

Chapter 13 Indirect effects of acidifying compounds on natural systems: critical loads

Introduction

Historical data provide evidence of increasing transport of sulfate (SO_4^{2-}) in natural systems, by factors of up to 2 and 3.5 in 1950 and 1980, respectively, in comparison to preindustrial levels (1). Sulfur dioxide emissions are largely due to the combustion of fossil fuels for power generation, for industrial production processes and by households. These emissions provide a significant contribution to air pollution in urban areas and on a regional or wider scale. Sulfur dioxide emissions have declined over the last decade in Europe and North America following international agreements on abatement policies, but have increased rapidly in areas with high economic growth such as south-east Asia. In Europe, about 41 Mt of sulfur dioxide were emitted in 1990 compared to about 55 Mt in 1980 (2). In the Asian-Pacific region, emissions in 1990 reached about 35 Mt and are expected to increase rapidly (3). The risk of widespread damage to natural systems (i.e. acidification) due to sulfur emissions may have decreased in Europe, but is highly likely to increase in fast-developing economies. However, sulfur is not the only acidifying compound of concern; nitrogen deposition, which in addition causes eutrophication, can also cause damage. There are indications that the ability of vegetation to take up nutrients is dependent on acid stress (4).

In this chapter, the effects and risks of sulfur dioxide emissions and resulting deposition are described for soils in general and for forest soils and surface waters in particular. The risk of damage due to acidification is emphasized. It should be noted that allowable exposure to acidification as a basis for deriving guidelines for acceptable levels of sulfur deposition should not be assessed in isolation from nitrogen compounds. The effects of nitrogen compounds and the direct impacts of air concentrations of sulfur dioxide and nitrogen oxides are considered in Chapters 10, 11 and 14.

Routes of exposure of natural systems

Oxidized sulfur can affect terrestrial and aquatic ecosystems. Emissions of sulfur dioxide in the air are transformed to sulfate, which constitutes the major compound of acid deposition. Nitrate is another important component (5).

Indirect effects on terrestrial ecosystems

Damage to forests in Europe, including defoliation, discoloration, growth decrease and tree dieback, have been reported over the last decade. Terrestrial ecosystems can be affected directly owing to the impact on foliage and indirectly via soil acidification.

Soil acidification was originally defined as a decrease in the acid neutralizing capacity (ANC) of the inorganic fraction of the soil, including the solution phase (6,7). ANC is in turn defined as the sum of the basic components (cations) minus the strong acidic components, i.e. the strong acid anions. Soil acidification in this sense is directly dependent on the net supply of base cations (from weathering and deposition) and the net supply of anions (deposition minus retention) in the mineral content. It has since been recognized that accumulation of sulfur (and

nitrogen) compounds in organic matter should also be included, leading to a definition of soil acidification in terms of “a decrease in ANC of the total solid (mineral and organic) and solution phase of the soil” (8). A distinction is thus introduced between actual and potential acidification, i.e. acidification caused by immediate cation removal (actual) and acidification delayed by anion retention (potential).

Soil acidification not only occurs as a consequence of the deposition of acidifying or potentially acidifying compounds. Several other causes can be identified (9,10) including the following:

- decreased deposition of acid-neutralizing compounds;
- increased primary productivity (and/or biomass harvest);
- increased rate of nitrification or sulfur oxidation;
- changes in land use, e.g. afforestation, introduction of acidifying species, and changes in forest management;
- reduced decomposition rate of litter and soil organic matter; and
- increased production and vertical movement of organic acids.

It is generally accepted that soil acidification is caused by the uncoupling of element cycles in relation to the cycling of hydrogen (H^+) (11,12). The most important processes identified by Van Breemen *et al.* (13) and modified by De Vries and Breeuwsma (8) are summarized in Table 1.

