

Environmental impacts on freshwaters: acidification as a global problem

Roland Psenner

University of Innsbruck, Institute of Zoology, A-6020 Innsbruck, Austria

Abstract

Nutrient and acid loads are the two main global impacts on freshwaters. Recognition of problems began in 1960–1970 both for eutrophication and acidification, but changes in lake ecosystems depending on nutrient inputs started 10 years earlier, whereas acidification of sensitive lakes began approximately 100 years ago. Freshwater acidification is no longer restricted to the northern hemisphere but it affects industrialized regions of the whole world. Reduction of eutrophication in large lakes of developed countries was partly successful, but effective measures against acidification are still missing, although SO₂ emissions and deposition of sulfuric acid are decreasing: sulfate in acid rain has largely been replaced by nitrogen compounds. Moreover and in contrast to chemical viewpoints, recent studies have shown that biological effects of acidification start very early (approx. pH 6.5); for sensitive organisms the chemical threshold of acid water, i.e. pH 5.5 or zero alkalinity, represents the end point rather than the onset of acidification. The application of models and the reconstruction of the pH history of high altitude lakes in the Alps showed that very sensitive lakes can acidify even at low acid inputs and that climate changes can strongly interact with anthropogenic emissions of acids.

Key words: Nutrient load; Eutrophication; Acid load; Acidification; Freshwaters; High altitude Alpine lakes; Anthropogenic impacts

1. Introduction

Eutrophication by enhanced phosphorus loading was one of the main global impacts on freshwaters since the early sixties. Problems of eutrophication still persist in many industrialized and developing countries (Forsberg, 1987; Gächter, 1987), but for a long time countries of the northern hemisphere were regarded as the only ones suffering from freshwater acidification until Rodhe and Herrera (1988) showed that also acidification has become an issue of global dimensions. Although acidification was studied under the aspect of oligotrophication — one of the possible side-effects of acid loading — and moderate nutrient input was envisaged as a remedy for acid lakes (Davison 1987; Davison and Woof 1990), sources, sites and mechanisms of both environmental threats are different (Table 1).

Eutrophication is a result of increased nutrient

input into lakes and rivers leading to problems such as algal blooms, low transparency, oxygen deficiency and hydrogen sulfide production. Solutions to the problems must be sought in a better management of sewage and a reduction of nutrient application in the catchment. Acidification results from atmospheric depositions of SO_x and NO_x and affects the chemical behaviour of aluminum, iron and other heavy metals by low pH values (Table 1). On the long term, its effects can be reduced solely by curbing emissions of acid precursors. Nonetheless, acidification and eutrophication have some effects in common: changes in species composition, extinction of sensitive species, toxicity for plants and animals (H⁺ and Al³⁺ in acid waters; H₂S and cyanobacterial toxins in eutrophic lakes). Also remedies and effective restoration measures have the same rationale and background: reduction of loading. All other methods have only transitory effects and

Table 1
Comparison of eutrophication and acidification as the two major freshwater problems

	Eutrophication	Acidification
Agent		
Primary	P	H ₂ SO ₄
Secondary	N	HNO ₃ , NH ₃ , organic acids
Sources		
Natural	Weathering, dust, organic matter, sediments	Volcanism, plant growth, oxidation of pyrite and ammonium
Man-made	Detergents, waste water, tourism, agriculture	Industry, traffic, argiculture, heating
Main transport routes	From catchment	Long-range transport
Sites	Lowland lakes	Remote lakes
Geology	Carbonate, silicate	Silicate
Beginning of impact	1950–1960	1850–1920
Recognition of problems	1960	1970
Delay of recognition	5–10 years	50–100 years
Effects		
Chemistry	Oxygen depletion, low transparency, H ₂ S and odour emission	Low pH values, high aluminum and heavy metal concentrations
Biology	Changes in (algal) species composition, high biomass of algae and bacteria	Changes in species composition, extinction of sensitive invertebrates and fishes
Human use	Problems for drinking water supply, bathing, recreation	Toxicity for plants, animals, and humans; corrosion
Restoration	Since 1960	Since 1970
Within lake	Oxygenation, deep-water renewal, iron or aluminum precipitation, biomanipulation	Liming, soda ash briquettes, (addition of organic matter and nutrients)
Within catchment	Reduction of loading , sewage diversion, agricultural changes	Liming
Outside catchment	Reduction of P and N fluxes in the biosphere	Reduction of SO_x and NO_x emissions

Effective measures are highlighted with **bold** letters

may affect the ecosystem in an unpredictable manner, e.g. by extreme pH pulses during liming of acid lakes (Christophersen et al., 1990). A striking difference is observed in the recognition of the problems (Table 1). Measures against eutrophication were undertaken with a time lag of one decade or so, although in some cases not very efficiently: some large lakes in Europe are still eutrophic. Acidification, on the other hand, can be regarded as an unrecognized problem were

effects and counter-measures differ by almost one century. Palaeolimnological data clearly demonstrate the early onset of acidification in sensitive lakes (approx. 1900, Fig. 1).

