FRESHWATER LIMING

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Abstract. Operational liming of surface waters is part of Sweden and Norway's strategy to counteract freshwater acidification caused by air pollutants. Smaller scale liming efforts are performed as research or experimental programs in other countries. Yearly, approx. 300,000 tons of fine-grained limestone ($CaCO_3$) is spread in lakes and streams and on wetlands to raise the pH in surface water at a cost of approximately 40–50 million $US. The chemical target is set by the biological goals and objectives. A total of over 11,000 lakes and streams are treated on a continuing basis. Dose calculations consider pH, inorganic monomeric Al, dissolved organic matter and the necessary buffering. Lake liming, limedosers at streams and terrestrial liming are used. A mix of different liming techniques is often preferred to get an optimal result. The vast majority of changes are desirable and expected. Undesirable effects may appear and damaged wetlands are probably the most serious ones. Cost-benefit analysis show that liming may be profitable for the society. Recovery of the systems can take up to 10–20 years. Liming will in the long run restore the ecosystems but will not make them identical to what may be the original ones. In some cases, complementary measures, e.g. facilitation of recolonization, are necessary to enhance recovery. Reduced emissions of acidifying pollutants according to signed protocols will decrease the need for liming, but still liming is needed for several decades in large regions to preserve biodiversity.

1. Introduction

Acid deposition has changed the natural water chemistry, and thereby has affected the biological community in 50,000–100,000 lakes and watercourses in Europe and North America (e.g. Brodin, 1995a). Aquatic biodiversity is affected as well as the possibility for human use of natural resources. The accumulation of heavy metals like mercury in top carnivores is a latent threat to animal species and human health.

The deposition of sulphur has been reduced in Europe during the last 15 years, but further reductions are needed. Although many European countries have agreed upon a sulphur reduction of 70–80 % by the year 2010 relative to 1980 (UN, 1994), acidification will still be a problem for many decades in large areas (Henriksen and Hindar, 1993; Brodin, 1995a). No large scale reductions in nitrogen deposition have been achieved and acidification due to nitrogen compounds seems to be more important relative to sulphur in the future.

One way of counteracting acidification of freshwaters is to add neutralizing agents to the water. Sweden and Norway have chosen large-scale liming as a national strategy in order to preserve species threatened by acidification (e.g. Baalsrud *et al.*, 1985; Hindar and Rosseland, 1991; Henrikson and Brodin, 1995a; Romundstad and Sandøy, 1995; Svensson *et al.*, 1995). Smaller scale liming efforts, mainly as research or experimental projects, have been conducted in several other countries, e.g. Canada, Finland, the United Kingdom (Wales and Scotland) and USA. In Sweden, liming has also been used to reduce the content of mercury in fish in acidified lakes (Meili, 1995).

In this paper, we present an overview of the need of countermeasures and the experiences of liming in different countries. Earlier surveys on liming and effects of

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liming have been presented by Brocksen and Wisniewski (1988), Weatherley (1988), Olem (1991), Olem *et al.* (1991), and Henrikson and Brodin (1995a).

2. Acidification status and the critical load concept

Aquatic organism status is correlated to a set of water quality parameters, such as pH, dissolved inorganic aluminium (Al) and calcium (Ca) (Økland and Økland, 1986; Wood and McDonald, 1987; Brown and Sadler; 1989; Rosseland *et al.*, 1990; Herrmann *et al.*, 1993; Brodin, 1995a). Ca may ameliorate the toxic effects of Al in acidic water and dissolved organic matter forms complexes with Al and thereby reduces the inorganic concentrations. Iron and manganese have been shown to form toxic species in Sweden (e.g. Nyberg *et al.*, 1995).

Aquatic species are subjected to sublethal and lethal effects at different pH/Al-levels. Differences have been found between fish species, fish strains, invertebrate and phytoplankton species (e.g. Almer *et al.*, 1978; Eriksson, *et al.*, 1983; Engblom and Lingdell, 1984; Raddum and Fjellheim, 1984; Brett, 1989; Kroglund *et al.* 1992; Lien *et al.*, 1992). The critical chemical value of pH or a combination of pH, Al or other components may therefore vary from species to species. Regional differences in pH/Al-tolerance may be found due to differences in the natural water quality and climate.