Table 1. The relationship between H^+ -producing and H^+ -consuming processes in the soil

H^+ sources	H^+ sinks
Uptake of cations	Uptake of anions
Mineralization of anions	Mineralization of cations
Oxidation reactions	Reduction reactions
Dissociation of weak acids (carbon dioxide, organic acids)	Association of weak acids (carbon dioxide, organic acids)
Weathering, desorption of anions, precipitation, adsorption of cations	Weathering, desorption of cations, precipitation, adsorption of anions

Source: De Vries and Breeuwsma (8).

Insight into the natural and anthropogenic causes of soil acidification is obtained by investigating the acid-producing and acid-consuming processes listed in Table 1 in relation to the complete cycles of elements in the plant–soil system. The management of natural systems, e.g. harvesting practices, may make an important additional contribution to the geochemical balance. A complete overview of cycles is published elsewhere (8, 14).

Deposition of acidifying compounds such as sulfur dioxide, nitrogen oxides and ammonia leads to soil acidification by oxidation to sulfuric and nitric acids and leaching of SO_4^{2-} and NO_3^- respectively, with accompanying cations. Leaching occurs when vegetation does not sufficiently reduce or retain SO_4^{2-} (NO_3^-). The extent to which soil is acidified depends on the H^+ cycle and interacting element cycles, which vary over a broad range in forest soils. It is virtually impossible to account for all sinks and sources of H^+ for all the biogeochemical constellations of

forest soils. To clarify the understanding of soil acidification, Ulrich (15,16) introduced the concepts of buffer capacity and buffer rate. Buffer capacity is the total reservoir of buffering components in the soil. The buffer rate is taken as the actual rate of release of buffering compounds from their pools (e.g. calcium carbonate in calcareous soils).

This general systematic description of acid buffering reactions in soil solution distinguishes four buffer ranges. The first is the carbonate buffer range ($\text{pH} \geq 6$), in which the dissolution rate of calcium carbonate is high enough to buffer most of the existing acid loading rates. The second is the silicate buffer range, in which base cations are produced by silicate weathering. The rate of silicate weathering tends to be low because of the high resistance of silicate rock to decomposition and dissolution. The third range is the cation exchange range, in which exchangeable base cations in the soils are depleted by replacement of magnesium (Mg^{2+}), calcium (Ca^{2+}), sodium (Na^+) and potassium (K^+) by H^+ and aluminium (Al^{3+}). Cation exchange occurs on the surfaces of soil particles, and the total amount of the available exchange surfaces is known as the cation exchange capacity (CEC). The buffer capacity of the cation exchange range is defined as a fraction (i.e. base saturation) of CEC. Since cation exchange reactions are fast, an effective buffering of acidity is obtained. Nevertheless, the buffer capacity may be fairly low, especially in soils with low quantities of base cations. Finally, the fourth buffer range is the aluminium and iron buffer range, where the pH ranges between 3.8 and 4.2. In this range, aluminium is from oxides and hydroxides, which change acids (H^+) to acid cations (Al^{3+}); these are toxic to root systems. Soils with a pH of around 3.8 leach toxic heavy metals and nutrients, inhibiting flora and fauna. The soil processes affecting the rate of soil acidification in all of these various buffer ranges has been described mathematically by the SMART model (14,17).

From the above it becomes clear that the dynamics of forest soil acidification are very site-specific and depend on the soil characteristics such as weathering rate, sulfate adsorption capacity and CEC. However, by ignoring all time-dependent processes and finite pools, the ultimate steady state chemical status of the soils can be calculated (18). Thereafter, by defining the relationship between the chemical status (base cation and aluminium concentrations in the soil solution) and vegetation response, the so called critical load for that particular ecosystem can be derived. This critical load can be interpreted as a guideline for the protection of natural systems against acidification. A similar reduction of the complexity of element cycles can be formulated for the assessment of the quality of surface waters.