Acidification of freshwaters was recognized as a major environmental issue only in the seventies (Drablos and Tollan, 1980) and effective measures to reduce SO_x emissions were started around 1980. Nonetheless, improvements in water quality are hardly seen (Fig. 2), partly because of

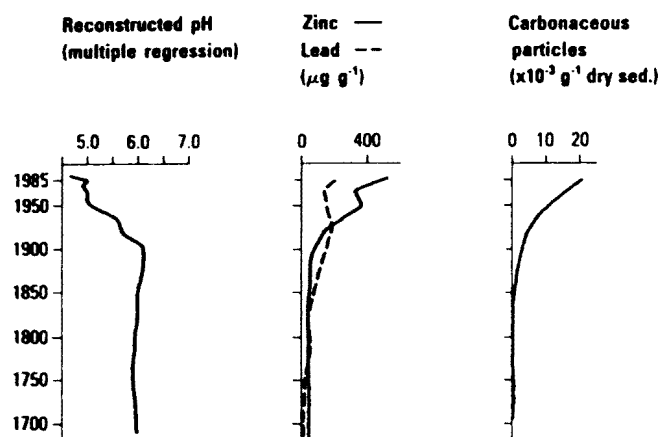


Fig. 1. Reconstructed pH, zinc, lead and carbonaceous particles for Llyn y Bi, Wales. From Battarbee et al. (1988).

NO_x emission increase during the last decade as shown for lakes in southern Norway.

The history of eutrophication shows us that also large lakes with water residence times of several years such as lake Constance or Lago Maggiore can be restored (Fig. 3) if nutrients are diverted.

Acidification of freshwaters, however, was an unidentified problem for many years; affects remote lakes often representing a reservoir of endangered species or the remainders of ecosystems in a densely populated world; counter-measures

still did not substantially improve the situation; and restoration measures are efficient only on a global scale.

2. Freshwater acidification from a chemist's point of view

Lakes and rivers become acidic when the input of acids exceeds the amount of bases produced in the catchment by weathering of rocks or in the lake itself by reduction of acid anions, such as sulfate and nitrate (Schnoor and Stumm, 1986).

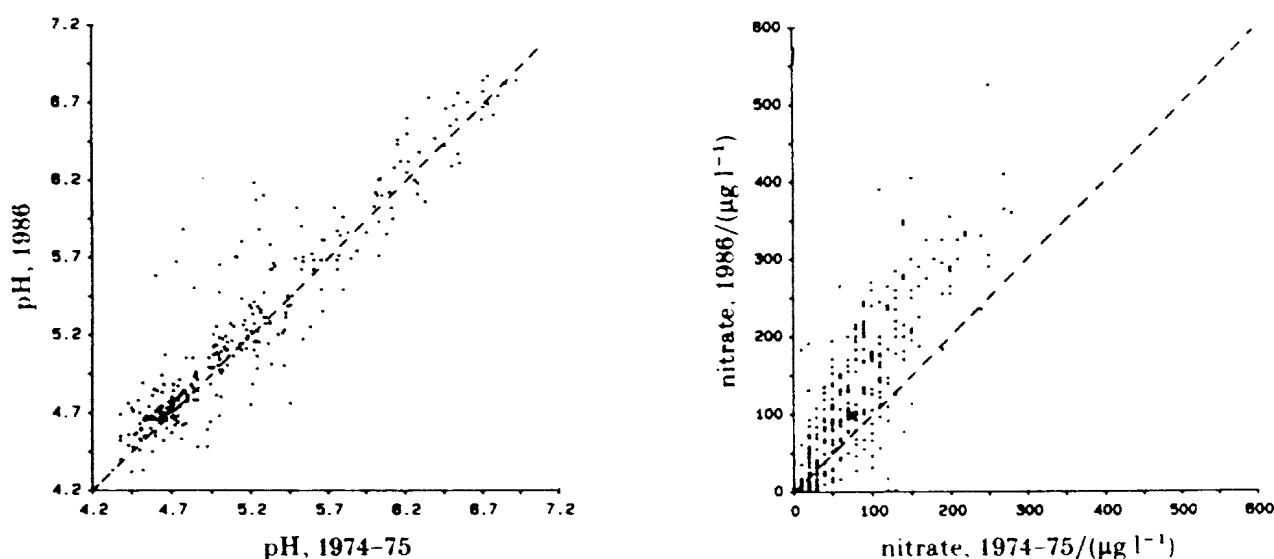


Fig. 2. Comparison of pH (left) and nitrate (right) in 1986 with those in 1974–75 for 305 lakes in southern Norway. From Henriksen et al. (1990).