Fish status, although related to the direct toxic effects of pH and Al, is well (and better) correlated to acid neutralizing capacity (ANC) (Lien *et al.*, 1992; Bulger *et al.*, 1993). Lien *et al.* (1992) found that intact Brown trout *Salmo trutta* populations in most cases corresponded to ANC values higher than 20 μeq L^{-1}. Exceedance of the critical load is reflected by lower ANC than 20 μeq L^{-1}. A variable ANC_{limit} has recently been introduced to take into account the very low (even lower than 20 μeq L^{-1}) naturally occurring ANC-levels in parts of Norway (Henriksen *et al.*, 1995). Exceedances and thereby anticipated damage to aquatic life due to acidification are manifest in large areas of Scandinavia (Henriksen *et al.*, 1992).

Biodiversity is the variation at genetic, species, and ecosystem level, and the ecological processes of the ecosystem. All levels of aquatic biodiversity are affected by acidification. Genetic unique populations of the Salmonidae family have been damaged (Appelberg and Degerman, 1991; Bergquist, 1991; Hesthagen and Hansen, 1991; Snucins *et al.*, 1995). The total number of species is lower in acidified compared to non-acidified waters (e.g. Brodin, 1995b) even though some tolerant species may be favoured and colonize acidified waters (e.g. Henrikson and Oscarson, 1981; B.-I. Henrikson, 1988). Although biodiversity is affected at species level we only know of one species, the Spring-spawning Cisco *Coregonus trybomi*, that may be threatened on a global or national scale (cf. Henrikson and Brodin, 1995b). On a regional and local scale several species may be considered as threatened, e.g. European Roach *Rutilus rutilus*, Freshwater Pearl Mussel *Margaritifera margaritifera* but also semi-aquatic species like Red-throated Diver *Gavia stellata* (Eriksson, 1987; Brodin, 1995b; Henrikson, unpubl. data). Changes at the ecosystem level are manifested by altered community structure as well as ecological processes, e.g. retarded breakdown of leaves (e.g. Appelberg et al., 1993). If the acid deposition decreases the biodiversity will increase (e.g. Keller and Gunn, 1995; Raddum and Fjellheim, 1995).

3. Liming as a countermeasure

3.1 EXTENT, OBJECTIVES AND TARGETS

Yearly, approximately 300,000 tons of fine grained limestone is spread in lakes and streams and on wetlands in different countries to raise the pH in surface water at a cost of approx. 40–50 million $US. A total of over 11,000 lakes and streams are treated on a continuing basis in Sweden and Norway including a maximum of 150 in other countries. The liming in Norway and Sweden is financed either in large part or in total by the respective governments.

At the start of operational liming of acidified surface waters in the 1970's, the main motive was the concern of fish populations for recreational fishing. Now, the aim and direction have been broadened and are focused on the preservation or recovery of biodiversity, but also human health. In Sweden (cf. Svensson *et al.*, 1995) the official aims of today are: (1) The biological aims are to detoxicate the water for the continued existence or recolonization of natural flora and fauna, (2) The chemical aims are to raise the pH above 6.0 and alkalinity above 0.1 meq L^{-1}. In Norway (Romundstad and Sandøy, 1995) the aims are (1) to improve conditions for recreational fishing and (2) to preserve biological diversity. The chemical targets vary according to the biological target. In Norwegian salmon rivers the chemical target is differentiated at an annual basis. Target pH is higher (pH 6.5) during February–June than during the rest of the year (pH 6.2–6.3) due to the extreme sensitivity of salmon smolt.

3.2 LIMING AGENTS AND DOSES

Many kinds of deacidification agents, such as carbonates, oxides, hydroxides and industrial waste products, have been used to neutralize acid waters (Dickson and Brodin, 1995). Calcium carbonate ($CaCO_3$) as finely grained dry powder may stabilize pH at intermediate levels (pH 6–8), is cheep and easy to handle, has low content of contaminants, and is therefore most widely used (>95 %).