Indirect effects on freshwater aquatic ecosystems

The quality of aquatic ecosystems is affected by sulfur compounds deposited directly on the water surface and through the runoff from terrestrial catchment through acidified soils. Acidic deposition has caused acidification of surface waters, dieback of fish populations, alterations of biogeochemical cycles and other ecological changes in large areas of northern Europe and eastern parts of North America. Southern Norway is probably the most severely affected area in Europe with respect to fish mortality. Acidic water has probably been the cause since as far back as the beginning of the twentieth century (19), but it was only at the end of the 1950s that the link to acidic deposition was established (20). The extensive reductions in fish populations in Norway from the 1960s onwards correlate in space and time with the escalation of atmospheric emissions of sulfur and nitrogen compounds in Europe (21). The total land catchment area now affected by acidification damage to fish populations in Norway has been estimated at about 86 000 km² (22).

Studies of Swedish lakes have shown that significant ecological changes, including loss of fish populations attributed to anthropogenic acidification, can be dated back to the 1910s or 1920s (23,24). Today, about 13 700 lakes in Sweden are considered to be significantly acidified by acidic deposition (25). In Finland, the proportion of acidic or poorly buffered lakes is fairly large. While acidic lakes are spread over most of the country, they are concentrated in south-western, central and eastern regions (26). Acidification has caused reductions mainly in roach and perch populations (27–29).

Sulfate is normally a mobile anion in glaciated catchment areas. Increased sulfate concentrations in runoff owing to increased acidifying inputs are accompanied by an increase in base cations and a decrease in bicarbonates, resulting in an acidifying effect on surface waters. The acidification of dilute surface waters with low concentrations of dissolved organic carbon has generally been due to inputs of sulfate anions. Sulfate and nitrate are deposited in a catchment, and acidic surface waters result when the concentrations of these strong acid anions in the runoff exceed the concentrations of base cations, delivered mainly by mineral weathering and cation exchange processes in the soil and by base cation deposition. The efficiency of these processes in counteracting the incoming acidifying deposition determines how much of the mobile strong acid anions are balanced by base cations and how much by hydrogen anions.

Under natural conditions, most of the nitrogen deposited on terrestrial catchment is taken up by vegetation, leaving only low concentrations of ammonia and nitrate in the runoff. In some areas of Europe, however, including Denmark, southernmost Norway and southern Sweden, nitrogen concentrations in runoff water appear to be above background values. In such cases, nitrogen deposition exceeds the rate of nitrogen retention mechanisms, i.e. growth uptake, denitrification and immobilization, thus disrupting the nitrogen cycle. When nitrate becomes mobile and appears in surface waters, it contributes to acidity in the same manner as sulfate.

The long-term dynamics of surface water acidification are determined by soil processes taking place in the terrestrial catchment (described in the previous section) and processes taking place in the water itself. As in the case of forest soils, the end-point steady state status of surface waters can be estimated for a given deposition level by ignoring the time-dependent processes and finite pools. Moreover, as surface water quality integrates the processes over the whole catchment area, the steady state acidity status of surface water can be modelled on the basis of present water quality (30). By assigning critical limits for certain chemical components, which are based on knowledge of the ecological tolerance of sensitive fish species to water chemical conditions (31), critical loads for surface waters can be derived by applying principles of the steady state water quality model (32–34). The ANC, defined as the difference between non-marine base cations and strong acid anions, is used as the chemical criterion for sensitive indicator organisms (usually fish) in surface waters. The computed critical loads can be used to determine suitable air quality guidelines for the acidifying deposition.

Dose–response relationships and critical loads

Acidity

The importance of ANC and the relationship between base cations and aluminium for assessing the quality of surface waters and forest soils, respectively, has been described above. The concept of investigating these parameters as a basis for assessing the risk of damage is at the basis of the concept of critical loads.

