aggregate or powdered form. It consists predominantly of calcium carbonate and magnesium carbonate with some impurities. Dolomite is limestone that contains a relatively high proportion of magnesium carbonate (Table 3). Limestone is quarried in Nova Scotia but not in the Southern Upland of Nova Scotia where the rivers have been affected by acid rain. Limestone can be handled safely without protective equipment but spreading the powdered form produces considerable dust. Dolomite has slightly higher potential neutralising capacity per unit weight but is less easily dissolved so that it is less effective for liming watercourses.

Table 3. Calcium and magnesium content of calcitic and dolomitic limestone used for lake liming in Nova Scotia.

	Calcitic limestone	Dolomitic limestone
Calcium	36.0 %	17.1 %
Magnesium	1.5 %	11.3 %
Calcium carbonate equivalence	96.1 %	89.2 %

In theory, limestone can raise the pH of acid water to about 8.3 but laboratory and field tests seldom raise the pH much above 7.0 even when excess limestone is added. This limited acid-consuming ability is actually an advantage as there is no danger of creating overly high pHs through overdosing. On the other hand, a portion of the limestone added to watercourses settles uselessly on the bottom and takes little part in the neutralisation of the lake. The high calcium content of limestone may provide additional benefits to fish, beyond those created by improving the pH.

Sodium carbonate

Sodium carbonate has similar chemical properties to limestone but is considerably more reactive. It should be used with caution as it can raise the pH up to 11 or 12. This material may irritate exposed throat and nose skin and irritate or burn the eyes. Personnel handling sodium carbonate should wear protective clothing.

Calcium hydroxide and magnesium hydroxide

Calcium hydroxide or slaked lime is widely used for treatment of acidic mine wastes. It is highly soluble and reactive. Complete dissolution is easy to achieve. Calcium hydroxide can raise the pH above the tolerance limits of fish and so the dose must be carefully regulated and the limed water fully mixed.

Inhalation of calcium or magnesium hydroxide dust irritates the nose, throat and upper respiratory tract. Severe exposure may cause serious injury to the respiratory tract. In contact with the skin and eyes, it can cause severe burns. It can cause permanent injury and blindness. Personnel must wear protective equipment and clothing.

Sodium hydroxide

Sodium hydroxide (caustic soda) is dangerous to handle. It is readily dissolved in water and can raise the pH above the lethal range for fish and concentrated solutions cause severe burns to the eyes and skin. There is a strong exothermic reaction with water.

Sodium hydroxide irritates or burns the skin, nose, throat, and mucous membranes. Particles in the eyes can cause serious burns. Personnel must wear protective equipment and clothing including a long-sleeved shirt, long pants extending over tops of the work boots, gauntlet-type work gloves, eye goggles and face mask.

Calcium oxide and calcium-magnesium oxide

Calcium oxide (quicklime) is dangerous to handle. It undergoes a violent exothermic reaction with water to produce calcium hydroxide. The reaction can generate enough heat to ignite combustible materials. It is easy to dissolve 100% of CaO in water and it can raise the pH above the lethal level for fish.

Calcium oxide irritates or burns the skin, nose, throat, and mucous membranes. Particles in the eyes can cause serious burns. Personnel must wear protective equipment and clothing including a long-sleeved shirt, long pants extending over tops of the work boots, gauntlet-type work gloves, eye goggles and face mask.

Other materials

Other materials can be used to neutralise acidified waters. Certain industrial wastes such as basic slag $CaO\text{-}SiO_2$ can also be used but these should be accepted with caution. They may cause the pH to rise beyond the tolerance limits of fish and besides, may contain metals or other toxic contaminants.

Chemical reactions of neutralisation

The chemistry of acid neutralisation depends on the particular substance used and on the environmental conditions in which the reaction takes place. The following equations describe the dissolution of limestone ($CaCO_3$) in an acidic aqueous solution:

$CaCO_3 + H^+ \rightarrow Ca^{++} + HCO_3^-$ (pH<6) ---(2)
$CaCO_3 + H_2O \rightarrow Ca^{++} + HCO_3^- + OH^-$ (pH >6) -------------------------------------(3)

The pH determines which reaction is the more important. The first reaction predominates at pHs below 6; the second above pH 6. Magnesium behaves similarly to calcium and may be substituted for it in these and the following equations:

The dissolution reaction of sodium carbonate is similar to the high pH reaction of calcium carbonate.