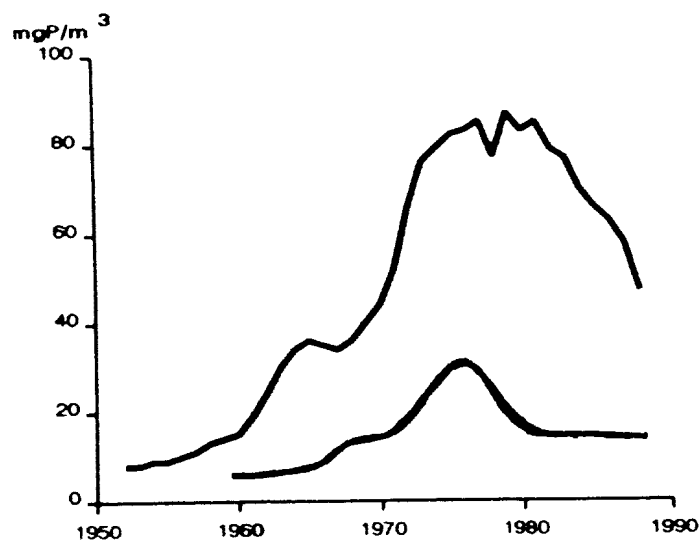


Fig. 3. Concentrations of soluble reactive phosphate (mg P m^{-3}) in lake Constance (upper curve) and Lago Maggiore (lower curve). From Geller and Güde (1989) and Mosello (1989).

Acidification occurs in siliceous catchments with low weathering rates. Schnoor (1990) reports empirical data for weathering rates measured by cation export from siliceous rocks which are generally between 20 and 100 $\text{mEqiv m}^{-2} \text{ a}^{-1}$. Sensitive bedrock geology is found in several parts of Europe (Fig. 4) with acidic depositions equal to or higher than cation release from rocks and soils. In such areas, the alkalinity balance

$$\begin{aligned} \text{ALK} &= [\text{Ca}^{2+}] + [\text{Mg}^{2+}] + [\text{Na}^+] + [\text{K}^+] - \\ &\quad [\text{SO}_4^{2-}] - [\text{NO}_3^-] - [\text{Cl}^-] \\ &= [\text{HCO}_3^-] + [\text{CO}_3^{2-}] + [\text{OH}^-] - [\text{H}^+] \end{aligned} \quad (1)$$

may become zero, and proton concentrations equal bicarbonate ions, the pH value at this point being around 5.5. The pH of 5.5 represents the lower end of the bicarbonate buffering system