The critical load is defined as “the highest deposition of compounds that will not cause chemical changes leading to harmful effects on ecosystem structure and function” (35,36). Critical loads for acidity have been used as a basis for negotiations on the further reduction of European sulfur emissions in the framework of the Convention on Long-Range Transboundary Air Pollution of the United Nations Economic Commission for Europe. In general, the aim of the critical load (for acidity) approach is to achieve protection of ecosystems, e.g. forest soils and surface waters, by sufficiently reducing emissions of acidifying compounds such as sulfur dioxide, nitrogen oxides and ammonia. In fact, the critical load for acidity is a measure for allowable acid deposition, which is indifferent to whether the acidifying stress is caused by sulfur or by nitrogen compounds. However, in the remainder of this section, a critical load of acidity is applied as an indicator for acceptable sulfur deposition only. The effects of nitrogen are treated in Chapters 11 and 14. Methods (not discussed here) have recently been developed to depict combinations of sulfur and nitrogen deposition at which protection against acidification and eutrophication can be achieved (37,38). Thus, the proposal in this section is that sulfur emission reductions are considered sufficient when the resulting sulfur deposition does not exceed critical loads for acidity.

To set a critical load for acidity, an indicator organism is chosen, for which the response to different levels of acidification is known. For each indicator organism, chemical criteria have to be established such that no damage occurs according to current knowledge. In practice, reliable limits have not been established separately for each individual species.

A relationship has been established between increased aluminium concentrations in the soil solution and adverse effects to roots and growth (39–45). For example, it has been shown that the root length of Norway spruce decreases when the base cation (i.e. calcium) to aluminium (BC:Al) ratio is smaller than 1 (46,47). Other limits for forest soils are the aluminium concentration in soil solution and the pH.

An extensive overview of both laboratory and field experiments with respect to the relationship between BC:Al ratios and damage to trees and ground vegetation can be found in Sverdrup & Warvfinge (44). Laboratory results of aluminium damage indicate that tolerance to this element varies among tree species. For example, a growth reduction of 80% has been demonstrated at a BC:Al ratio of 0.1 for the northern white cedar (*Thuja occidentalis*) and of 4 for the masson pine (*Pinus massonii*). Sverdrup & Warvfinge (44) found that, as a general rule, a BC:Al ratio ≥ 1 seems to provide appropriate sustainability for European forests. However, species that grow in aluminium-oxide-rich, non-glaciated, old soils, such as teak, guapira, orange and cotton, seem to be more accustomed to aluminium than trees from the temperate zone. Thus computation of critical loads in Europe (37,38,48–50) have generally applied a BC:Al ratio of 1, whereas preliminary critical loads computed in Asia have used varying BC:Al ratios (51). Table 2 gives an overview of average limits that have been established for computing critical loads (52,53).

For surface waters, ANC has been considered as a chemical criterion that can be used to explain the increasing risk for fish damage. For critical load calculations, the critical chemical value, termed the ANC limit, has been derived from information on water chemistry and fish status obtained from the 1000 lake survey carried out in Norway in 1986 (30,31). The Scandinavian countries have decided to use an ANC limit of 20 $\mu\text{eq/litre}$ as the critical chemical value for fish in surface waters, the value being well justified by the Norwegian survey (32).

Table 2. Acceptable limits for chemical compounds in forest soils and freshwater systems

Compound	Unit	Forest soil	Fresh water	Groundwater
Aluminium	mol _e /m ³	0.2	0.003	0.02
BC:Al ratio	mol/mol	1	–	–
pH	–	4.0 ^a	(5.3, 6.0) ^b	6.0
ANC	mol _e /m ³	–	(0.02, 0.08) ^b	0.14
Nitrate	mol _e /m ³	–	–	0.8

^aAssuming log K_{gibb} of 8.0 and $[Al] = 0.2 \text{ mol}_e/m^3$.

^bA pH of 6.0 relates to peak flow situations and is associated with an acid neutralizing capacity of 0.08 mol_e/m³.

The selected ANC limit has been assessed by examining the relationship between the critical load excess and the damage to fish, again using data from the Norwegian 1000-lake survey. The probability of damage to fish populations increases clearly as a function of the critical load excess (54). When the critical load is not exceeded, there is only a small probability of damage to fish, but as soon as the critical load is exceeded, the probability of fish damage starts to increase.