$Na_2CO_3 + H_2O + H^+ \rightarrow 2Na^+ + HCO_3^- + H_2O$ --(4)

The reactions of $Ca(OH)_2$ and $Na_2(OH)_2$ are also similar, the result of both reactions being the production of OH^- ions that react with H^+ to produce water.

$Ca(OH)_2 + 2H^+ \rightarrow Ca^{++} + 2H_2O$ ---(5)
$Na_2(OH)_2 + 2H^+ \rightarrow 2Na^+ + 2H_2O$ ---(6)

CaO reacts with water to produce $Ca(OH)_2$ according to the following equation. The $Ca(OH)_2$ then reacts as shown above.

$CaO + H_2O \rightarrow Ca(OH)_2$ --(7)

When the pH of water is raised, dissolved metals form complexes with OH^- ions that are relatively insoluble. The metal hydroxides tend to precipitate around the lime particles and shield the particles from further reaction.

METHODS OF APPLYING LIME

The discharge of running waters in many Nova Scotian rivers normally varies over a range of two orders of magnitude. The H^+ concentration may vary over one unit. Furthermore the pHs and flows are so related that the lowest pHs tend to occur during the highest flows. Therefore, the rate at which hydrogen ions are carried past a given point on a river will likely vary over a range of several orders of magnitude and the rate at which neutralising substances are added must be similarly adjusted. This requirement complicates the task of treating acid rivers since the rate of treatment must vary continuously to compensate for continuously changing pHs and flows. Effective treatment requires a release of lime proportional to the discharge and acidity of the water that is to be neutralised. Liming of flowing water must be conducted continuously throughout the year to avoid extreme fluctuations of pH. It is essential that the liming equipment be able to function reliably with few breakdowns at all seasons. Lake liming is usually carried out intermittently, usually at annual intervals. With lake liming one has a choice between liming open water or liming in the winter on the ice. If lake liming is undertaken the relative advantages of the different seasons should be weighed.

Lake Liming

In Nova Scotia, lakes are limed in order to protect fish habitat in the rivers downstream from the lakes. The improvement of habitat within the lake is not the primary purpose of the operation. In this respect, lake liming in Nova Scotia differs from similar operations carried out in other areas where improvement of fish habitat in the lakes themselves is the objective. Ideally lakes limed for this purpose should be as far upstream as possible so as to correct the pH in the largest possible amount of downstream habitat and there is little advantage in liming lakes near the estuaries of acidified rivers. Excess alkalinity usually must be added to the lake so as to compensate for dilution downstream towards the estuary.

Most of the neutralising substances in Table 2 are highly soluble and no difficulties will be met with in achieving complete dissolution. The main considerations in their use will be avoidance of over-dosing and safe handling of these dangerous chemicals. Limestone and dolomite are considerably less soluble than the others. The methods used to spread them will determine the proportion dissolved. The dissolution rate for limestone is controlled by the diffusion rate of the reacting ions and by the surface area of the limestone particles. The diffusion boundary layer is thicker around particles that are at rest with respect to the water than around sinking particles. The rate of dissolution for particles sinking through the water column is greater than for particles at rest on the bottom and a considerable portion of the limestone that settles on the lake bottom will be removed permanently from the reaction. Furthermore, limestone particles in the lake sediments become coated with metal hydroxides on the bottom and thereafter contribute little to the neutralisation (Sverdrup 1984). In some environments, the lime will be covered with silt.

Nevertheless evidence of dissolution from the bottom was seen during periods of overturn during the year following liming in Nova Scotian lakes. Significant dissolution from the bottom evidently ceased approximately one year following liming in Nova Scotia experiments (Watt *et al*. 1984, White *et al*. 1984). However, in the Norwegian Lake Stora Hovatn, perceptible dissolution from the bottom continued for 8 years after liming (Rosseland and Hindar 1988). About 40 to 70 percent of limestone will be permanently lost to the bottom in lake liming operations (Table 4).