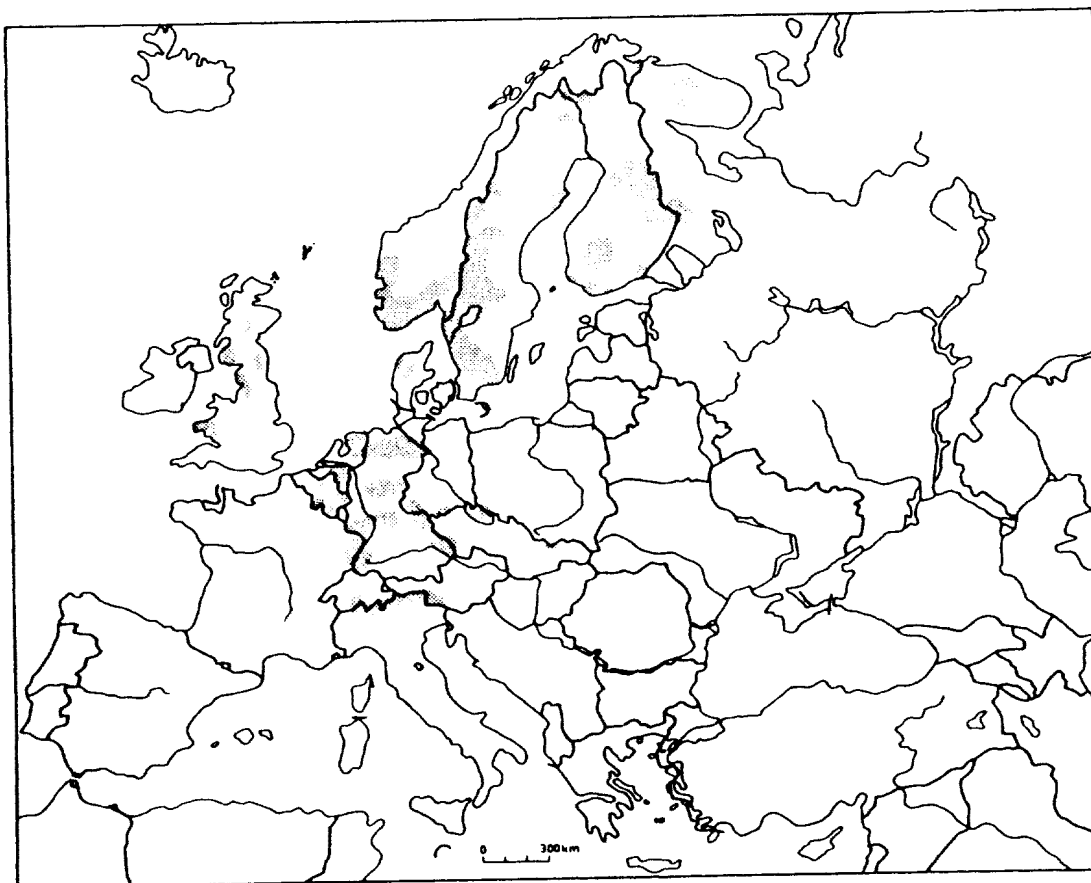


Fig. 4. Areas in Europe with acidified surface waters. From Merilehto et al. (1988).

and a crucial point for the release of aluminum ions: aluminum in the form of Al^{3+} , $\text{Al}(\text{OH})^{2+}$ and $\text{Al}(\text{OH})_2^+$ has extremely low concentrations at pH 6.0–7.0 but it may reach toxic values for fish at pH 5.0–5.5. The release of aluminum cations, however, prevents waters from assuming very low pH values when acidic depositions exceed the acid neutralizing capacity of soils and bedrock (Zobrist et al., 1987). Acid waters have, by definition, a pH of 5.5 or less; for sensitive species, however, such as molluscs, crustaceans and fishes, the pH of 5.5 represents the end point rather than the beginning of acidification.

3. Case study: high altitude lakes in the Alps

Surface water acidification in the whole ECE region has been documented by Merilehto et al. (1998) in the form of a survey (Fig. 4), and extensive studies and comparisons have been made, e.g. in Scandinavia and England (Mason, 1990). Lake water acidification in the Alps was reported since 1980 (Stumm and Righetti, 1982; Mosello, 1984; Psenner et al., 1988; Psenner, 1989). Quantification on a statistical basis was done by Mosello et al. (1992) who studied 413 lakes in the Italian, Swiss and Austrian Alps situated at altitudes between 820 and 3001 m a.s.l. For a better comparison, 210 lakes lying in siliceous watersheds were chosen. The 'typical' high mountain lake of this study was small (approx. 1 ha of surface area), situated at an altitude of about 2400 m a.s.l. and receiving approx. 1400 mm of precipitation per year. The ionic composition showed a dominance of calcium among cations and sulfate among anions (Table 2). Nitrate was quantitatively important as acid anion in high altitude lakes of the Alps. These findings were in contrast to observations in Norway before 1980 where nitrate was considered as unimportant in the acid-base balance (Henriksen, 1980). A survey of 1000 Norwegian lakes in 1986, however, showed that nitrate concentrations have increased (Fig. 2) while sulfate decreased in one decade of observations (Henriksen et al., 1990).

Nine sampling stations for atmospheric depositions, situated between 1140 and 3105 m a.s.l.

Table 2

Ionic composition of 210 mountain lakes from the Alps lying in siliceous catchments

Variable	Unit	Mean	Min	Median	Max
Conduct.	$\mu\text{S}_{18^\circ\text{cm}}^{-1}$	22	5	18	254
pH		6.57	4.64	6.59	8.51
Alkalinity	$\mu\text{Equiv l}^{-1}$	95	–23	51	1521
Calcium	$\mu\text{Equiv l}^{-1}$	152	15	109	2495
Magnesium	$\mu\text{Equiv l}^{-1}$	34	3	21	526
Sodium	$\mu\text{Equiv l}^{-1}$	17	2	15	106
Potassium	$\mu\text{Equiv l}^{-1}$	10	0	8	123
Ammonium	$\mu\text{Equiv l}^{-1}$	2	0	1	71
Sulfate	$\mu\text{Equiv l}^{-1}$	91	17	67	1441
Nitrate	$\mu\text{Equiv l}^{-1}$	18	0	18	51
Chloride	$\mu\text{Equiv l}^{-1}$	6	0	5	60
Silica	$\mu\text{mol l}^{-1}$	32	1	27	231

From Mosello et al. (1992).

from the western to the eastern Alps, showed potential acid loads ($\text{SO}_4 + \text{NO}_3 + \text{NH}_4$) of 32–172 mEquiv $\text{m}^{-2} \text{a}^{-1}$ with an average of 70 mEquiv $\text{m}^{-2} \text{a}^{-1}$ for alpine catchments (Mosello et al., 1992). Sulfur to nitrogen ratios were significantly less than 1 on an equivalence basis. During the period of 1981–1989, nitrate concentrations in precipitations at Lago Toggia, 2160 m a.s.l., increased by 0.8 $\mu\text{Equiv l}^{-1} \text{a}^{-1}$, whereas sulfate decreased by 1.6 $\mu\text{Equiv l}^{-1} \text{a}^{-1}$.