The calculation of critical loads is based on the steady state mass balance (SSMB) method, which assumes a time-independent steady state of chemical interaction involving an equilibrium between the production and the consumption of acidic compounds (37,52). The ratio of base cations to aluminium and the aluminium concentration are important indicators that reflect whether the ecosystem is protected or not within an infinite time span. By assigning critical chemical values for various indicators, it is possible to compute the allowable acidification for each ecosystem. Critical loads for acidity and sulfur have been calculated and mapped in Europe (37) for forest soils and surface waters and in Asia for 14 vegetation–soil combinations (51).

It should be noted that the critical load is an indicator for long-term sustainability. Research on the relationship between current excess of critical loads for acidity on broad regional scales and actual damage is required to further develop the applicability of the critical load as a guideline.

Guidelines

In Europe, critical loads have been established at the resolution of EMEP¹ to allow for comparisons between critical loads and (sulfur) deposition values and to identify areas where critical loads are exceeded. Critical loads of acidity, as calculated by the SSMB method, are predominantly dependent on the rate of base cation weathering. For terrestrial ecosystems, the weathering rate can be estimated by combining information on soil parent material and texture properties. Table 3 lists the ranges of critical loads in relation to combinations of parent material and texture classes.

¹ Cooperative Programme for the Monitoring and Evaluation of Long Range Air Pollutants in Europe.

Table 3. Critical load ranges used for the various combinations of parent material and texture classes in terrestrial ecosystems

Guideline range of critical loads (eq/ha per year)	Parent material	Texture class ^a
< 250	Acidic ^b	Coarse
250–500	Acidic	Coarse-medium
	Intermediate ^c	Coarse
	Basic ^d	Coarse
	Acidic	Medium, medium-fine
500–1000	Intermediate	Coarse-medium, medium
	Basic	Coarse-medium
	Intermediate	Medium-fine
1000–1500	Basic	Medium
	Intermediate	Fine
> 1500	Basic	Medium-fine

^a Coarse – clay content < 18%; medium – clay content 18–35%; fine – clay content >35%.

^b Sand(stone), gravel, granite, quartzine, gneiss.

^c Gronodiorite, loess, fluvial and marine sediment.

^d Gabbro, basalt, dolomite, volcanic deposits.

Additional factors, such as vegetation cover, further modify the value of the critical load. To calculate precise critical loads for a given geographical area it is recommended that the mass balance equation be used.

For surface waters, the weathering rate can be estimated on the basis of water quality and quantity variables, of which base cation concentrations and runoff are the most influential. Table 4 lists the ranges of critical loads in relation to combinations of base cation concentration and runoff classes. For each critical load class, at least 50% of the critical load values computed on the basis of lake data from Finland, Norway and Sweden fall within the class boundaries, given the ranges for present base cation concentrations and runoff. In just two cases, the boundaries for the base cation concentration classes overlap two critical load classes, when the class boundaries were set on the basis of the 25th and 75th percentile base cation concentrations for given runoff classes. For these cases the critical loads are determined more by other factors than base cation levels and runoff; the guideline values set are therefore more uncertain than those without overlap.

An assessment of the cumulative distribution of critical load data available for terrestrial ecosystems and surface waters in Europe, as collected in the framework of the UN/ECE mapping programme, shows the following: about 1% of the natural systems have critical loads lower than 250 eq/ha per year (at a median of 170 eq/ha per year); the range of 250–500 eq/ha per year covers about 3% (at a median of 414 eq/ha per year); and about 11% of the area has critical loads of 500–1000 eq/ha per year (at a median of 821 eq/ha per year). Thus about 15% of terrestrial and surface water ecosystems have critical loads for sulfur lower than 821 eq/ha per year.