The reaction with acid takes place at the lime particles' surfaces. Small particles have relatively greater surface areas and are dissolved more completely than larger particles. Furthermore, small particles sink through the water column more slowly and so are exposed to the lake water for a relatively longer period. Therefore, the lime used for lake liming should be the most finely ground available. If lime is spread on lakes as a dry powder, smaller particles adhere to the larger ones. The larger particles sink relatively rapidly and carry the smaller particles with them. Mixing the limestone powder into a slurry before spreading it on the lake can prevent this problem. Dry application can reduce the dissolution efficiency 40-50% relative to slurrying (Dodge *et al*. 1988, Sverdrup and Warfvinge 1988, Warfvinge *et al*. 1984). However, in winter liming operations, forming a slurry might be impractical owing to freezing of the apparatus and in any case the slurry so formed would not enter the lake water in liquid form.

Lime that is spread over the deepest parts of the lake is exposed to the lake water for a longer period as it sinks through the water column and will dissolve more completely than lime spread over shallower water (Warfvinge *et al*. 1984). However, particles that settle near the shore above the wave base may be re-suspended by wave action (Wright 1985).

The rate of limestone dissolution varies directly with temperature. On the other hand the viscosity of water varies inversely with temperature so that particles sink more slowly through cold water. The longer exposure to the water column in cold water may counteract the effects of the slower chemical reaction. Lake liming differs from liming with limestone gravel which is very temperature-dependent. The percent dissolution of lime varies inversely with pH since the rate of reaction depends on the concentration of H^+ ions (Sverdrup (1984).

Table 4: Summary of lake liming operations and percent dissolution of lime in Nova Scotia (data from Watt 1986 and Watt and White 1992).

Lake	Type of limestone	Season	Dose (X acidity)	Pre-liming pH	Percent dissolution
Big	Dolomite	Summer	0.7	4.6	62
Peterson	Dolomite	Summer	2.0	4.4	50
Eastern	Dolomite	Summer	2.7	4.6	47
Sandy	Calcite	Summer	3.3	4.8	59
Scott	Calcite	Winter	8.2	4.5	45
Officers Camp	Calcite	Winter	2.5	4.5	49
Timber	Calcite	Winter	1.2	5.2	32
Coolen	Calcite	Winter	8.2	5.1	34
Square	Calcite	Winter	6.3	5.2	32

Temperature stratification in winter can reduce the effectiveness of lake liming. Acid surface runoff enters the lake at 0°C in the winter. Being less dense than the warmer limed water already in the lake, it fails to mix and this runoff forms a shallow layer of highly acidic water at the lake surface (Hultberg 1977). Ice cover augments this situation by preventing wind-driven mixing. The limed water underlying this surface layer contributes virtually nothing to the lake's discharge and the water that leaves the lake's outlet may be almost unaltered acid rain (Watt *et al*. 1984, White *et al*. 1984). Liming in the winter can overcome this problem. The least acidic water is then at the lake surface and at a temperature near 0°C. It mixes easily with the acidic surface runoff that enters the lake and each new rainfall until the spring break-up brings more lime to the lake.

In winter liming operations, the lime will likely be applied as a dry powder to prevent freezing of the liming apparatus. Adhesion between particles will reduce the percent dissolution. Some of the limestone delivered in winter will contain large frozen lumps that will sink quickly and occasionally jam the lime-delivery apparatus. Although winter liming is less efficient in the use of limestone, it is relatively cheap in other respects and treats the lake surface just before the spring runoff and fry swim-up.

Liming the headwaters of East River, Chester caused a gradual increase in the pH of East River from slightly above pH 5.0 before liming in 1986 to above 6.0 five years later. The increase was detected at all downstream sites but was greater near the headwaters than at the mouth of the river. The mean pH at the mouth of the river rose from slightly above 4.5 before liming to 5.0 by 1991. Coinciding with this improvement in water quality, the numbers of juvenile salmon increases but the increase was slight until three years after liming began (Watt and White 1992). This result was expected since the number of juveniles were not expected to increase until one salmon generation after the first liming and after the juveniles present in 1986 when liming began had returned and spawned.

