New concepts and methods for effect-based strategies on transboundary air pollution

Synthesis Report, April 2002

ASTA
The Mistra Programme:
International and National Abatement Strategies
for Transboundary air Pollution

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Preface

This report was prepared in connection with the scientific evaluation of the ASTA programme in May, 2002. It consists of a comprehensive summary of the approaches and results from the different ASTA sub programmes. ASTA is presently in its fourth and last year of the first phase. The report is not intended to give a full description of all relevant aspects of the problem of transboundary air pollution but rather discuss some crucial problems and their possible scientific solutions.

After the preparation and submission of this report, ASTA has been positively evaluated and will continue into its second phase. The evaluation reports as well as a letter of intent for the second phase of the ASTA programme are available on the ASTA web page (http://asta.ivl.se). Further information of the programme is also available at the web page. For those who wish to receive more information of the programme, there is a list of contact persons at the end of the report.

Many of the ASTA phase 1 activities are currently in a state of intense evaluation and reporting and additional scientific results as well as synthesises and assessments will be prepared during the remainder of 2002.

Göteborg 16 August 2002

Peringe Grennfelt	John Munthe
Programme Director	Deputy director, main editor of the report

The ASTA-programme 1999-2002

ASTA is a 4-year research programme focussed on transboundary air pollution. The overall aim is to develop scientifically based support to international agreements on reductions of transboundary air pollution in Europe.

The ASTA programme includes experimental and modelling work on acidification and recovery of soils and surface waters, impact of surface ozone on crops and forest trees, nitrogen impact on terrestrial ecosystems and sources and transformation of atmospheric particles. The ASTA programme also includes specific social science studies of the process of developing science based policy. National issues are the focus of a specific project where interactions between land-use and transboundary air pollution are investigated.

A schematic structure of the ASTA programme components is presented in Figure 0-1.

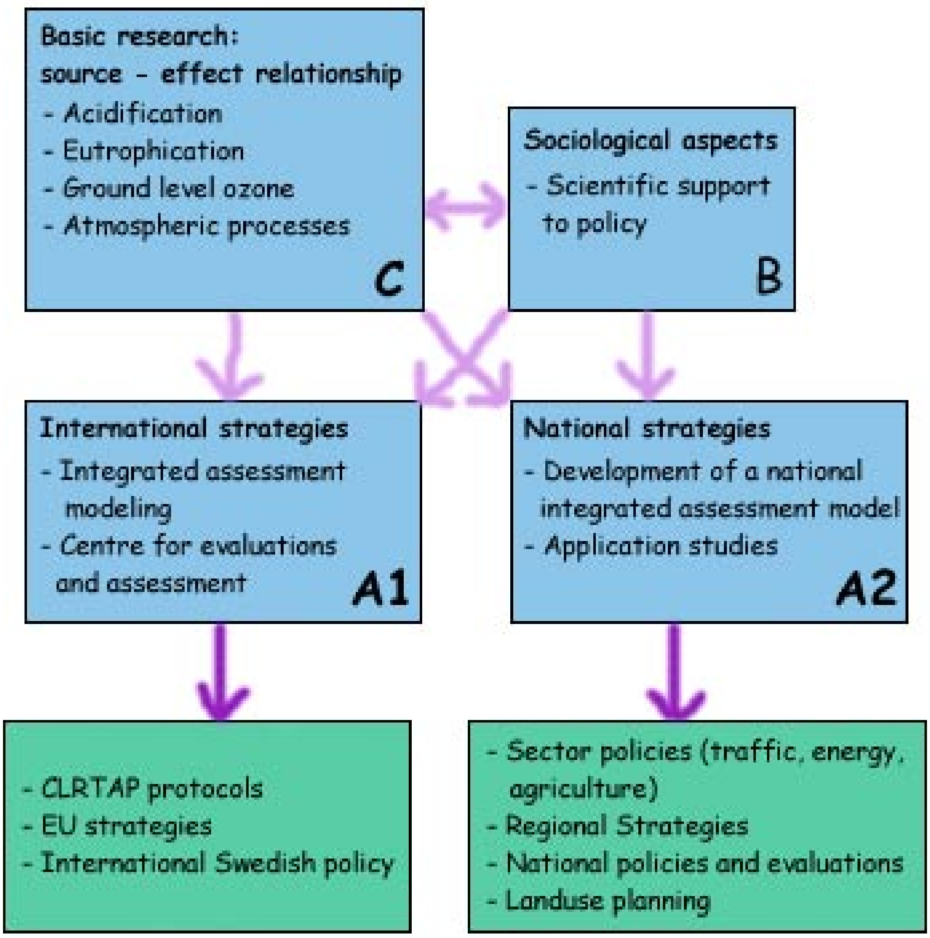

Figure 0-1 Structure of the ASTA programme.

More information is available on the ASTA website: http://asta.ivl.se

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1 Introduction

The UN ECE Convention on Long-Range Transboundary Air Pollution (CLRTAP) was established in 1979 for the control of regional air pollution problems in Europe. Under the Convention several protocols have been signed, most recently the so-called Gothenburg Protocol directed towards sulphur dioxide, nitrogen oxides, volatile organic compounds and ammonia in one joint strategy (Table 1.1). The purpose of the protocol is to control several environmental problems; acidification, eutrophication, effects of ozone to vegetation and human health and include national emission control requests for all signing countries. The protocol was based on the assumption that a certain environmental targets should be reached by the least cost.

Table 1.1 Emission reductions in comparison to 1990 levels in Europe according to the Gothenburg Protocol (UN ECE 1999).

Pollutant	Emission reduction (%)
SO_2	63
NO_x	41
VOC	40
NH_3	17

Scientific research has played an important role for the development of abatement strategies all the time from the discovery of the regional air pollution problems (in particular acidification) until today. There are several examples on how scientific assessments have influenced the policy process. The first international assessment of significance was probably Sweden's Case study for the UN Conference on the human environment in Stockholm in 1972. In this study the environmental problems related to the emissions of sulphur dioxide were compiled and quantified. The future development was studied for Europe as well as for Sweden in terms of three scenarios. The European scenarios included one scenario which could be characterised as business as usual, one as a levelling out of the emissions in 1965 and one assuming a 50% reduction between 1965 and 1982 and after that constant yearly emissions. The control scenario also included estimates on abatement costs; all together the report contained an overall approach very similar to today's integrated assessment models.

Another early example on how science interacted with the policy process is the OECD study on the transboundary transport of sulphur over Europe. The study took place between 1971 and 1975 (OECD, 1977). In this study, emissions, atmospheric transport and deposition were studied by means of measurements and model calculations. The study included both policy discussions and actions but also intense scientific research. Other early examples are the production of background documents for a number of policy conferences on transboundary air pollution and acid deposition, e.g. the conference in Stockholm in 1982 and the MP conference organised by the Nordic Council of ministers in 1987 (Nordic Council of Ministers, 1986). The Norwegian project on the effects from acid deposition to forests and lakes

(SNSF Project) is probably the best example on a scientific research programme exclusively directed towards the development of policies (Overrein et al. 1980).

From the discussion above, it is obvious that Sweden and Scandinavia already from the early days of acid rain and transboundary air pollution took a strong international position, and contributed to both science and policy. There are probably several reasons for that; the discovery of the problem was to a large extent made by Swedish scientists; in particular by Svante Odén but important contributions were also made by other scientists such as Cyrill Brosset and Torsten Ahl. A second reason was the magnitude of the problem as it was observed in Swedish lakes and a third the obvious geographical location of Sweden, downwind the large European source areas.

Early results showed Scandinavia's vulnerable location downwind of the large industrial areas in Europe (OECD 1977). Other studies showed that the sensitive ecosystems in Sweden and Norway made the problems more pronounced and easier to detect than elsewhere in Europe (Odén 1968; Hultberg and Stensson 1970). The disappearance of fish in lakes and streams were signs that could not be neglected by anyone, even if many argued that acidification and fish disappearance could be explained by other causes. Acidification was for a long time considered being the most important environmental problem in Sweden and Norway and received large attention in both political and scientific arenas.