Table 4. Critical load ranges used for various combinations of base cation concentration and runoff for surface waters

Guideline range of critical loads (eq/ha per year)	Base cation concentration (meq/m ³)	Runoff (m)
< 250	< 45	> 1
	< 100	0.3–1
	< 270 ^a	< 0.3
250–500	45–70	> 1
	100–190	0.3–1
	250–400 ^a	< 0.3
500–1000	70–103	> 1
	190–290	0.3–1
	400–650	< 0.3
1000–1500	103–170	> 1
	290–465 ^a	0.3–1
	650–1300	< 0.3
> 1500	> 170	> 1
	> 350 ^a	0.3–1
	> 1300	< 0.3

^aThe class boundaries overlap.

The median critical load of acidity of each of the ranges, i.e. 0–250, 250–500, 500–1000, 1000–1500 and higher, is proposed as the representative value for these ranges. Table 5 lists the median values for each range, including the related protection percentage of the total European natural area.

Table 5. Median values of critical load ranges for terrestrial and surface water

Guideline range of critical loads (eq/ha per year)	Median (eq/ha per year)	Ecosystem area protected in Europe (%)
< 250	170.0	99.2
250–500	414.0	96.2
500–1000	821.0	85.3
1000–1500	1269.0	73.7
> 1500	2295.0	29.9

References

1. FINKEL, R.C. ET AL. Changes in precipitation chemistry at Dye 3, Greenland. *Journal of geophysics research*, **910**: 9849–9855 (1986).

2. TUOVINEN, J.-P. ET AL. *Transboundary acidifying pollution in Europe: calculated fields and budgets 1985–93*. Oslo, Norske Meteorologiske Institutt, 1994 (Technical Report No. 129, EMEP/MSC-W Report 1/94).
3. BARRETT, M. Environmental impacts of fossil fuels. In: Ramani, K.V. et al. *Burning questions: environmental limits to energy growth in Asian Pacific Countries during the 1990s*. Kuala Lumpur, Asian and Pacific Development Centre, 1993.
4. BRYDGES, T. & SUMMERS, P.W. The acidifying potential of atmospheric deposition in Canada. *Water, air and soil pollution*, **43**: 249–263 (1989).
5. BOBBINK, R. & ROELOFS, J.G.M. Nitrogen critical loads for natural and semi-natural ecosystems: the empirical approach. *Water, air and soil pollution*, **85**: 2413–2418 (1995).
6. VAN BREEMEN, N. ET AL. Acidification and alkalization of soils. *Plant and soil*, **75**: 283–308 (1983).
7. VAN BREEMEN, N. ET AL. Acidic deposition and internal proton sources in acidification of soils and waters. *Nature*, **307**: 599–604 (1984).
8. DE VRIES, W. & BREEUWSMA, A. The relation between soil acidification and element cycling. *Water, air and soil pollution*, **35**: 293–310 (1987).
9. BERDÉN, M. ET AL. *Soil acidification, extent, causes and consequences. An evaluation of literature information and current research*. Stockholm, National Swedish Environmental Protection Board, 1985 (Report 3292).
10. KAUPPI, P. ET AL. Modeling soil acidification in Europe. In: Alcamo, J. et al., ed. *The RAINS model of acidification*. Dordrecht, Kluwer Academic Publishers, 1990.
11. ULRICH, B. & PANKRATH, J.L., ED. *Effects of accumulation of air pollutants in forest ecosystems*. Dordrecht, D. Reidel Publishing, 1983.
12. ULRICH, B. & MATZNER, E. *Abiotische Folgewirkungen der weiträumigen Ausbreitung von Luft Verunreinigungen*, Bonn, Bundesminister d. Innern., 1983 (Forschungsbericht 10402615).
13. VAN BREEMEN, N. ET AL. *Reprints of publications on the project “Effects of acid atmospheric deposition on soils and water”*. Wageningen, Agricultural University, 1985 (Project No. J 050-792, 1982–1985).
14. DE VRIES, W. *Soil response to acid deposition at different regional scales: field and laboratory data, critical loads and model predictions*. Dissertation, Agricultural University, Wageningen, 1994.
15. ULRICH, B. & SUMNER, M.E., ED. *Soil acidity*. Berlin, Springer, 1991.
16. ULRICH, B. Soil acidity and its relation to acid deposition. In: Ulrich, B. & Pankrath, J., ed. *Effects of accumulation of air pollutants in forest ecosystems*. Dordrecht, D. Reidel Publishing, 1983.
17. DE VRIES, W. ET AL. Simulation of the long-term response to acid deposition in various buffer ranges. *Water, air and soil pollution*, **48**: 249–390 (1989).
18. DE VRIES, W. ET AL. Impacts of acid deposition on concentrations and fluxes of solutes in acid sandy forest soils in the Netherlands. *Geoder*, **67**: 17–43 (1995)
19. HUITFELDT-KAAS, H. Om aarsaken til massedød av laks og ørret i Frafjordelven, Helleelven og Dirdalselven i Ryfylke høsten 1920 [On the cause of mass death of salmon and brown trout in Frafjordelven, Helleelven and Dirdalselven in Ryfylke harvest 1920]. *Ambio*, **20**: 23–27 (1991) (in Norwegian).
20. DANNEVIG, A. Nedbørens innflytelse på vassdragenes surhet og på fiskebestanden [Influence of precipitation on river acidity and fish population]. *Jæger og fisker*, **3**: 116–118 (1959) (in Norwegian).