After lime is added to a lake, the concentration begins to fall as untreated water from the tributaries replaces the limed water. The change in concentration at any time following liming can be approximately calculated from equation 8.

$$C_t = C_o e^{t/T} \quad \text{--}(8)$$

Where: C_t = concentration of lime at time t
C_o = concentration of lime at time 0 immediately after liming.
T = water retention time of the lake.

This equation would be completely valid if limed and unlimed water were perfectly mixed and if no lime were released from the bottom. Temperature stratification causes seasonal deviations from the calculated value by preventing complete mixing. The release of lime from the lake sediments consistently increases lime concentrations above the predicted values. After one water retention period has passed the concentration of lime should be approximately 37% of the initial concentration. After 2 periods it will be about 14%. A limed lake should have a water retention period of several years or else the liming must be repeated at frequent intervals. Liming must be repeated at least annually in most Nova Scotia lakes owing to their short water retention times.

Calculation of lime dose: The initial dose of lime for a lake can be calculated by multiplying its volume by the acidity of its water. Incomplete dissolution can be allowed for by dividing the product by the expected proportion of lime dissolved. Based on the data in Table 3, this correction factor should be about 0.5 or 0.6 for summer liming and 0.35 for winter liming. The dose required to maintain the pH at a suitable level can be estimated by multiplying the annual volume of discharge through the lake's outlet by the lake's average acidity. The product should be divided by the same correction factor as for the initial liming. Any useful liming program in Nova Scotia will be conducted over a fairly long period. Therefore, a site-specific correction factor can be determined experimentally after one or two years' monitoring data have been gathered.

Costs of lake liming: Watt *et al*. (1984) estimated that the average cost of liming lakes by boat in the summer was $72 per tonne in 1984 Canadian dollars. In 1998 dollars, this translates into a cost of $113 per tonne. The present-day costs of liming Sandy Lake without any environmental studies are estimated as follows.

135 tonne lime @ $30/tonne	$ 4,050
Labour 45 person-days	6,750
Equipment, supplies	3,500
Total	$14,300
Total cost per tonne	$ 105

This estimate is close to the $113 given in Watt *et al*. (1984).

This site is easily accessible and is within the Halifax City limits. A similar project in a remote area would be more expensive. This estimate is based on the assumption that major equipment including boats and vehicles are available for the three weeks of the liming operation and do not have to be purchased. If these items must be purchased or rented especially for lake liming, the cost per tonne would be considerably higher. Bagged lime is more expensive than bulk lime. It is assumed here that lime would be delivered in bulk unlike the actual operation when the lime was bagged. White *et al*. (1984) described the apparatus used for summer liming. Watt and White (1992) reported the cost of liming Sandy Lake by boat in the summer at $210/tonne in 1990 dollars, equivalent to $244 in 1998. The higher estimate was made to compare summer lake liming with other methods and includes estimates of all costs.

Watt and White (1992) reported that 330 tonnes were spread on the ice of several lakes in the headwaters of East River, Chester. The cost in 1990 dollars was $100/tonne equivalent to $116/tonne in 1998.

Limestone gravel bars

The rate base needed to neutralise a river varies with the flow of the river. The fluctuations in lime requirements are magnified by the inverse relationship that exists between flow and pH of rivers. Experiments have been conducted in Nova Scotia to de-acidify acidic rivers by spreading limestone gravel on the stream-bed (Watt *et al*. 1984). Gravel limestone in streams was effective at low flows but the effect fell markedly at higher flows. Unfortunately, the pH of acidified rivers also varies inversely with discharge (Watt *et al*. 1984) so that the effect of the limestone gravel on pH is least when it is most needed.

Lacroix (1992) experimented with limestone gravel bars on Fifteen Mile Brook, Nova Scotia, a tributary to the Medway River. He observed an average increase in pH of 0.05-0.1 unit over each bar and a total increase of 0.4 units. Calcium in the water column also increased by 0.38 mg/L. The difference in pH between limed and control and limed areas ranged from 0.01 to 1.31 units. Severe acidic episodes were observed during high flows in limed and unlimed areas alike. However liming reduced the frequency of episodes where salmon were completely eliminated. The difference in pH between limed and unlimed sections of the stream decreased gradually over the three years following liming as the limestone was gradually washed away or coated. Nevertheless, the gravel bars continued to improve pH three years after they were installed.