Two-thirds of all lakes and more than 80% of lakes in acidic catchments (mainly composed of granites and gneisses) were classified as sensitive to acidification (alkalinity < 200 $\mu\text{Equiv l}^{-1}$). One-third of all lakes and 50% of lakes in acidic catchments must be regarded as very sensitive to acidification (alkalinity < 50 $\mu\text{Equiv l}^{-1}$). The pH of the lakes was independent of altitude but the scatter (pH 4.64–9.14) was higher in small lakes.

Alkalinity losses or gains were calculated by using a variable *F*-factor model (Brakke et al., 1990). This factor

$$F = \frac{\Delta (\text{Ca} + \text{Mg})}{\Delta (\text{SO}_4 + \text{NO}_3)} \quad (2)$$

indicates whether increasing inputs of acids ($\text{SO}_4 + \text{NO}_3$) are buffered by enhanced weathering of basic cations (Ca + Mg) or not. *F* may assume values between 0, i.e. no increase of cation weath-

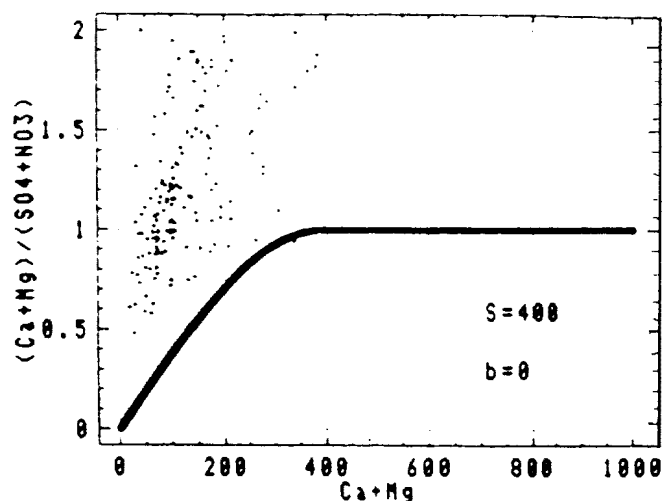


Fig. 5. The ratio of (calcium and magnesium) to (sulfate and nitrate) in dependence of (calcium and magnesium) concentrations in mountain lakes of the Alps. The curve corresponds to Eqn. (3) with an S value of $400 \mu\text{Equiv l}^{-1}$ and a sulfater and nitrate background of zero. From Mosello et al. (1992).

ering (for instance on bedrock very resistant to weathering), and 1 corresponding to a complete neutralization of acid inputs (Fig. 5). The data were fit to

$$F = \sin \left(90^\circ \frac{\text{Ca} + \text{Mg}}{S} \right) \quad S \dots 400 \mu\text{Equiv l}^{-1} \quad (3)$$

assuming that in lakes with concentrations of $([\text{Ca}^{2+}] + [\text{Mg}^{2+}])$ exceeding $400 \mu\text{Equiv l}^{-1}$, all acid inputs are completely neutralized ($F = 1$). By taking into account a variable F -factor, alkalinity changes gave more narrow and reasonable limits with median values around $28 \mu\text{Equiv l}^{-1}$ (Fig. 6b) than calculations based on the assumption of $F = 0$ (Fig. 6a).

4. Critical loads of acids to sensitive ecosystems

This variable F -factor was used to calculate target loads (L_t) for each individual lake by assuming that a critical alkalinity level ($\text{ALK}_{\text{limit}}$) of $20 \mu\text{Equiv l}^{-1}$ be maintained. This level is regarded as a minimum requirement for the survival of fish. Very sensitive species (molluscs, crustaceans, diatoms), however, may suffer or become extinct even at higher alkalinity levels