21. OVERREIN, L.N. ET AL. *Acid precipitation – effects on forest and fish. Final report of the SNSF project 1972–1980*. Oslo, Norwegian Institute for Water Research, 1981 (SNSF-project FR 19/80).
22. HESTHAGEN, T. ET AL. *Mapping of fish community status in relation to acidification in Norway. The Norwegian Critical Load Project (Naturens Tålegrenser)*. Trondheim, Norwegian Institute for Nature Research, 1992 (Report No .8) (in Norwegian with English summary).
23. ALMER, B. ET AL. Effects of acidification on Swedish lakes. *Ambio*, **3**: 30–36 (1974).
24. RENBERG, I. & BATTARBEE, R.W. The SWAP Paleolimnology Programme: a synthesis. In: Mason, B.J., ed. *The Surface Waters Acidification Programme*. Cambridge, Cambridge University Press, 1991, pp. 281–301.
25. BERNES, C. *Acidification and liming of Swedish freshwaters. Monitor 12*. Stockholm, Swedish Environmental Protection Agency, 1991.
26. KÄMÄRI, J. ET AL. Finnish lake survey: present status of acidification. *Ambio*, **20**: 23–27 (1991).
27. RASK, M. ET AL. Losses and recoveries of fish populations in acidified lakes of Southern Finland in the last decade. *Water, air and soil pollution*, **85**: 315–320 (1995).
28. RASK, M. Effects of acid deposition on fish populations in small lakes of southern Finland. In: Perry, R. et al., ed. *Acid rain: scientific and technical advances*. London, Selver Ltd, 1987, pp. 528–532.
29. LAPPALAINEN A. ET AL. Acidification affects the perch (*Perca fluviatilis*) populations in small lakes of southern Finland. *Environmental biology of fish*, **21**: 231–239(1988).
30. HENRIKSEN, A. ET AL. Lake acidification in Norway: present and predicted chemical status. *Ambio*, **17**: 259–266 (1988).
31. HENRIKSEN, A. ET AL. Lake acidification in Norway: present and predicted fish status. *Ambio*, **18**: 314–321 (1989).
32. HENRIKSEN, A. ET AL. *Critical loads to surface waters in Fennoscandia*. Copenhagen, Nordic Council of Ministers, 1990 (Miljørapport 1990: 124).
33. KÄMÄRI, J. ET AL. Critical loads of sulfur and nitrogen for lakes. II. Regional extent and variability in Finland. *Water, air and soil pollution*, **66**: 77–96 (1993).
34. POSCH, M. ET AL. Critical loads of sulfur and nitrogen for lakes. I. Model description and estimation of uncertainty. *Water, air and soil pollution*, **66**: 173–192 (1993).
35. NILSSON, J., ED. *Critical loads for sulphur and nitrogen: report from a Nordic working group*. Copenhagen, Nordic Council of Ministers, 1986.
36. NILSSON, J. & GRENNFELT, P., ED. *Critical loads for sulfur and nitrogen. Report from a workshop held at Skokloster, 19–24 March 1988*. Copenhagen, Nordic Council of Ministers, 1988 (Miljørapport 1988: 15).
37. HETTELINGH, J.-P. ET AL. The use of critical loads in emission reduction agreements in Europe. *Water, air and soil pollution*, **85**: 2381–2388 (1995).
38. POSCH, M. ET AL., ED. *Calculation and mapping of critical thresholds in Europe*. Bilthoven, National Institute of Public Health and the Environment, 1995.
39. ABRAHAMSEN, G. Effects of acidic deposition on forest soil and vegetation. *Philosophical transactions of the Royal Society, London, B*, **305**: 369–382 (1984).
40. ASHER, C. J. Effects of nutrient concentration in the rhizosphere on plant growth. In: Hintze, D., ed. *Transactions of the Twelfth Congress of the International Society of Soil Science*. Wageningen, International Society of Soil Science, 1987, pp. 209–216.
41. BERDÉN, M. ET AL. *Soil acidification, extent, causes and consequences*. Stockholm, National Swedish Environmental Protection Board, 1985 (Report 3292).