Salmon spawned in Fifteen Mile Brook before liming but survival of juveniles was negligible. Salmon selected limestone gravel for spawning, possibly on physical criteria alone. High mortalities occurred in these redds in the first year following liming owing to instability of the newly laid gravel. The mean pH of interstitial water in the limestone bars was about neutral (pH 7.1) and much higher than in the controls (pH 5.4). Acidity-related mortalities in the limed areas were greatly reduced during incubation and early fry stages. The density of age 0+ salmon fry was higher in the limed sections than in the controls but few older parr were present even after good fry survival, possibly owing to emigration of these older fish.

Salmon parr over-winter under stones 20 cm across or greater (Rimmer *et al*. 1984). This choice of habitat might bring them into contact with the interstitial water of high pH. It might be worth while to investigate whether over-winter parr survival could be enhanced by including stones of this size on the surface of limestone gravel bars so as to attract overwintering parr to limed substrate.

Gravel bars tend to be washed away during high flows. The bars placed in straight, shallow riffles and runs are more stable than those in winding, deep and uneven stream bottoms (Mayhew 1989). Limestone gravel beds eventually become inactivated owing to the formation of metal hydroxides on the particles' surfaces (Zurbuch 1984). The inactivation of limestone particles can be prevented to some extent by placing the limestone in fast-running turbulent water that agitates the gravel and cleans off the surface deposits. However, gravel placed in such conditions may be swept away during floods.

Pearson and McDonnell (1975) give design criteria for limestone barriers to neutralise acid mine waste. Their methods require the establishment of a head of water behind the barrier so as to create enough current through the barrier to keep it clear of silt that would otherwise coat the limestone and halt the reaction. A disadvantage of their technique is that it might obstruct fish migrations.

Calculating the required dose: Equation 9 describes the relationship between the pH increase, amount of gravel spread, river discharge and temperature. This equation can be used to calculate the amount of limestone gravel needed to produce a given pH increase for any site provided the likely range of flows and pHs have been determined.

$$\Delta \text{pH} = 0.237(\log_e \text{DOSE}) + 0.008(t) - 0.809 \text{ ------------------------------------}(10)$$

where:
ΔpH = change in pH
DOSE = rate of lime application/water discharge (tonne/m^3/sec)
t = temperature (^{o}C)

Equation (10) indicates that the neutralisation reaction is temperature-dependent. For any rate of discharge, a given increase in pH would require about twice as much lime at 0^o C as at 20^o C. A temperature of 0^o C can be assumed because the most severe combinations of pH and discharge generally take place in the late autumn and winter. The increase in pH caused by a gravel bed will vary inversely with water discharge.

Enormous quantities of limestone gravel would be required to prevent severe acidic episodes during periods of high flow and low temperature. The pH of the Gold River, N.S. at a discharge of 50 m^3/sec would be about pH 4.8 (Watt *et al*. 1984) which is within the lethal range for salmon. At 0^o C and that flow and pH, 3400 tonnes of gravel would be needed to raise the pH to the minimum value of 5.0. Doubling the amount of limestone spread could purchase an additional 0.2 pH units increase to pH 5.2.

Costs of limestone gravel bars: Watt *et al*. (1984) estimated the cost of distributing limestone gravel at $32/tonne. In 1998 dollars, that would be $41/tonne. The cost of raising the pH of the Gold River to pH 5.0 under the above conditions of temperature and flow would be $139,000. On the other hand, the effects of liming with limestone gravel might last a number of years whereas the effects of lake liming last one or two years for most lakes.

Revolving drums

A rotary drum system was developed in West Virginia to overcome the problem of coating and inactivation of the limestone particles (Zurbach 1984). Each drum functions as an overshot water wheel and the crushed limestone aggregate is placed into a perforated metal cylinder that turns about the hub of the water wheel. This system does not need an external source of electrical power but there must be a hydraulic head equal to at least the diameter of the drum (1.3 m. in the drums that were tested). The drums have to be regularly inspected and refilled. One 4-drum installation was operated for 15 years. Freezing temperatures did not interfere with operation of the drums except during the lowest flows and drums resumed operation within a few hours of an increase in flow.