Today, acidification and other transboundary air pollution problems are not receiving the same attention as 10-20 years ago. The political arena is taken over by other problems, in particular climate change and chemicals. One obvious reason is that emissions have decreased and are expected to decrease even further up to 2010. Another reason is that there is a much higher degree of consensus about causes and effects, which tends to decrease at least medial interest for the issue.

Science and policy have been closely linked throughout the whole history of transboundary air pollution. Researchers as well as politicians consider the work within the LRTAP Convention an exemplary form of co-operation between science and policy. The science-based character is usually viewed as an explanation of the success of the protocols, in which scientifically founded environment standards rather than arbitrarily chosen proposals are the principle of regulation. A schematic sketch of the links between science and policy is presented in Figure 1-1.

But this success could also be a problem. A too close co-operation between researchers and policy makers could lead to a narrow regulative process, where transparency and stakeholders' involvement are vanishing, as well as the interest of the general public. Recently the European Commission has launched a white paper on governance highlighting the importance of democratising expertise in order to strengthen the legitimacy of the policy process. A technocratic process is considered the enemy, in which an expert elite carries out the work in an opaque way difficult for outsiders to follow and understand, i.e. inaccessible to public scrutiny. Therefore, the necessary tension between the co-operation of research and policy on one hand, and transparency and stakeholder involvement on the other, should be given increased attention in the efforts to further develop scientifically credible as well as politically legitimated abatement strategies in the field of transboundary air pollution.

Within the area of health effects, air pollution still is of large importance to both science and policy. Several international studies show that air pollution contributes significantly to human health and mortality. Fine particles are considered to be a main factor for the observed effects and reduction of atmospheric particle concentrations is today perhaps the primary focus in air pollution policy. Since the fine particle fraction to a large extent is linked to the same pollutants as those considered in the present air pollution strategies (sulphur dioxide, nitrogen oxides, ammonia and VOC), the policy work on transboundary air pollution has adopted health effects as an important topic for future strategies.

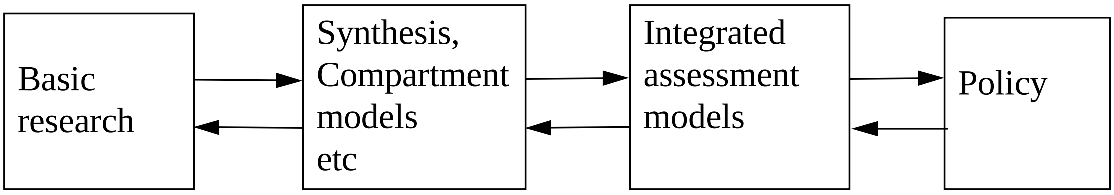


Figure 1-1 The links between basic scientific research and policy in the present strategies of transboundary air pollution.

1.1 The role of ASTA. The objective of the report.

The ASTA research programme was outlined in 1998 and the decision for support was taken by the Mistra foundation in October 1998. At that time, agreements on substantial reductions of emissions of sulphur dioxide, nitrogen oxides, VOC and ammonia were expected to be reached within a year or two. It was also clear that these reductions would not be sufficient to reach long term objectives for the protection of human health, welfare and ecosystems in Europe. A new round of negotiations was expected to take place some years after 2000 and ASTA was formed as a scientific research programme to support the development of these new policies. A number of specific research areas were chosen primarily based on expected needs for improvements in the coming strategies and competence in Sweden.

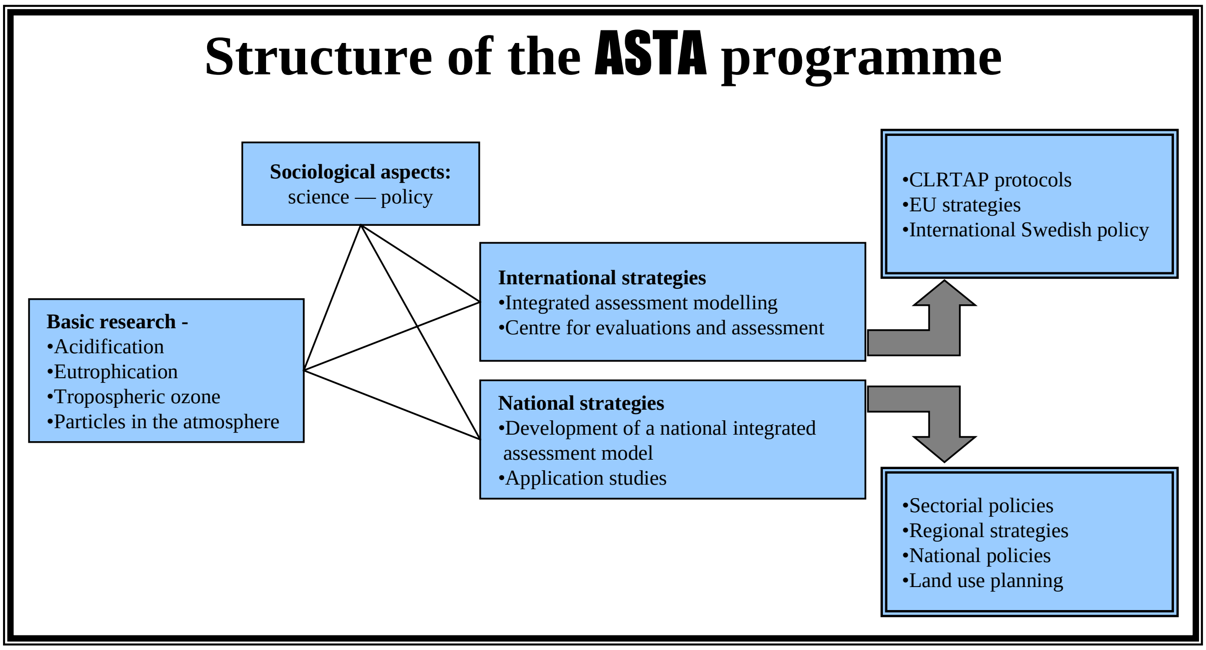


Figure 1-2 Schematic structure of the ASTA programme.

During the planning of the ASTA programme, it became obvious that national issues on energy, transport and environment also were crucially dependent on the same type of background material as generated for the international negotiations. A national research programme was then formed directed primarily towards relations between air pollution and forest land use. A schematic diagram of the different program components and their links with policy makers is presented in Figure 1-2.

The ASTA programme is now at the end of its first phase comprising the period 1999-2002 and many results have been achieved in relation to the objectives originally outlined in the proposal. Before entering the second phase, we have found it important to summarise and communicate the results in the perspective of their relation to integrated knowledge and policy.

Since the results are finally aimed to support the CLRTAP and EU work on transboundary air pollution, the report has taken its starting-point in what we feel are the main needs of scientific support. Much of the research is still in progress and will be further refined and communicated during the next 1-2 years.

2 New approaches

2.1 Third generation strategies – approaching critical loads

The negotiations on further reductions in air pollution emissions after the Gothenburg Protocol and the National Emissions Ceilings Directive are primarily directed towards control measures with target years 2015 or even 2020. The new negotiations will also direct measures towards environmental targets that are reaching or getting very close to critical loads and levels. The NEC directive has outlined a request that critical loads should be reached in all (EU-)Europe at this time.

Critical loads was defined from a perspective of environmental effects. International strategies have so far been directed towards minimising or avoiding these effects in European ecosystems. Other effects-based approaches are also possible and transboundary air pollution control strategies can also be considered in the context of sustainable development. In such an approach environmental measures should be directed towards levels where human health and ecosystems will not be threatened, but also to levels which allow a sustainable exploitation of ecosystems without deterioration for coming generations. In such a strategy, ecosystem stability becomes more important and also the recovery of ecosystems damaged by air pollution. Recovery processes, their relation to the critical loads concept and abatement strategies were not considered in the development of air pollution strategies; neither for the Gothenburg Protocol nor for the National Emissions Ceilings directive. In these, it was more or less understood that reaching critical loads would be enough for the recovery of damage ecosystems. The picture is, however, not that simple. The definition of critical loads implies that the system will be balanced when deposition is at the critical loads; i.e. damaged ecosystems will not change, neither towards further deterioration nor towards recovery. But there is of course in any air pollution strategy an expectation that acidified lakes should recover and sustain ecosystems of the same kind as before the acidification; and that eutrophied ecosystems damaged by nitrogen deposition should recover and become pristine. The time for recovery will be dependent on several factors, but it is obvious that it will depend on how far the actual load is below the critical load. A deposition far below the critical loads will reduce the time for recovery compared to if deposition is just below the critical loads.