Dose calculations may be on a theoretical basis and consider the concentrations of H^+, inorganic monomeric Al, dissolved organic matter and the necessary buffering. A more direct approach is titration analyses. Common doses are 5–30 mg $CaCO_3$ L^{-1}. The reactions with dissolved inorganic Al result in a change of Al-species from a mixture of inorganic monomeric species to longer chains of inorganic and organic complexes. Temperature and the concentrations of ligand-forming compounds may affect the kinetics and reaction products of these reactions (Lydersen, 1990).

Carbonates dissolve as a function of pH, dissolved carbon dioxide, powder size distribution and time and conditions for dissolution (Sverdrup, 1985). These variables are taken into account, together with the fraction of carbonates in the powder, in mathematical models for both dose calculations and simulation of lake reacidification after liming. However, run-off regimes other than the average may result in longer or shorter duration of the liming intervals than expected. Long-term dissolution of sedimented limestone powder may contribute significantly to the liming effect, stabilizes the water quality and may therefore increase the duration in some lakes. Limestone doses for rivers may be even more difficult to calculate due to the variability in water quality and run-off both within and between years. Doses for wetland liming depend on pH, the limed area relative to total catchment area and liming frequency. Common doses are 0.3–1 ton ha^{-1} catchment area corresponding to 3–25 tons ha^{-1} limed area.

A model based on calculations of the critical load exceedance has been developed for dose calculations on a catchment or regional scale (Henriksen and Hindar 1993; Hindar and Henriksen, 1995). Both present and expected future sulphur and nitrogen deposition data are used and scenarios for future liming have been estimated.

3.3 LIMING METHODS AND STRATEGIES

Both lake liming, limedosers and terrestrial liming may be used (Rosseland and Hindar, 1988; Dickson and Brodin, 1995), but a mix of different liming techniques is often preferred to get an optimal result. Increased knowledge and experience may result in changes in the liming strategy over time (cf. Alenäs *et al.*, 1995).

Lake liming is most common, but often insufficient because of severe acidification of the littoral zone during snow melt (Abrahamsson, 1993), which may affect sensitive littoral species (Henrikson, 1988; Barlaup and Åtland, 1995). To avoid this, lake liming should be supplied with other techniques, such as liming of the littoral area, wetland liming or doserliming of inlet streams.

Terrestrial liming techniques such as whole-catchment liming or wetland liming have several advantages compared to lake liming. Most important is terrestrial retention of Al and deacidification of melt water (Traaen *et al.*, 1995; Hindar *et al.*, 1995; Gubala and Driscoll, 1991; Dalziel *et al.*, 1992). Although the pH-increase in streams after forest soil liming may be small (Brahmer, 1992; Westling and Skärby, 1993) forest soil liming programmes may be coupled to aquatic liming strategies because both pH and Ca increase, Al decreases and because a more stable water quality may be achieved (Hindar *et al.*, 1995).

Dosers for dry limestone powder or slurried powder are used in both small streams and large rivers. The most advanced are equipped with automatic dosing control based on pH upstream or downstream and water flow (Hindar and Henriksen, 1992). Close-downs may be detrimental, especially to salmon populations if no other liming measures are included.

Adequate water quality, with elevated pH and low concentrations of inorganic monomeric Al, is necessary to reach the biological targets. Al must have time to reach more stable forms when pH increases to avoid mixing zones, which may be even more toxic than the acid water itself (Rosseland and Hindar, 1991; Rosseland *et al.*, 1992). Mixing zones are probably especially significant in salmon rivers due to the extreme sensitivity of salmon smolts. Acid, Al-rich tributaries of some size should therefore be limed, although the biological targets in these particular rivers may be insignificant.

3.4 ECOLOGICAL EFFECTS OF LIMING

Generalizing about the ecological effects after liming is difficult, partly because of a lack of data records. Long-term biological changes have been documented 16 years after liming (Appelberg, 1995) but such changes may be related to delayed re-colonization of extinct species rather than liming itself. Expected effects related to the chemical and biological targets may be considered as desirable, others as undesirable.