An installation can contain several drums that share a single sluice. Variations in river flow can be compensated for by arranging the supply of water so that only one drum would be in operation at low flows and the number of operating drums would increase progressively as the flow increased. As flow increases or decreases, water will reach and power more or fewer drums. The drums will rotate more rapidly as the flow increases. The average person-day requirement from a 4-drum installation was 51 man-days per year for operation and maintenance. Lime can be stockpiled at the site so that transportation over rough roads can be carried out at the most convenient time of year.

Water downstream from drum installations is turbid and deposits of fine lime collect on the river bottom downstream from these devices. This is not entirely undesirable as the lime deposits may protect the river from acid shock if the drums should malfunction. On an occasion when a drum ceased operation, the pH did not fall for 2 days afterwards, probably because of the ground limestone deposits accumulated on the streambed downstream from the installation (Zurbuch 1963).

The production of limestone fines varies directly with the hydraulic power transmitted to a drum. The rate of production of fines is a function of the hardness of the stone used. A drum's turning rate varies as its load is depleted, and is lowest when the drum is half full. The rate of limestone fine production varies similarly. The rate of production in a recently filled drum will decline at first until it is half full and increase again as the drum empties further (Pearson and McDonnell 1978). Zurbuch (1963) reported that over 98 % of fines produced by rotating drums could pass a 74 μ m sieve.

The drums are more efficient when only lightly loaded. When a drum was replenished at 0.5 hr intervals and the load held to about 100 kg. the rate of limestone consumption could be increased by a factor of six. Continuous human attendance would be needed to keep a drum operating at maximum efficiency by this means. A self-feeding drum has been developed that continuously replenishes the limestone as it is consumed and allows the load to be kept light without constant human attendance. A self-feeding drum was also 6 times more efficient than a hand-fed drum.

Calculating the required capacity for rotating drums: The necessary capacity of a drum installation can be determined from the flood frequency distribution and acidity of the river that is to be limed. When a maximum acceptable frequency has been chosen for acidic episodes, the discharge corresponding to the frequency is determined from the flood frequency distribution. The rate of discharge is then multiplied by the acidity likely to occur at that discharge. Acidity increases with flow but the relationship is not linear. The river is unlikely to be more acidic than the rain and so the value for the acidity of rain could be used in this calculation. Using the acidity of winter rain is justified because the most severe acid episodes usually occur when runoff flows directly over frozen ground into watercourses without significant dilution by groundwater. The product of flow times acidity must be divided by the fraction of the limestone dissolved to allow for incomplete dissolution. This fraction would have to be determined experimentally for Nova Scotian lime.

Costs of revolving drums: Zurbuch (1963, 1984) gives estimates for the costs of this device. The cost in 1963 of two four-drum installations was $31,240 (U.S. dollars) which is $3,905 per drum. These were hand-fed drums. The cost in 1998 Canadian dollars would be $23,788 per drum and this would amount to $3,350 per year amortised over 10 years at 6.75% interest. Each drum required 13 person-days for maintenance, inspection and replenishing the lime. The average lime consumption of each drum was 23 tonnes.

The annual costs of operating a single drum are estimated as follows:

23 tonne lime @ $30/tonne	$ 690
Labour 13 person-days	1,950
Capital amortised over 10 years	3,350
Total	$5,990
Total cost per tonne	$ 260

The factor that limited production of lime in the stream was the availability of water power. A drum operating at full capacity all year might consume 80 tonne/year. At this rate of consumption, $2,400 worth of lime would be consumed. The annual cost of labour and capital would remain constant. The annual cost would rise to $7,750 but the cost per tonne would fall to $96.

A self-feeding drum is equal in capacity to six ordinary hand-fed drums. Furthermore, a self-feeding drum would require only one-sixth the flow required to turn six hand-fed drums. The cost of a self-feeding drum in 1998 dollars would be $110,500. Amortised over 10 years that would be $15,550/year.