When the critical loads concept was developed actual levels and loads were far above the critical loads in large parts of Europe and the measures to be taken were only limited steps of the overall needs. The deposition and concentration levels had for all-important trace constituents been more or less constant for at least a decade. In this situation it was important to achieve a notable change in the situation and the critical loads concept gave a clear starting-point for this.

In the development of abatement strategies for the Gothenburg Protocol and the NEC directive, variability in factors such as land-use and climate were not taken into account. It is however obvious that there are a number of anthropogenic and natural circumstances that may change over time and also affect the critical loads concepts and the strategies based on the concepts.

Land-use is always changing and critical loads made very simple assumptions with respect to land use. Several European policies indicate today an increased use of biological fuels for stationary as well as mobile sources. Such policies may also be of importance for acidification and nutrient balances of forest soils. Present critical loads concepts do not take into account an increased extraction of biomass.

Another factor is the climate change expected to take place due to increased greenhouse gas concentrations. Factors to be taken into account may be increased concentrations of carbon dioxide, increased temperature and changes in precipitation. ASTA has far not considered the relations between the effects from transboundary air pollution and climate.

2.2 Dynamics in environmental effects and their relations to abatement strategies

2.2.1 Acidification

Present IAM models (e.g. RAINS) rely completely on results of static calculations of critical loads as performed by individual states. Acid deposition has been reduced substantially in Europe over the last decade and signs of recovery have been noticed. Evaluating the time lag between deposition reduction and ecosystem recovery has been the focus of a number of national and international research activities during the last 10 years. One general conclusion of these activities is that recovery of damaged ecosystems is a slow process and that further reductions in deposition will increase the rate of recovery. In high sensitivity regions, the accumulated impact of acidification have caused damages, which may take centuries to recover and are in practice irreversible. These findings present a picture of the acidification status in Europe that greatly differs from e.g. maps of critical load exceedances. It clearly points to the need for further efforts to reduce acid deposition and that recovery time should be taken into account in the process of emission reduction optimisation.

These findings also point out the necessity of changing the basis for control strategies, i.e. to include dynamic aspects and results from dynamic modelling in the critical loads concepts. The complexity of the currently available dynamic models for acidification and recovery, however, precludes a direct coupling to integrated assessment models such as RAINS. The question of dynamic modelling and how to couple the dynamic recovery models to the basically static RAINS has been the focus of a large part of the ASTA programme. ASTA has played a central role in the on-going work to transfer the knowledge gained from experiments and advanced models on recovery into models that can be coupled to integrated assessment models.

2.2.2 Nitrogen

2.2.2.1 N deposition and effects to biodiversity

Biodiversity loss is an important and often overlooked consequence of regional air pollution although it is ranked as an important environmental issue within Europe. Nitrogen deposition has had a devastating effect in many nutrient-poor ecosystems and the importance of acid deposition to biodiversity loss in lakes and surface waters is well recognised. Even if there are clear signs of biodiversity changes there is still very little known on the mechanisms and dynamics on biodiversity change caused by anthropogenic stresses.

The underlying science for establishing critical loads and parameters for dynamic assessments of physiological and ecological responses to increased (and decreased) nitrogen input is currently not satisfactory. Experimental studies forming the basis for methods to calculate critical loads for nitrogen with respect to biodiversity have often been performed in areas which have received high nitrogen input for decades. Thus, they describe changes from a state already affected by nitrogen induced vegetation changes to a state of even higher impact.

Biodiversity loss is also linked to many other environmental problems, in particular climate change. Climate change, acidifying compounds and nutrient nitrogen are expected to have individually significant effects on the biodiversity but none of them can be considered in absence of the others. In model approaches, there is thus a need to develop models that may include all parameters in parallel. State of the art models available for evaluating some of these effects are considering each pollutant effect in isolation for a very limited repertoire of plants, mainly trees. An integrated assessment tool is yet missing for (1) the total combined impact of climate change, acidification and eutrophication on biodiversity in a regional sense, and (2) the degree and importance of individual causes (most severe problem) and the importance of feedback between climate change, acidification and eutrophication on the final effect on biodiversity.

An operational framework is needed for analysing terrestrial ecological systems from local to national scales and, in so doing, aims to define the role GIS and environmental modelling have to play in the conservation of biodiversity as reported by the European Environmental Agency (EEA). ASTA has recognised the problem and activities are started in order to develop models for predicting pollution impacts on biodiversity components not only for large trees in production, but for multiple components of the ecosystems. These models are aimed to be at hand during phase two of the ASTA programme.

2.2.2.2 Recovery

The problems of recovery of ecosystems damaged by eutrophication due to large inputs of nitrogen are similar to those for acidification. The dynamic aspects in N effects to ecosystems were recognised already when the critical loads for nitrogen were developed (Nilsson and Grennfelt 1988). At that time many ecosystems received atmospheric deposition far above the critical loads without showing (at least based on the knowledge at that time) any signs of damage and there was an interest to assess the time-scales for the development of observable ecosystem changes.

Recovery of forest ecosystems from N deposition effects has been studied both in roof experiments and by halted additions of N to formerly fertilised systems. These studies convincingly show that soil properties such as levels of NO_3^- and NH_4^+ in the soil solution, rates of mineralisation and leakage of NO_3^- rapidly changes when the N load decreases (cf Quist et al. 2000). Also levels of N in leaves or needles and in the below ground tissues decrease rapidly following cessation of N additions. Thus, the majority of results from studies using roofs to decrease N loads and from studies with halted N additions points to a rapid recovery of forest ecosystems, at least with respect to chemical and physiological responses on a plant level.

Within ASTA these more general results and assumptions are challenged by studying N transformations in forest ecosystems. Because N added to ecosystems may be immobilised to a great extent in the soil, there is a potential for long-lasting effects on the biota. Furthermore, it seems probable from experimental studies of N fertilisation that N effects may remain some half-century after cessation. A slow recovery may include both a long lasting effect on N turnover in the soil-plant system and slow replacement of the N favoured vegetation by the original vegetation. Both these factors are important to consider in the development of concepts and methods for assessing the dynamic effects in control strategies for nitrogen-induced effects to ecosystems.

The research within ASTA has focused on nitrogen transformations and levels in N fertilised forest ecosystems some years after fertilisation was terminated. (Strengbom et al. 2001).

2.2.3 Level II ozone

In the last few years important new, dynamic concepts have been introduced in the field of ozone effects on plants. They are presently in a process of intensive development and are likely to be finalised around 2005.

The first generation of critical levels for ozone, identified at a workshop in Bad Harzburg, Germany in 1988, were based on traditional concepts in ecotoxicology where average concentrations for different integration periods were used. The second generation was more sophisticated in that it was based on the accumulated exceedance of a certain cut-off concentration. AOT40 – the accumulated exposure over a concentration threshold of 40 ppb ozone – was the main indicator used at this stage. This process was initiated at a workshop in Egham, UK in 1992. Further steps on this path were taken at similar meetings in Bern, Switzerland in 1993 and in Kuopio, Finland in 1996 (Kärenlampi and Skärby, 1996). AOT40 is the main exposure index for ozone effects on plants in the background documents for the Gothenburg protocol and in the EU Ozone directive. It was also used by WHO for assessment of ecotoxic effects of ozone.

AOT40 is calculated for the daylight hours of the day. This represents a first adaptation to the fact that the ozone uptake by the plants occurs mainly through the stomata. Ozone uptake thus depends on the ozone concentrations in the air and on the stomatal conductance. Stomata are typically closed during the night. Consequently the ozone uptake is small during the dark hours. AOT40, however, suffers from the inherent assumption that the toxicologically important uptake of ozone is directly related to the exceedance of 40 ppb, regardless of the stomatal conductance. The stomatal conductance is sensitive to environmental factors such as solar radiation, temperature, the vapour pressure deficit of the air and the soil water availability. The scientists in the field were aware of this limitation already in the 1990s, but only in the last few years data and models have been available to quantitatively cope with this problem (see e.g. Pleijel et al., 2000, Karlsson et al., 2000).