The experiences from large scale liming in Sweden and Norway as well as liming in USA and Scotland are that the vast majority of chemical, biological and ecological changes are desirable (Baalsrud *et al.*, 1985; Porcella, 1989; Hindar, 1992; Howells and Dalziel, 1992; Henrikson and Brodin, 1995b).

3.4.1 Desirable effects

The primary effects on water quality are increases in pH, alkalinity, ANC, Ca content and decreases in toxic metal species (cf. Wilander *et al.*, 1995). Biologically most important, together with elevated pH, is the decrease of the toxic species of Al and other metals like iron and manganese.

In polyhumic lakes the content of dissolved organic substances (water colour) decreases, whereas increases as well as decreases can occur in waters of lower humic content. Such changes may secondarily affect the light climate of the water. Immediately after liming there is an increased turbidity and a transient decrease in transparency. In the long term the transparency may increase as well as decrease, which may be related to the phytoplankton production and the occurrence of humic substances.

Short-term studies show a transient elevation of the phosphorus (P) content, probably due to phosphorus in the liming agent and release of P bound in the sediments or in decaying plants due to increased mineralization. Also, decreased P after liming, probably due to adsorption to the carbonate particles, has been measured (Blomqvist *et al.*, 1993). Long-term studies indicate unchanged or decreased contents. However, the accessibility of phosphorus to algae may increase. In the few studies of nitrogen, both increases or decreases have been documented.

The most obvious desirable biological effects after liming are successful fish reproduction, increased density of sensitive species and re-colonization of eradicated species (Appelberg, 1995; Bergquist, 1995; Degerman *et al.*, 1995; Larsson, 1995). Another desirable effect is the decrease or elimination of species favoured by acidification.

In figure 1 important factors and processes for the biological development after liming are illustrated. The abiotic changes are the triggers for all other changes but in the long run biotic mechanism, i.e. competition and predation, will determine the community structure.

The community structure will stabilize within the new abiotic limits set by the mix of liming techniques used. The community will be more complex, as seen by increased species number and trophic levels (Appelberg, 1995). Ecosystem processes like decomposition will become more normal (Gahnström, 1995; Gahnström and Andersson, 1995). Ecosystems severely damaged by acidification will experience the most profound changes in the community after liming (Henrikson *et al.*, 1985; Degerman *et al.*, 1995).

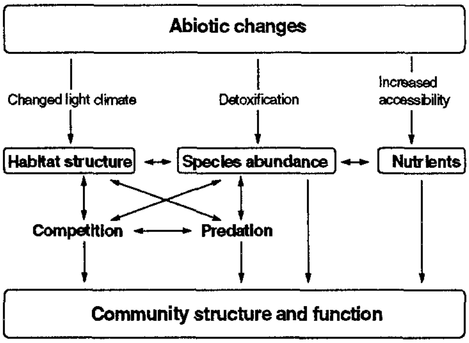

Fig. 1. Schematic illustration of important factors and processes for the biological development after liming. The direct action of altered water quality will affect species abundance, nutrient conditions and habitat structure. The detoxification will facilitate the fish reproduction and populations increase for fish and other sensitive organisms and the recolonization of eradicated species. This will affect the species abundance. The accessibility of the important plant nutrient phosphorus will increase when the Al content decreases or less toxic species of Al is formed, which will favour the production of algae. If the transparency is decreased, the habitat structure is changed, which may affect visual dependant predators like birds. The decrease of *Sphagnum* mat will change the habitat for benthic species. The primary changes caused by the new abiotic environment will release further biotic changes caused by competition and predation. The result is a "new" structure of the organisms community. As a consequence of changes in community structure also the functions within the ecosystems are changed. One example is increased breakdown of leaves due to increased number of shredders.

3.4.2 *Undesirable effects*

The overwhelming desirable effects of liming must be weighted against undesirable effects which also have been documented after liming. Undesirable effects may be divided into terrestrial and aquatic effects.