The annual costs of operating a single drum are estimated as follows:

138 tonne lime @ $30/tonne	$4,140
Labour 13 person-days	1,950
Capital amortised over 10 years	15,550
Total	$21,640
Total cost per tonne	$ 156

A self-feeding drum operating at full capacity for a year would consume 480 tonne. The annual operating costs would rise to $31,900 but the cost per tonne would fall to $66. It should be remembered that a self-feeding drum would require less water to operate and so would operate nearer full capacity than an equivalent installation of six hand-fed drums.

Lime dosers (silos)

Lime dosers are stationary automatic devices that dispense powdered lime into running water from a vertical silo or other container. An automatic mechanism feeds the powder into the river at a controlled rate. The rate at which lime is dispensed must be continually adjusted to keep pace with the changing amount of acid passing it. This is accomplished by placing a sensor in the river connected to an electronic controller on the lime feeding mechanism. Systems regulated directly by pH are unreliable because unattended pH electrodes are fragile and subject to drift. They also freeze easily. Controllers linked to water level sensors are more dependable. Flow and pH are correlated and so water level is a good surrogate for both acidity and flow. The lime may be dispensed directly to a river as a dry powder or fed into a slurrying apparatus where it is mixed thoroughly with water before being added to the river. Bjerle *et al*. (1983) estimate that dissolution efficiency is 40% for dry powders and 100% for slurries.

Some designs need a source of electrical power to operate the lime dispensing mechanism and controller. This is a disadvantage in Nova Scotia where many potential sites have no nearby electrical power. Water powered dosers have been built that overcome this limitation(Olem 1991). Lime dosers must be accessible by road both to permit delivery and installation of the equipment and to deliver lime.

Calculation of required capacity of lime dosers: High flows will sometimes exceed the capacity of a lime doser to dispense lime. The required capacity of a lime doser can be determined from the flood frequency distribution and acidity of the river that is to be limed. The flood frequency distribution and the acidity of the river determine the required feed rate of a lime doser. When the acceptable frequency has been chosen for acidic episodes, the discharge corresponding to the frequency is determined from the flood frequency graph. The rate of discharge is then multiplied by the acidity likely to be associated with that discharge. Acidity increases with flow but the relationship is not linear. The river is unlikely to be more acidic than the rain and so value for the acidity of rain could be used in this calculation. The product of these two numbers must be divided by the fraction of the limestone dissolved to allow for incomplete dissolution.

Costs of Lime dosers: Watt *et al*. (1984) estimated the cost of liming with silos or lime dosers at $120/tonne in 1983 dollars. The equivalent cost in 1998 would be $188/tonne. The state of Maryland has released the operating costs for two lime dosers through the internet (http://www.mde.state.md.us/wma/minebur/doserdat1.html). The capital costs in 1993 were $92,000 and $93,000 U.S. respectively. These costs are equivalent to $127,000 and $128,000 Cdn in 1998. The costs of liming are $100/tonne and $155/tonne. The costs per tonne differ because one doser dispenses 362 tonnes annually while the other dispenses 770.

If similar assumptions regarding the cost of lime and labour are made as for the rotating drums above, the annual cost of applying 1000 tonne of lime with a lime doser in Nova Scotia may be roughly estimated as follows:

Capital costs	$127,500
Period of depreciation	10 years
Interest	6.75%
Annual costs	
Capital amortised over 10 years	17,942
1000 tonne limestone @ $30	30,000
Labour 14 person-days @$150/day	2,100
Total	35,042
Total cost/tonne	$50

Diversion wells

The diversion well is a simple liming device in which limestone chips are kept in motion by the upwardly flowing water (Bjerle *et al*. 1983). The agitation causes collisions between particles that abrade them and prevent their surfaces from becoming coated with metal hydroxides. Dissolution of the limestone is not complete. When the particles have been worn down to a certain size, the current will carry them out of the well and these particles presumably take no further significant part in the reaction. Widening the top of the well reduces losses by this process. The current is slowed in the wide section below the fluidisation velocity and the particles then drop back into the well. Losses can be reduced by this means to about 10 % of lime.