The development, which is presently underway, represents the third generation of critical levels for ozone. The first formalised steps within the CLRTAP were taken in 1999 with a workshop devoted to these problems in Gerzensee, Switzerland (Fuhrer and Achermann, 1999). There are two major challenges here. One is to develop and validate reliable models to predict the stomatal conductance, and hence stomatal ozone uptake, under different environmental conditions. Another is to relate the predicted ozone uptake to effects observed in controlled experiments, obtained from biomonitoring systems or obtained from the field for important receptors among forest trees, crops and other plants.

It is possible to discern two parallels with the critical loads for acidification in this development. First, the ozone uptake models for effects on plants are based on a flux from the atmosphere to the plants, similar to the flux of sulphur transferred from air to soil in the case of acidification. Second, a more fine-tuned representation of the sensitivity of the receptor is obtained. In hot and dry conditions stomata will be closed to a large extent even during the day, while in cooler and more humid climates stomata will be open to larger extent. This kind of variation is not considered by the AOT40 approach. This is to some extent comparable to the sensitivity to acidification which shows a large variation mainly linked to variations in mineral weathering rates in different soil types.

2.3 Airborne particles and human health – importance of time for action

Since the findings of clinical effects of inert particles on animals and a clear relation between ambient particle concentrations and human health by epidemiological studies, the political pressure has been growing to implement new limit values for particles in the atmosphere as well as to restrict emissions from major sources. The basic knowledge on airborne particles is however limited. There are large gaps in our understanding of the occurrence of ambient particles, their physical and chemical characteristics, sources, transportation, transformation and deposition mechanisms. In addition to this, the relations between occurrence of particles and human health are not well established. In present assessments, the relations to health effects are expressed in terms of particle mass. However, there is little evidence that particle mass is the actual property harmful to human health. There are indications that several other characteristics of air pollution are harmful such as certain gases and chemical components in particles. Other parameters suggested are particle number and particle surface.

Presently particle mass, PM10 or PM2.5, attracts most interest as several major studies show a clear correlation with different health effects making these parameters major output parameters for the models. A Swedish network for monitoring PM2.5 /PM10 has been set up providing data for background, urban background and street sites in five urban areas and at two rural background sites. This network shows a dominant influence of long rang transported particles.

This calls for a more detailed characterisation of local as well as regional ambient aerosols in order to facilitate a better evaluation of possibly harmful components in the transboundary airborne particle pollution and a to provide a better basis for control measures. To facilitate aerosol characterisation and to quantify the relation to sources, support by models is needed. However, these models are still in a development phase and verification of model results with field measurement data is yet to be done.. Reliable emission data, models and measurements are necessary for evaluating the risk of human health impact from particles. Improved descriptions of particle dynamics will also serve to further refine the model description of more traditional pollutants such as sulphates and nitrates.

The new approaches need a strong link to the international community, partly because of the international dimension of the issue, partly because of the complexity of the problem. Several groups from different scientific disciplines should collaborate to achieve scientific progress and to facilitate the formation of a consensus around the issue. Within the ASTA programme contacts have been established to the EMEP community for model development and the establishment of a monitoring programme. A Nordic network has also been built up for co-ordinated continuous monitoring of particle size distribution and some other parameters. The data will be used to test detailed process models, parameterised aerosol modules as well as for evaluation of the final EMEP aerosol model.

The approach of the ASTA particle programme is to provide a key to evaluate the consequences of new limit values, to provide a better understanding of source – receptor relationships and to support a further development of the understanding of health effects of particles.

2.4 National assessment

Sweden is to more than 50% covered by forests. Forests are large receivers of air pollution and there is a strong interaction between air pollution impact and forest production, most obvious with respect to the deposition of sulphur and nitrogen compounds and the nutrient balances in forest soils.

The acidified areas in Sweden often coincide with highly productive forests, which can create certain conflicts. Both acidification and biomass harvesting will cause loss in alkaline compounds in the soils and thus contribute to acidification. The acidification caused by biomass extraction has not been fully taken into account in the critical loads concept and may cause problems in connection with increased biomass harvesting for energy production. Those products normally considered as most important for energy purposes also have proportionally higher contents of alkaline nutrients.

The objective of the national part of the ASTA programme is to apply the concepts, methods and tools developed for the international control strategies in national assessment work. In practise this means that critical loads concepts and experiences from dynamic modelling will be used for land use planning, in particular for productive forests. The results are particularly important for the forest and energy sectors.

Due to the complexity of the forest systems and the various impacts (climate, pollution load and forest practices), there is a need of integrated analysis to assess the options to reach important environmental goals combined with demands on land use. These options include issues such as high productivity and preservation of biodiversity.

Today, the responsibility for national measures falls upon different sector organisations and authorities. The combination of emission control and sustainable methods in forestry can influence future development of the sectors substantially. ASTA aims to support policy development both for environmental needs and for needs related to increased use of biomass.

2.5 Synthesis and policy-related activities – communication and consensus

Scientific research at a basic level is seldom directly applicable to policy. As pointed out in chapter 1, there are often several steps between the basic production of scientific knowledge and the final use of the results for policy. Within the field of transboundary air pollution, the development of strategies based on integrated assessment modelling provided a framework for the integration of knowledge from the basic levels (determination of rate constants for atmospheric chemistry models, monitoring single pollutants in the atmosphere etc.) over compartment models and data compilations to the integrated assessment models. This framework is based on underlying concepts, models, environmental data etc. The legitimacy and acceptance of the outcome from integrated assessment models are to a large extent dependent on the reliability of the underlying material. Reliability and scientific credibility has been achieved through a number of activities, from making data and models transparent and open to reviews and analyses of uncertainties of models and monitored data.

In the ASTA programme the integration and communication of scientific knowledge has been an important part of the programme. All research activities were planned with the objective to contribute in a practical and known way to the development of policy.

2.6 Credibility and transparency in scientific processes behind abatement strategies

The protocols under the LRTAP Convention are usually understood as being based on scientific knowledge. The scientific knowledge has been able to influence policy-making in a "clean" and rapid way leading to a science-based policy, which is politically fair and beneficial for the environment. The problem with this view, dominant among policy analysts, is the risk of supporting a technocratic policy approach.

The problem of technocracy has recently been highlighted by the European Commission, in its White Paper on Governance, focusing on what is called "democratising expertise" (European Commission 2001a). By this it is not meant "majority voting in science", but that the *process* in the way expertise is developed, used and diffused should be democratic. Democratised expertise is recognised by several specific characteristics. One of the most important is transparency, i.e. the visibility in how experts are recruited and how the process of development, use and diffusion of expertise is managed. The reason for this initiative is the crises which in the last few years have been witnessed in the European Union concerning expert knowledge and policy regulation, foremost in relation to food, health and the environment, e.g. BSE, GMO and climate change. Conclusions have been drawn, indicating a crisis for the credibility of expertise.

The objective of democratising expertise is to strengthen the quality and credibility of expertise in relation to policy makers and the public. It is argued that the credibility of expertise has to be high, otherwise regulation will not be acceptable to citizens. Since regulatory policy-making is considered an important feature of European integration as such, a questioning of the credibility of expertise can potentially affect the legitimacy of the whole Union.

In the Clean Air for Europe (CAFE) programme – launched in May 2001 in order to propose an integrated air quality policy – one main objective is to strengthen the links between research and policy, and at the same time increase transparency and stake holder involvement. In December 2001, a draft of the CAFE work-plan was presented. This plan gives an overview of the work to be finished in March 2004 when the first CAFE report will be published. The process is divided into 201 main blocks, assessed as critical in order to achieve the new strategy. One objective of publishing this detailed work plan is to increase the transparency of the CAFE programme, “both in the day-to-day proceedings and in the way research data and technical analysis are used for policy developments. Stakeholders have the opportunity here to present evidence and comments, giving as much clarification as possible about the technical justification and political motivation behind them” (http://europa.eu.int/comm/environment/air/index.htm).