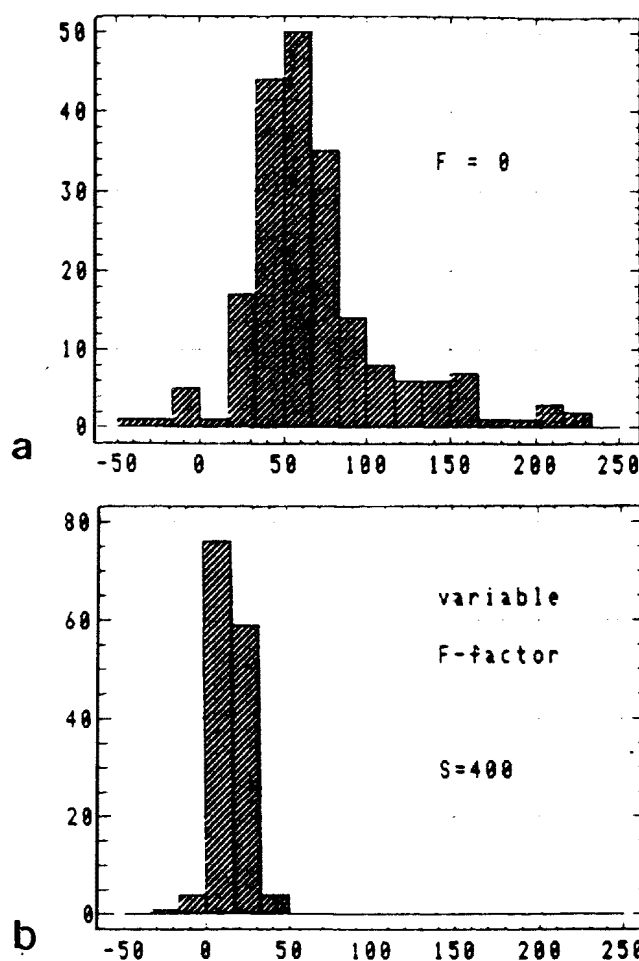


Fig. 6. Alkalinity losses ($\mu\text{Equiv l}^{-1}$) in mountain lakes of the Alps having less than $400 \mu\text{Equiv l}^{-1}$ of (calcium and magnesium); (a) calculated under the assumption of $F = 0$ (no increase of the weathering rate upon increased acid deposition); (b) calculated assuming a variable F factor (enhanced weathering of basic cations if acid depositions increase). From Mosello et al. (1992).

between 20 and $50 \mu\text{Equiv l}^{-1}$.

$$L_t = \frac{L_d (1 - F) + (\text{ALK}_{\text{lake}} - \text{ALK}_{\text{limit}}) (P - E)}{(1 - F)} \quad (4)$$

where L_t is target load ($\text{mEquiv m}^{-2} \text{ a}^{-1}$); L_d , acid load ($\text{mEquiv m}^{-2} \text{ a}^{-1}$); ALK_{lake} , lake alkalinity ($\mu\text{eq l}^{-1}$); $\text{ALK}_{\text{limit}}$, critical alkalinity ($20 \mu\text{Equiv l}^{-1}$); P , precipitation (m a^{-1}); and E , evaporation (m a^{-1}).

The target load for the most sensitive lakes in the Alps is near zero, but we may accept that 10% of the most sensitive lakes have alkalinity values below $20 \mu\text{Equiv l}^{-1}$. In this case, the