Diversion wells require a minimum head of water of about 1.5 meters to supply water with enough pressure to keep the limestone in the well in a fluidised state. They are suitable for sites where the flow variations are not excessive. A liming well in Piggeboda, Sweden can dissolve 5g/m^3 limestone or 150 tonne/year but the authors state that with modifications to the well, this capacity could be doubled.

As with lime dosers, high flows will sometimes exceed the capacity of a diversion well to dispense lime and fish will be killed on these occasions. The procedure for estimating the necessary capacity of a diversion well is similar to the method for lime dosers and can be determined from the flood frequency distribution and acidity of the river that is to be treated. The calculations are identical but as there is no controller on a diversion well, the dose can not be so finely adjusted to the flow.

Costs of Diversion wells: Bjerle *et al*. (1983) estimate the capital costs for a liming well with a capacity of 2,900 tonne/year to be 150,000 Kr. In 1983. This is equivalent to $36,300 Cdn 1998.

Capital costs	$36,000
Period of depreciation	10 years
Interest	6.75%

Annual costs

Capital amortised over 10 years	$ 5,110
1000 tonne limestone @ $30	30,000
Labour 14 person-days @$150/day	2,100
Total	$37,210
Total cost/tonne	$37

Table 5 compares the costs of the different methods of liming. The method of placing bars of limestone gravel in rivers is not included because the efficiency of dissolution with this method is not comparable with those of the other methods. The different methods vary considerably in relative costs but the most cost-effective method for any given site will likely depend on the individual characteristics of the site. For example, silos are very cost-effective where large amounts of lime are required but may prove less so on smaller streams. A large part of the cost of silos is the initial capital cost. This cost may not fall proportionately as the lime requirement is reduced.

Table 5. Summary of annual costs and cost/tonne of liming by different methods

Method	Tonnes of lime	Total cost of lime	Capital and equipment	Labour	Cost per tonne
Lake liming	$ 135	$ 4,050	$ 3,500	$ 6,750	$ 105
Revolving drum (manually fed)	$ 23	$ 690	$ 3,350	$ 1,950	$ 260
Revolving drum (self-feeding)	$ 138	$ 4,140	$ 15,550	$ 1,950	$ 156
Lime doser (Silo)	$ 1,000	$ 30,000	$ 17,942	$ 2,100	$ 50

CONCLUSIONS

Liming natural waters may be an effective method of protecting fish populations that are threatened by acid rain. It raises the pH of waters where it is undertaken and may produce additional benefits by increasing calcium concentrations. Calcium increases the ability of fish to withstand low pHs. The method is expensive and is justified only where corresponding benefits can reasonably be expected. In Nova Scotia, mitigation of acid rain by liming has been undertaken mainly to protect the Atlantic salmon. A mean annual pH of at least 5.0 is necessary to protect this species. Liming where this pH can not be attained will not protect salmon. Limestone is the preferred liming substance owing to its relatively low cost and safety.

Different methods of applying lime to natural watercourses have been found effective but their relative costs vary considerably. Summer lake liming has the advantage that lime is more soluble in warm water but has the drawback that in the following winter when the lake is thermally stratified, the limed water may be overlain by a layer of more acidic water. The acidic water at the lake surface will then be discharged through the lake's outlet acidifying the fish habitat downstream. This latter problem can be overcome by winter liming. The lower solubility of lime in cold water may be compensated for by reduced cost of spreading the lime and by a relatively higher pH near the lake surface.

Flowing water must be limed continuously throughout the year. It is essential that the equipment for liming running water be able to function reliably with few breakdowns at all seasons. Revolving drums and diversion wells use the energy of flowing water to grind lime into smaller particles and to dispense it into rivers. Lime dosers have more complicated mechanisms that may be more prone to breakdown. Limestone gravel bars are the least efficient method of liming in terms of the pH rise produced by a given amount of gravel. However the pH of interstitial water in the limestone gravel may be higher than that of the water column and conditions may be favourable for juvenile salmon and salmon eggs in this interstitial water. Once liming has been carried out, the higher pH can only be maintained by frequent or continual liming.

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