What is set against transparency in the EU white paper – and therefore also in contrast to what is considered “democratised expertise” – is ”closed room expertise”, i.e. a too narrow assessment of quality and relevance, where the political majority could pick preferred expertise. Closed room expertise is driven by an expert elite and the work is carried out in an opaque way, which is difficult for outsiders to follow. According to the white paper, this leads to decisions apparently inaccessible to public scrutiny. Technocratic decision-making should be avoided because it undermines the legitimacy of the policy process.

The work in connection to the LRTAP Convention could be assessed as technocratic in the way that it has been driven by a small elite group and based on closed workshops. However, at the same time the work has been supported and considered credible among politicians and the general public. During the last years a decreasing interest among the general public could be noticed, due to less media coverage. In order to increase public interest and public support the work of the experts has to be opened up and the elite character be complemented.

ASTA includes a specific sub-programme on social science with the aim to contribute to an improved understanding of the role of experts in policy making. The intention is that ASTA as well as other more science-oriented research and development programmes would benefit from this sub-programme and use the results in order to orient the research towards an increased openness and understanding. The demand for increased transparency and critique of technocratic decision making are important phenomena which could be taken advantage of when further developing abatement strategies for transboundary air pollution. However, transparency should not be understood in a naive way, as just openness, but has to be combined with efforts to make the process of developing and distributing expert knowledge accessible to a wider audience. To strengthen the credibility of science means not only to disseminate scientific results in a public friendly way. A long-term strategy for gaining legitimacy is to involve stakeholders in the process of knowledge production, not least by creating different forums for knowledge exchange. Through public hearings, stakeholder dialogue etc. the perspectives and knowledge of researchers are spread in society, and at the same time the research community becomes more open for perspectives and knowledge that exist amongst other actors.

3 Achievements under the NEC and GP

3.1 Emission reductions – agreements and realities

The UN ECE Convention on Long Range Transport of Transboundary Air Pollution (CLRTAP) was established in 1979, as a first measure to reduce effects of acid deposition in Europe. As a part of this first agreement, emissions of sulphur dioxide were to be reduced by 30% in each of he countries signing the convention. This agreement has since been followed by a number of protocols as described in Table 3.1.

Table 3.1 Existing protocols within the UN ECE Convention (CLRTAP).

Year signed	Name	Species	Required reduction	By year
1985	First sulphur protocol	SO_2	30% in each country based on 1980 emissions	1993
1988	NO_x protocol	NO_x	Not exceeding 1987	1994
1991	VOC protocol	VOC	30% reduction based on emissions in a year between 1984 - 1990	1999
1994	Second sulphur protocol	SO_2	Differential, exceedance of CL reduced by 50%	2010
1999	Gothenburg protocol	SO_2, NO_x, NH_x, VOC	Differential (see Table 1.1)	2010

A good example of the results of these protocols is the decrease in SO_2 emissions (Figure 3-1). Drastic decreases occurred between 1975 and 1995. After this date, a slower reduction rate is expected and in 2010 the emissions are expected to decrease to just over 14 000 tons per year, roughly 25% of the emissions in 1980.

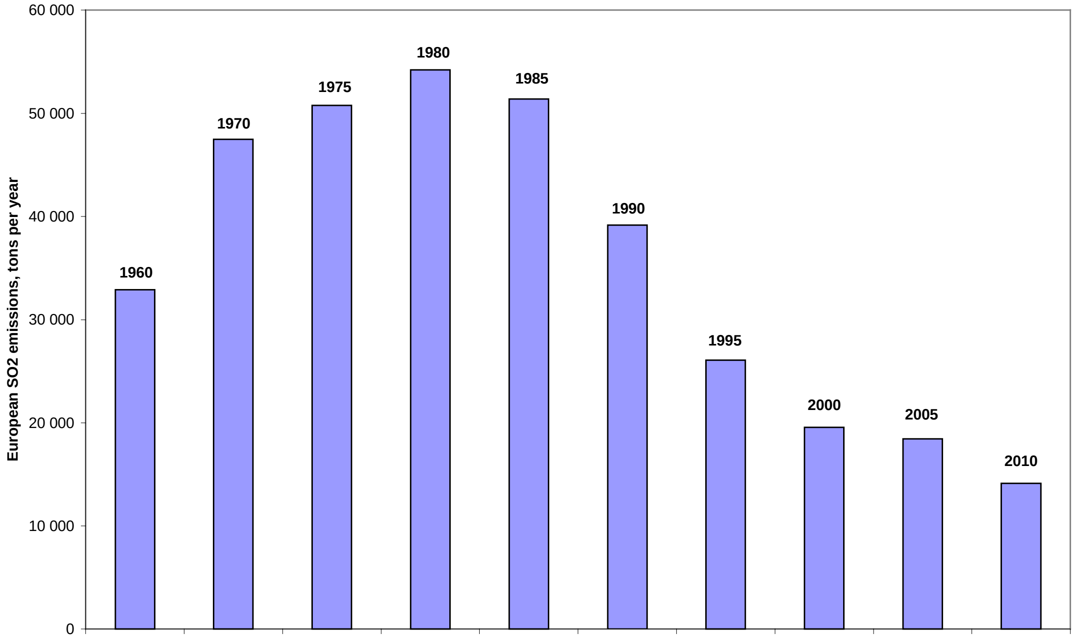
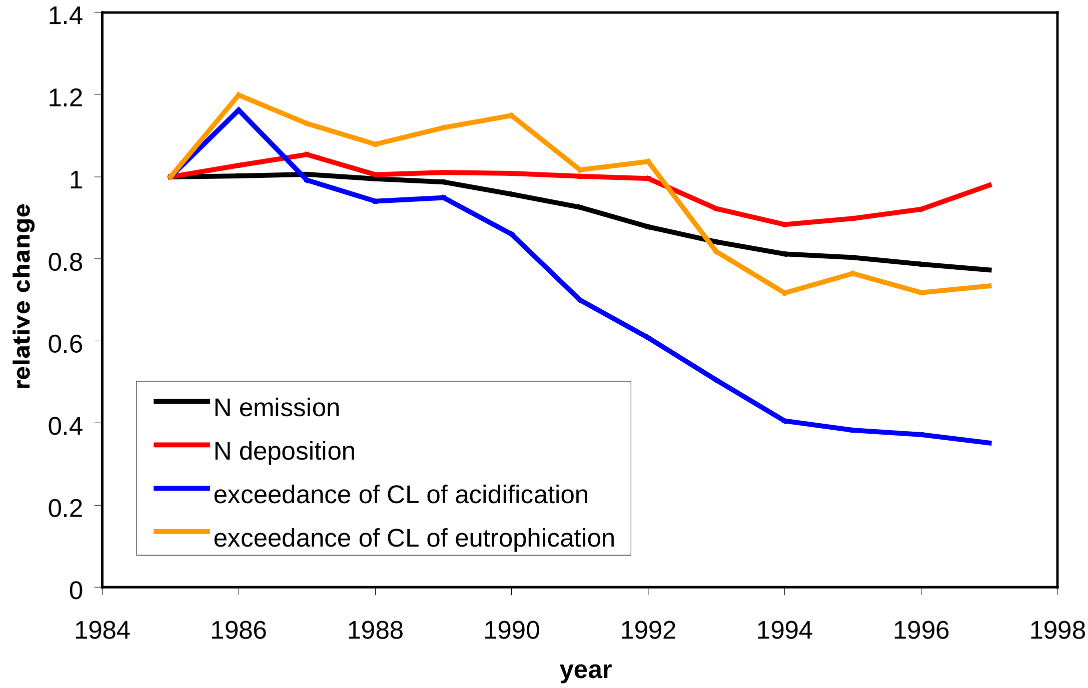


Figure 3-1 European SO_2 emissions 1960 to 2010 (Gothenburg protocol).

3.2 Changes in deposition/exposures and effects

The critical load concept was developed in order to control the acid deposition with an ecosystem effect-related concept. This connected for the first time ecological effects to measures. Instead of being at the focus of the process, demands for technological measures now became driven by environmental quality demands defined by ecological parameters. This was a total paradigm shift in terms of approaching environmental problems.