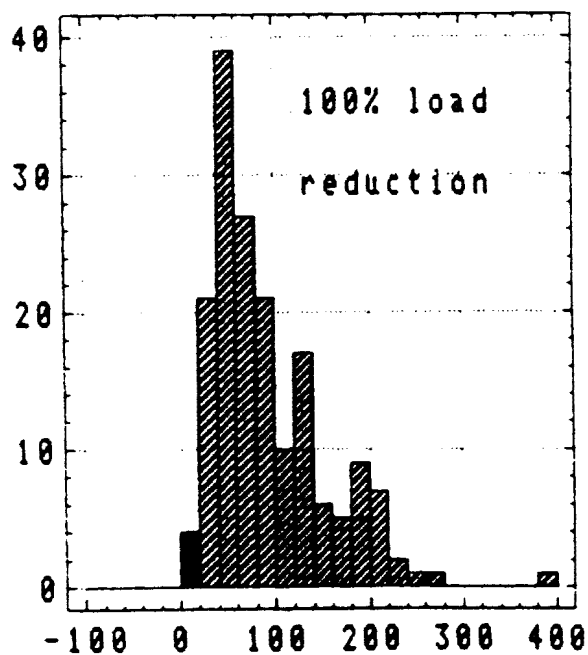
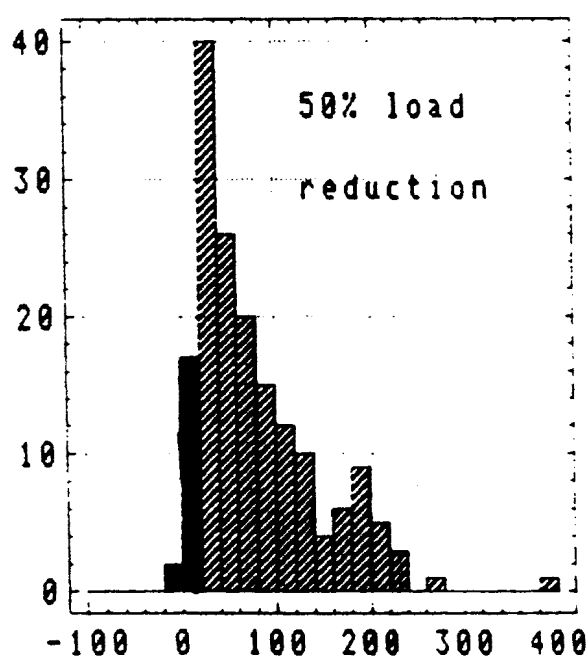
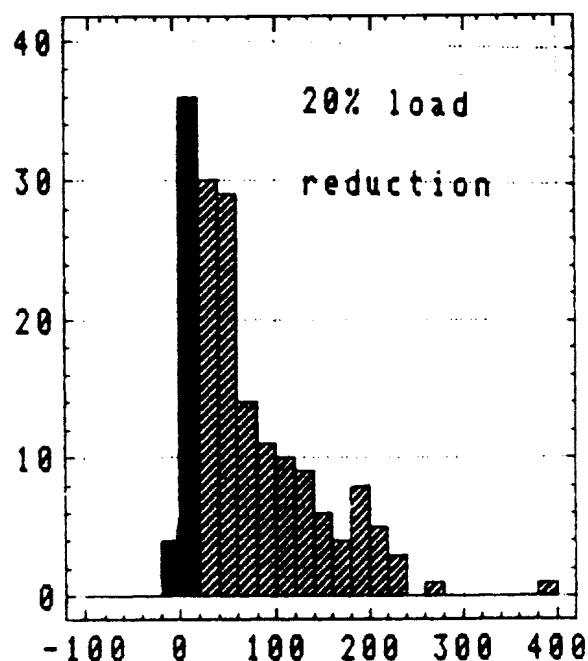
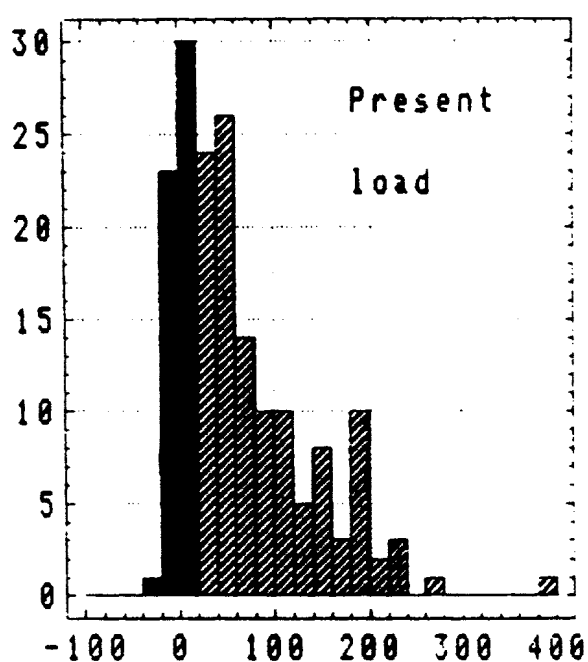


Fig. 7. Frequency distribution of lake alkalinity ($\mu\text{Equiv l}^{-1}$) in sensitive alpine lakes at the present situation (a) and under the assumption of a reduction of acid load by 20, 50 and 100%. Black bars: lakes below the critical alkalinity of 20 $\mu\text{Equiv l}^{-1}$. From Mosello et al. (1992).

critical acid load should be less than 34 mEq $\text{m}^{-2} \text{a}^{-1}$. At the present load, we have approx. 30% of sensitive lakes with alkalinities below the critical value of 20 $\mu\text{Eq} \text{L}^{-1}$ (Fig. 7a). If we reduce the acid load by 20%, there are still approx 20% of lakes remaining below the critical alkalinity, and a 50% reduction brings us to the 'acceptable' 10% of lakes below the critical alkalinity of 20 $\mu\text{Eq} \text{L}^{-1}$ (Figs. 7b,c). The authors of the study point out that the term acceptability does not imply consent to the fact that 10% of all lakes are not suitable for sensitive species. A 100% reduction of the present acid load still leaves 2% of the lakes below the critical alkalinity level (Fig. 7d) showing that some lakes in the Alps are so sensitive that even a total reduction of acid loads would not allow them to have alkalinities of 20 $\mu\text{Eq} \text{L}^{-1}$. If we envisage a 'realistic' or acceptable scenario, a 50% reduction should be achieved to keep 90% of high altitude lakes at alkalinity values above 20 $\mu\text{Eq} \text{L}^{-1}$.