42. BOSSEL, H. ET AL. *Dynamik des Waldsterbens: mathematisches Modell und Computersimulation* [The dynamics of forest death: mathematical model and computer simulation]. Berlin, Springer Verlag, 1985.
43. RYAN, P.J. *The role of acid and aluminium-rich media in the growth and nutrition of Pacific Northwest conifers*. Seattle, University of Washington, 1983.
44. SVERDRUP, H. & WARFVINGE, P. *The effect of soil acidification on the growth of trees, grass and herbs as expressed by the (Ca+Mg+K)/Al ratio*. Lund, Lund University, 1993 (Report 2: 1993).
45. WARFVINGE, P. ET AL. Modeling long-term base supply to acidified forest stands. *Environmental pollution*, **80**: 1–13 (1993).
46. TOMLINSON II, G.H. Die back of red spruce, acid deposition and changes in soil nutrient status – a review. In: Ulrich, B. & Pankrath, J.L., ed. *Effects of accumulation of air pollutants in forest ecosystems*. Dordrecht, D. Reidel Publishing, 1983.
47. SCHULZE, E.D. Tree responses to acid deposition into the soil. A summary of the COST workshop at Juelich. In: Mathy, P., ed. *Air pollution and ecosystems*. Dordrecht, D. Reidel Publishing, 1987, pp. 225–241.
48. HETTELINGH, J.-P. ET AL. *Mapping critical loads for Europe*. Bilthoven, National Institute of Public Health and Environmental Protection, 1991.
49. HETTELINGH, J.-P. ET AL. The use of critical loads in emission reduction agreements in Europe. *Water, air and soil pollution*, **85**: 2381–2388 (1995).
50. DOWNING, R.J. ET AL. *Calculation and mapping of critical loads in Europe. Status report 1993*. Bilthoven, National Institute of Public Health and Environmental Protection, 1993.
51. HETTELINGH, J.-P. ET AL. Deriving critical loads in Asia. *Water, air and soil pollution*, **85**: 2565–2570 (1995).
52. SVERDRUP, H. ET AL. *Mapping critical loads: a guidance to the criteria, calculations, data collection and mapping of critical loads*. Copenhagen, Nordic Council of Ministers, 1990 (Report Nord 1990: 98).
53. DE VRIES, W. *Methodologies for the assessment and mapping of critical loads and impacts of abatement strategies on forest soils*. Wageningen, Staring Centre, 1991 (Report 46).
54. HENRIKSEN, A. & HESTHAGEN, T. *Critical load exceedance and damage to fish populations*. Oslo, Norwegian Institute for Water Research, 1993 (Project Naturens Tålegrenser, Fagrapport No. 43).