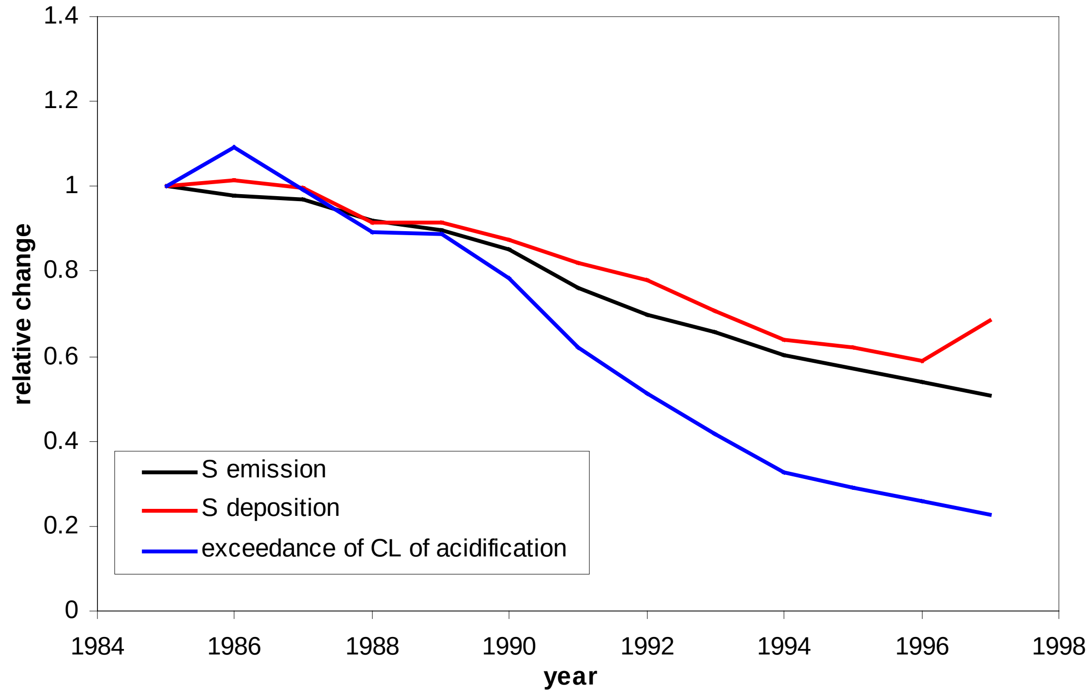


Figure 3-2 Relative changes in emissions/deposition/critical load exceedance for nitrogen (top) and sulphur (bottom) in Europe from 1985 to 1997 (data from http://www.emep.int, April 2002).

As a consequence of the decrease in deposition of the priority pollutants, deposition and critical load exceedances have also decreased markedly. In Figure 3-2, the normalised changes in emissions, deposition and critical load exceedance are presented.

3.2.1 Critical loads in Sweden

3.2.1.1 CRITICAL LOADS FOR FOREST SOILS

The critical load status is best shown by the exceedance maps, expressed as the 50-percentile, representing the average overload, and the 95-percentile, representing the overload if we want to protect 95% of all sites. For production in general, the 50-percentile maps will be most relevant. Be aware of the fact that the exceedance maps do not show the present situation in the forest, but only after a protocol has been fully implemented. This process normally takes several years, if not a decade, and only after that can the soil slowly recover. The recovery process is even slower that the political process and has been estimated to take from 20 to 250 years depending on site properties and soil chemistry state at the time of turning the deposition trend.

In 1988 the exceedances were very large, and the models predicted that a continuation of such deposition levels would lead to large scale damages to soil chemical state (base saturation, pH) and probably to growth damage of a same degree as in Germany and in the Black triangle. Damages would run into billions of SEK per year (Sverdrup et al 1995).

Table 3.2 Exceedance of critical loads in forest soils in Sweden.

Deposition year	Method	Assessment	Sites	Exceedance	
				% of area	10^6 ha
1980/81	PROFILE	1987	23	75	17,2
1980/81	PROFILE	2001	1883	66	15,1
1987	PROFILE	1989	1302	85	19,5
1987	PROFILE, SSMB	1991	1756	82	18,8
1987	PROFILE	1992	1804	76	17,4
1989	PROFILE	2001	1883	46	10,5
1992	PROFILE	1995	1883	52	11,9
1997	PROFILE	2001	1883	24	5,5
2010	PROFILE	2001	1883	14	3,2

In 1991, the second sulphur protocol, the "Oslo-protocol" was signed, for the first time based on the critical loads estimated for a number of European countries. It reduced sulphur emissions by 60% from the reference year (1985). This was a great achievement, but still far from what was required to remove the large scale threat against productivity in Swedish forestry. What was most important with the Oslo protocol was that it established a new principle; "effect-based mitigation of pollution", and "mitigation at the source". Both were more important than the actual emission reduction in the protocol itself, as this paved the way for the next protocol.

In 1997-2000 a new assessment was made and the databases revisited and updated. Table 3.2 shows the situation in the end of 2000/beginning of 2001. Exceedance is approximately 25-30% of the area (this is dependent on the input data and the true field exceedance is probably 35-45%). Some areas in the south still have significant exceedance, which will lead to continued acidification of forest soils. The Gothenburg protocol was a very large step towards compliance with no exceedance of critical loads, and when the protocol comes into full effect much of the threat to Swedish forest productivity will be reduced. In Sweden, there still is a significant exceedance with respect to protecting 95% of the sites under the protocol, but a large part of the territory is completely protected, approximately 60%, (Figure 3-3). There is exceedance in approximately 15-20% of the area (Considering scaling effects, the true field exceedance is probably 25-30% of the area under the Gothenburg protocol) and only a small part has large exceedance. Much of the remaining acidity exceedance is now caused by nitrogen deposition, a large part of this is in the form of ammonium deposition with large potential for acidification.