Direct damage to terrestrial vegetation after wetland or whole-catchment liming are probably the most severe of the undesirable effects. Terrestrial liming, especially wetland liming, is rather widely used as liming technique in Sweden. Death of *Sphagnum* mosses (Mackenzie, 1992; Hindar *et al.*, 1995; Larsson, 1995) changes the bog surfaces on the short term and probably alter both structure and function of bogs after some decades of repeated liming. More frequent use of smaller doses, coarser liming material and introduction of Mg-containing dolomite may reduce these unwanted effects.

In some lakes in Norway and Sweden increased expansion of *Juncus bulbosus* and *Myriophyllum alterniflorum* after liming has occurred (Brandrud and Roelofs, 1995; Dickson *et al.*, 1995; Larsson, 1995; Roelofs and Brandrud, 1995). This may have great impact on littoral flora and indirectly on littoral fauna and thereby on lake ecology. Increased availability of inorganic carbon in the littoral sediments as a result of carbonate addition may stimulate the increased growth.

3.5 SOCIO-ECONOMIC EFFECTS

Cost-benefit analysis in Norway and Sweden show that liming may be profitable (e.g. Navrud, 1990, 1993a, b; Krokan, 1992; Bengtsson and Bogelius, 1995). In these studies the cost of liming and fishery management has been related to benefit expressed as recreational value, i.e. the willingness to pay for fishing. The people around a limed Finnish lake was willingly to contribute to the costs of liming even if the lake only was used for outdoor recreation (Iivonen *et al.*, 1995). However, this kind of analysis only recognize a part of the economic aspects, not the value of e.g. preserved biodiversity.

Bengtsson and Bogelius (1995) also state that liming practices contribute to enhance environmental awareness among the people.

4. Synthesis

4.1 LIMING TO RECREATE PREVIOUS WATER QUALITY AND RESTORE ECOSYSTEMS?

Successful liming operations will lead to improved water quality and increased number of species susceptible to acidification, but do the ecosystems recover completely? Several difficulties must be considered. First of all, nobody knows the exact structure and function of the ecosystems before they were influenced by the anthropogenic acidification. Second, all ecosystems experience a developmental process and a certain degree of unstability between years even if the external conditions remain more or less constant (cf. Brink *et al.*, 1988).

Well aware of the uncertainties, we state that if the chemical target is met liming in the long run will restore the ecosystems but will not make them identical to what may have been original (Figure 2) (cf. Degerman *et al.*, 1995; Henrikson and Brodin, 1995b; Lingdell and Engblom, 1995; Wilander *et al.*, 1995). That the ecosystems are not completely recovered is not specific for liming but also for other attempts to repair disturbed habitats of aquatic as well as terrestrial systems (cf. Brink *et al.*, 1988; Cairns, 1988).

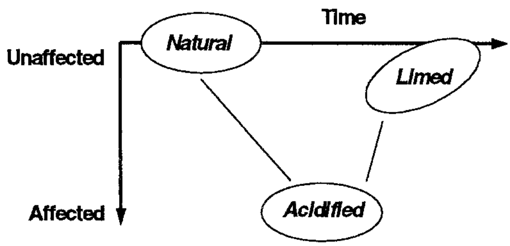

Fig. 2. Generalization of changes occurring in freshwater ecosystems influenced by acidification and liming. Acidification results in decreased ecosystem complexity. Successful liming entails a "normalization", i.e. the complexity and function of the ecosystem will be largely similar to unaffected ecosystems, yet the ecosystems will not become identical and liming alone might not bring back completely natural conditions. However, with time the similarity with unaffected waters will increase.

There may be several reasons why the limed waters differ from waters unaffected by acidification (cf. Appelberg, 1995; Degerman *et al.*, 1995). The most common reason is probably that the chemical target is not met due to insufficient planning or severe practical difficulties. Limed waters may become rather unstable if acidified waters continuously are discharged into the lakes and streams. Toxic water and the variable environment will affect the normal competition relations, favouring non-competitive "opportunistic" species. Also, changed nutrient availability and slow recolonization may affect the result of liming. Improved liming strategy and complementary measures may improve the success of liming.