5. Natural and anthropogenic impacts

Support for the calculations of anthropogenic acidification in the Alps came from palaeolimnological studies of diatom assemblages in the sediments of two high altitude lakes in South Tyrol (Psenner and Schmidt, 1992). Historic pH values inferred from the composition of diatoms in sediment layers (Fig. 8) showed fluctuation during the 19th century which could be attributed to climate changes: cold periods were associated with lower pH values: during cold years, stratification and decomposition processes were lower, flushing rates were higher and perennial snow fields may have formed in the catchments. These processes reduce the potential of alkalinity generation both in the watershed and the lake itself.

During warm periods with increased evaporation, longer water retention times, stronger stratification and enhanced assimilation/dissimilation, the pH was higher. The disappearance of snow fields in the watershed may have increased the dispersal of dust, therefore increasing the alkalinity of the water. Accumulation of organic matter and a corresponding shift of the iron to manganese ratio in different sediment layers support

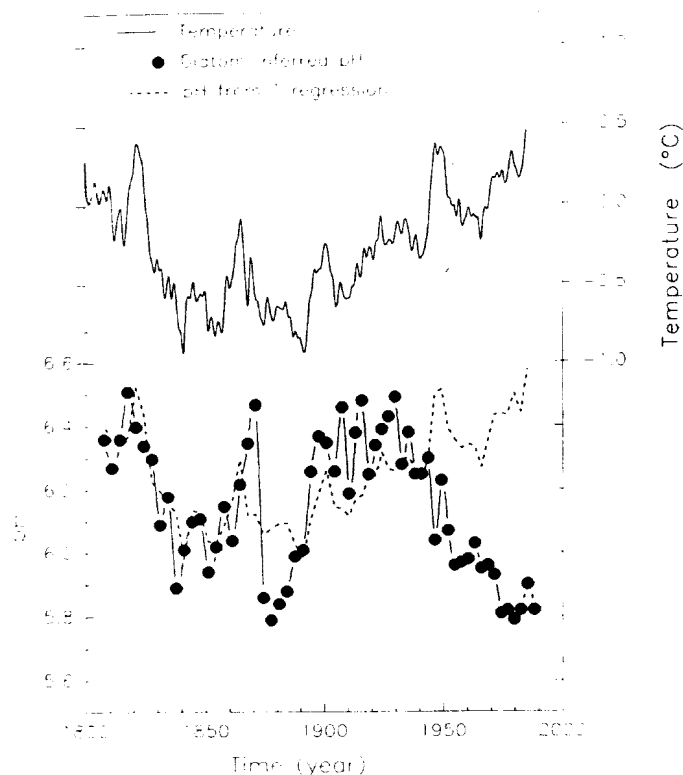


Fig. 8. Comparison of the diatom inferred pH from Rassas See, South Tyrol, Italy (2682 m a.s.l.), with mean Austrian temperatures (running averages, $n = 9$) and pH calculated from a linear temperature regression by using the data before 1907 ($n = 30$). The onset of acid depositions at the beginning of the 20th century lead to a decoupling of temperature and pH curves.

the view of in-lake processes influencing the acid-base equilibrium of sensitive lakes (Psenner and Schmidt, 1992). Terrestrial vegetation changes in the watershed were shown to be of less importance for freshwater acidification in Scotland (Battarbee et al., 1989) and seem to be even less important in high altitude regions of the Alps. These natural temperature-driven fluctuations of pH were disrupted by the onset of acidic depositions around 1900 (Fig. 8): recent pH values should be higher by approximately 0.8 units if the climate-pH coupling would still exist. The small pH peak around 1950 may reflect the influence of one of the warmest periods observed in the Alps for about 5300 years (Patzelt, pers. comm.). The trend reversal observed in the uppermost centimeters of the sediment can be interpreted as a response to reduced emissions of SO_x or to increasing temperatures since 1980.

6. Conclusions

Both natural processes and anthropogenic impacts influence sensitive lakes in the Alps. Acidic depositions since the beginning of this century have decreased the pH by approximately 0.8 units according to palaeolimnological data or by 28 $\mu\text{Eq l}^{-1}$ of alkalinity as determined from model calculations. Contrary to eutrophication where measures within the catchment can be effective, acidification can be mitigated only by global emission controls. We must reduce the deposition of acids by approximately 50% in order to arrive at tolerable levels of alkalinity in 90% of alpine lakes. Restoration measures both for eutrophic and acidic waters are often focussed on some selected features regarding the suitability for bathing, drinking, fishing, etc. The central objective of all measures, however, must be to bring aquatic ecosystems as close as possible to the previous or undisturbed state.

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