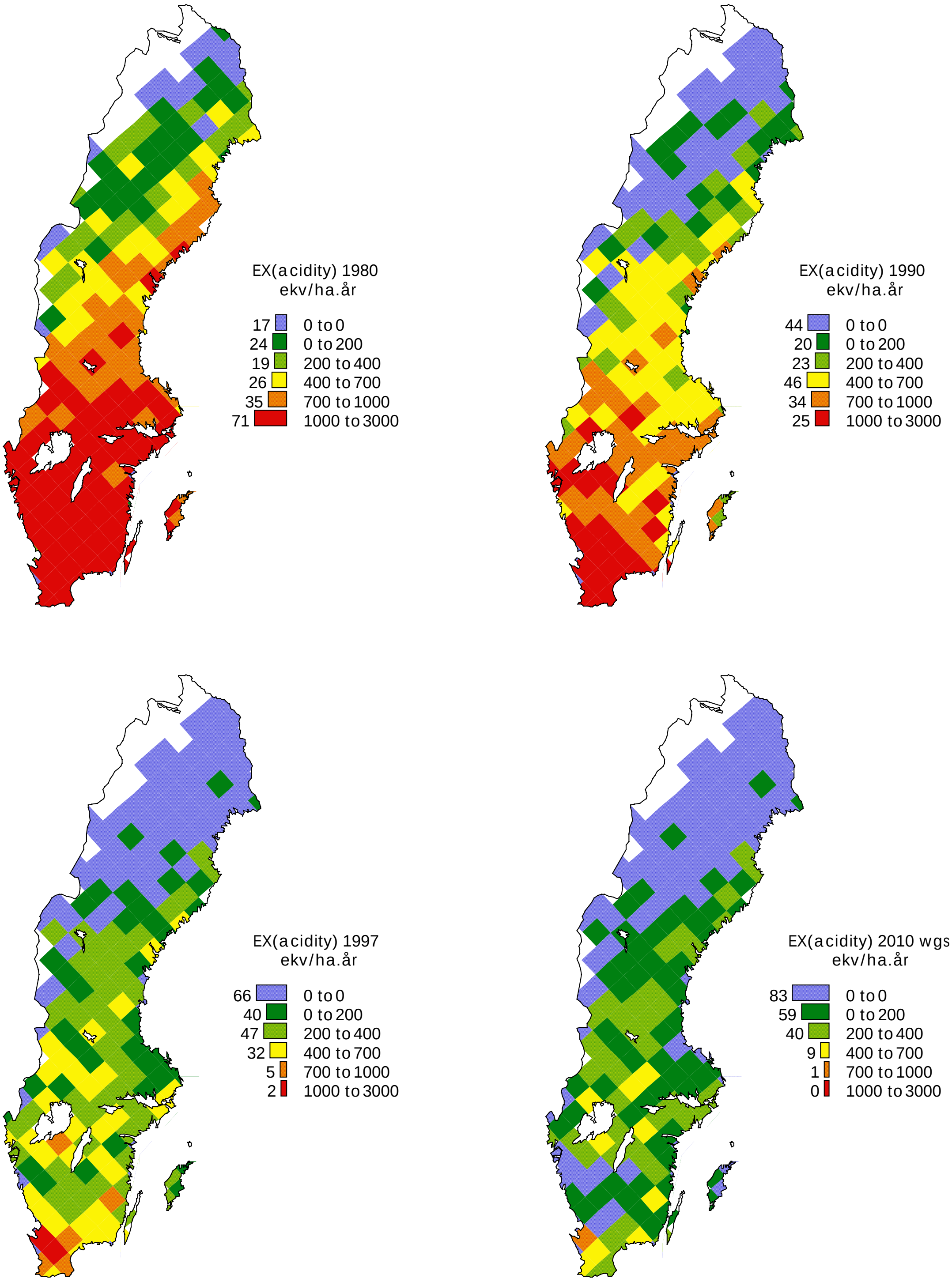


Figure 3-3 Exceedance of CL acidity 1980, 1990, 1997, 2010.

3.2.1.2 Critical loads for nitrogen

The expected development in the exceedance of critical loads for nitrogen is shown in Figure 3.6. Critical loads for N were estimated based on the mass balance method. The method is described and discussed in Bertills and Lövblad, 2001. A comparison of the inventories for 1997 and 2001 show an expected moderate decrease in the exceedances.

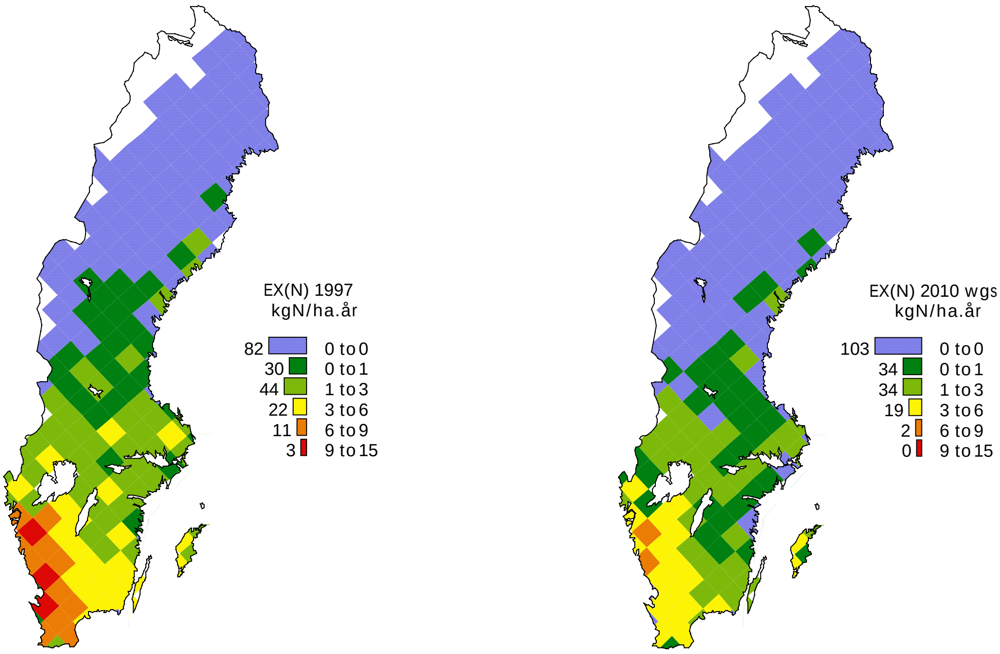


Figure 3-4 Exceedance of critical loads of nitrogen for 1997 (left) and 2010 (right) using PROFILE.

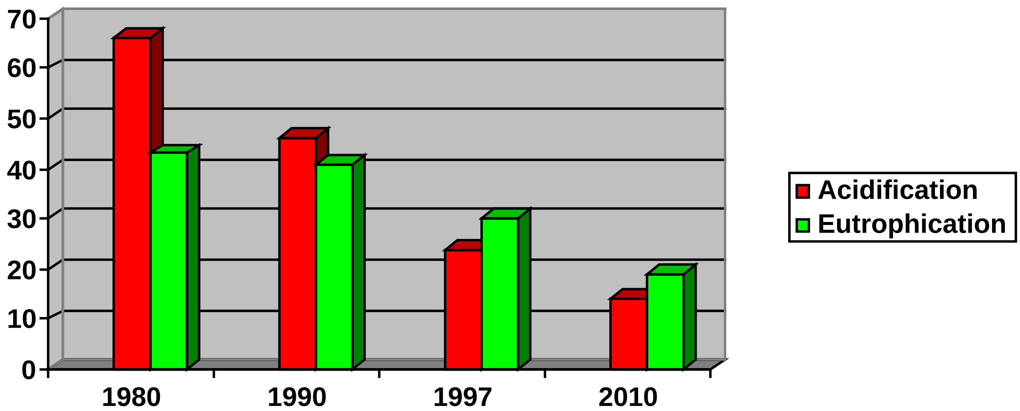


Figure 3-5 Comparing exceedance of critical loads of acidity and nitrogen over time in Sweden.

3.3 Signs of improvements and recovery

3.3.1 Acidification

Recovery from acidification generally refers to the return of ecosystems to a state which is similar to the state they were in before the onset of industrialisation in

Europe i.e. one or two centuries ago. In this chapter we will discuss signs of recovery in terms of chemical recovery of surface waters, chemical recovery of soils and biological recovery.

3.3.1.1 SURFACE WATERS

The earliest signs of recovery in a catchment as a result of decreased deposition are generally observed in the surface water chemistry. Changes in lakes, rivers and streams are also easier to monitor than soils. In Sweden we also have long and reliable records of surface water quality from a number of sites. Therefore there is a great potential for mapping surface water acidification status and trends. In areas where soils have not been significantly acidified and acidification problems are largely related to episodes, and particularly spring flood in northern Sweden, the recovery response to declining deposition is essentially immediate. Where soils are significantly acidified, the lag times in which the surface waters react to a changed deposition originates in part from the rate in which the catchment soils react to changed deposition.

Sulphate deposition has declined across most of Europe since about 1990, although the magnitude and exact timing varies (Mylona, 1996). After the first few years of decreasing deposition, the response in surface waters was mixed and no general improvement was apparent (Newell and Skjelkvåle, 1997). However, there are now a number of recent studies which convincingly demonstrate, that the decline in acidifying deposition had resulted in an improved chemical status of surface waters in sensitive regions of Scandinavia (Skjelkvåle et al., 2001; Fölster and Wilander, 2001; Moldan, 1999; Wright et al., 1994). Major syntheses of several regions, across the whole Europe and even North America have also found signs of recovery (Evans et al., 2001; Stoddard et al., 1999). The general pattern is that decreasing deposition of S and N causes decreased SO_4 concentrations in the lakes and streams and decreases in both base cations, H^+ and Al^{n+}. Consequently, there was an increase in alkalinity, which was generally largest at the most sensitive regions and small or absent in the regions without significant acidification. In a study of 114 liming reference lakes in Sweden sampled approximately between 1983 and 1997, there was a recovery in alkalinity in 83% of the lakes, with the degree of recovery generally correlated to the degree of acidification. The average increase in alkalinity was 1 μeq/yr, which was half of the average decline in sulphate concentrations (Wilander and Lundin, 2000). While going in the right direction, the observed annual rate of chemical recovery is so far much slower and smaller than the decline in acid deposition.

The lag and attenuation in the recovery response of surface waters at decreasing deposition originates in part from the rate at which the catchment soils react to changed deposition.