Complementary measures are a part of the Swedish liming programme. Bergquist (1995) defined two categories of "biological restoration": (1) measures aimed at facilitating natural re-colonization and re-establishment of species which have been eliminated, and (2) direct reintroduction of species by man. Examples are; elimination of migration obstacles, biotope reconstruction, and replenishment of individuals for species occurring in restricted numbers. Supply of nutrients, especially phosphorus, is another complementary measure in order to restore the productivity of limed waters. Hitherto, such measures have been taken only in a few research projects (Blomqvist *et al.*, 1993) and more experience is needed before these measures can be operational.

4.2 PERSPECTIVES ON LONG-TERM LIMING

As a consequence of deposition scenarios according to the commitments of the UN sulphur protocol (UN, 1994) liming in Norway will be reduced to one third some time after year 2010 (Henriksen and Hindar, 1993). Numbers in Table I are based on the introduction of a variable ANC_{limit} (Henriksen *et al.*, 1995).

TABLE I.
Effects of decreased exceedance of the critical load in southern and northern Norway according to the commitments of the UN (1994) protocol for sulphur deposition reductions. Figures are based on a variable ANC_{limit}, see Henriksen *et al.* (1995). (Data from A. Henriksen.)

	AREA EXCEEDED percent		AMOUNT OF LIME tons $yr.^{-1}$		COST mill. NKR	
	Today	Year 2010	Today	Year 2010	Today	Year 2010
South of Norway	55.0	21.2	390,000	149,000	312	120
North of Norway	6.4	1.2	22,000	5,600	23	4.3
Norway	29.7	10.8	412,000	155,000	335	124

Decreased exceedance of the critical load will probably result in decreased Al concentrations and to a decrease in the frequency and magnitude of acid episodes. Target-pH for liming may therefore be lower in this improved environment, thus reducing the liming costs further.

We are not yet able to estimate the exact timing of water quality improvements to given levels after future deposition reductions. Dynamic models, such as MAGIC (Wright *et al.*, 1988), have been used but lack of long data records during recovery makes the forecasts uncertain. Due to the time lags liming operations may probably be longer lasting than recognized from sulphur deposition scenarios, i.e. several decades.

Metals like Al may polymerize and eventually be precipitated as humic–metal complexes after liming (Egeberg *et al.*, 1995). Some has argued that easily available Al from such sediments could represent a threat to aquatic life during reacidification after liming and after close-downs of lime dosers. However, both theoretic considerations and monitoring data show that precipitated Al does not represent an additional Al-source of any significance for aquatic life during these circumstances (Hindar and Lydersen, 1995).

Wetland liming and whole-catchment liming may stimulate the decomposition of organic matter. A high carbon/nitrogen relation of the organic matter may reduce this stimulating effect and increased decomposition may therefore be less pronounced under humic/low-temperature climate as in Scandinavia. Nevertheless, liming for decades will probably change the structure and function of bog surface and the humic layer of e.g. forest soils. Knowledge of the quality and speed of these changes is needed.

Long-lasting liming obviously results in accumulation of undissolved minerals on lake bottoms and stream beds. So far, research activities have focused on the suitability of limed sediments as habitat for invertebrates and as spawning sites for fish. The ecological effects have not been studied.

In spite of increasing knowledge it is important to point out the short-comings and lack of scientific knowledge. We highlight the following (see also Henrikson and Brodin, 1995b; Kroglund *et al.*, 1995):

- long-term ecological effects, i.e. more than 20 years,
- concentrations and accessibility of nutrients,
- primary production,
- the structure and function of limed wetlands,
- re-colonization and reintroduction of species,
- strategies and methods to eliminate harmful effects of acidic episodes,
- improvements of liming techniques to reduce harmful effects in limed wetlands,
- identification of measurable biological targets others than fish and invertebrates.

5. Concluding remarks

We state that liming of acidified freshwaters preserves biodiversity and improves conditions for recreational fishing. Undesirable effects have been documented. Although further sulphur emission reductions are expected, acidification will still be a serious environmental problem and liming will be needed for several decades to come.

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