From an ecological standpoint, it has long been recognised that the mean annual or baseflow chemistry that is generally reported (as in many of the studies cited in this section), miss an important aspect of acidification, namely the short term episodes of pH decline that can occur during high flows, particularly spring flood. During recent years significant strides have been made in quantifying the sensitivity of spring flood to acid deposition (Bishop et al, 2000, Laudon 2000) as well as the rapidity of the response in spring flood to changed deposition (Laudon and Hemond, 2002, Laudon and Bishop in press Geophysical Review Letters). This is an important and immediate benefit that can now be ascribed to deposition declines in areas where soil acidification had not advanced significantly.

3.3.1.2 SOILS

The recovery of the acidified soils is a slow process in terms of alkalinity build-up. Consequently, to detect changes over a few years or even one decade is difficult, because the change is small and there is a substantial spatial and temporal variability. Furthermore, the soils subjected to an increased deposition of S accumulate a certain amount of sulphur, either as an adsorbed SO_4 (Alewell et al., 2001) or possibly also as organic sulphur, built in to the organic matter (Novak et al., 1996). When the deposition decreases the soils will loose some of the adsorbed sulphate and possibly even mineralise and release some of the organically bound S, as shown e.g. at the roof experiment at Gårdsjön (Mörth et al., 1995; Hultberg and Skeffington 1998).

Sweden's Integrated Monitoring program on four intensively studied catchments across Sweden has also documented S leaching for the superficial organic layer that is significantly higher than the deposition inputs of sulphate. This is a further evidence of previously deposited sulphur starting to leach from the soil (Löfgren et al., 2001).

Both desorption and mineralisation of sulphate generate acidity and they will cause a lag time in recovery. To date there is little evidence that there is a regional recovery of acidified soils anywhere in Scandinavia or Europe. The build up of soil base saturation could take place only when the input of base cations to a catchment (typically an atmospheric deposition and mineral weathering) exceeds the output (runoff leaching and uptake to the vegetation).

3.3.1.3 BIOLOGICAL RECOVERY

It is anticipated that once the chemical criteria improve in soils and waters, the biology will recover. There is abundant data on improvements of fish population and on invertebrates following liming of the acidified lakes and rivers. The evidence of recovery of terrestrial ecosystems after liming the forest floor is much weaker and sometimes even controversial (Binkley and Högberg, 1997). The data to show that it is actually happening are available from various studies, where the acidified ecosystems were limed. However, the evidence, that there is a biological recovery taking place at any part of ecosystem following the deposition decline of the last decade in Europe is also rather weak. There is a time lag from when the chemical conditions start to improve to when they actually reached a status that allows biological recovery. Time is also needed for species to change their abundance (either by re-colonisation or reproduction) Difficulties in detecting these changes are another reason why biological recovery is not observed to the same extent as chemical recovery. This lag may also indicate a need to remediate other human impacts besides acidification (such as physical habitat disturbance.)

The lack of biological data and complexity of predicting biological recovery may point to the need both for more biological research and the continued importance of employing chemical indicators of recovery.

3.3.2 Ozone

3.3.2.1 TRENDS IN AIR CONCENTRATIONS

Trends in European ozone are studied within the frame of "TRopospheric Ozone and Precursors – TRends, Budgets and Policy (TROTREP)", a project of the Thematic Programme for Environment and Sustainable Development within the Fifth Framework Programme. Hourly observations of ozone and daily means of NO_2 from three monitoring sites in Sweden; Esrange, Vindeln, and Rörvik, representing remote areas, and 15 German and Dutch sites, representing more polluted areas, are used. A statistical model that links observations with meteorological parameters was developed and applied to the German and Dutch sites. The average summer trend is –1.10 ppb/year for the 90th-percentile of daily maximum ozone and –1.46 ppb/year for the 90th-percentile of oxidant (Ox = O_3 + NO_2). The peak scavenging of ozone is at a rate that is about half of the rate of emission reductions of NO_x and NMVOC. Ozone winter trends are slightly positive +0.26 ppb/year over the 1992-2000 period, but, in general, virtually no trend (+0.02 ppb/year) was found for Ox. The positive ozone winter trend can therefore be attributed to a reduction in the titration reaction O_3+NO as a result of NO_x emission reductions. Changes in the chemistry seem very small as we regard the no-trend of oxidant. The result also indicates that lack of relevant meteorological information can result in a misinterpretation of measured trends.

At Rörvik (Sweden) a significant (p=0.05) upward trend of 0.25 ppb/year in background ozone is obtained for the period 1987-2000, mainly driven by the winter observations (Figure 3-6). The winter half-year ozone average has increased with 0.33 ppb/year (1.54%/year, p=0.005). The winter average of Ox has increased with 0.18 ppb/year (p=0.02), in spite of the decrease in NO_2, thus indicating a genuine upward trend in ozone. Also the daytime daily maximum is increasing during winter. No downward trend is found at any time of the year. The same tendency of increasing ozone levels is found at Esrange (1991-2000) for both seasons. In this case the results most certainly reflect an increasing hemispheric background. The development for Vindeln is quite different with a significant decrease in summer background ozone, -0.31 ppb/year (p=0.02), mainly explained by decreasing concentrations in July. Increasing levels are however found for individual months and percentiles.

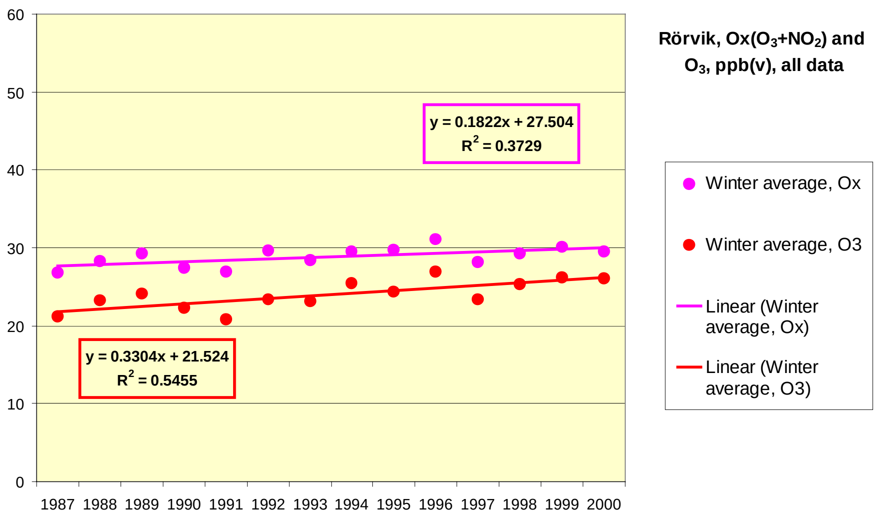


Figure 3-6 Winter averages of ozone and Ox at Rörvik, 1987-2000, based on all data.

3.3.2.2 Effects on vegetation

Effects of ozone are monitored at about 20 sites in Europe within the framework of UNECE ICP Vegetation (WGE 1998). Ozone sensitive and tolerant clones of white clover, as well as plants of several other species, have been used as indicators in standardised tests during the summer months 1992 to 1997. Visible injury occurred on at least one of the species at each of the sites and there is no clear evidence for a change in the effects of ozone during this time period. In general the extent of injury reflects the levels of ozone across Europe in any one year.

Ozone concentrations in ambient air in south Sweden have the potential to cause visible injury to leaves of sensitive plant species. During the summer 2001 there were several ozone episodes that caused clear visible injury on the ozone sensitive clone of white clover (Figure 3-7).

Figure 3-7 Visible leaf injury on the ozone sensitive clone of white clover (*Trifolium repens* cv NCS), caused by ozone in the ambient air at Östads Säteri, 45 km north-east of Gothenburg, Sweden. Photograph taken 2001-07-25 from an ICP-Vegetation experiment financed by the Swedish Environmental Protection Agency.

Furthermore, it was shown that ambient ozone levels during the summer 2001 also could cause visible leaf injury on leaves of *Centaurea jacea*, a plant growing naturally in south Sweden (Figure 3-8). Thus, it seems that the ozone levels in 2001 were high enough to cause visible injury on naturally growing plant species in south Sweden.

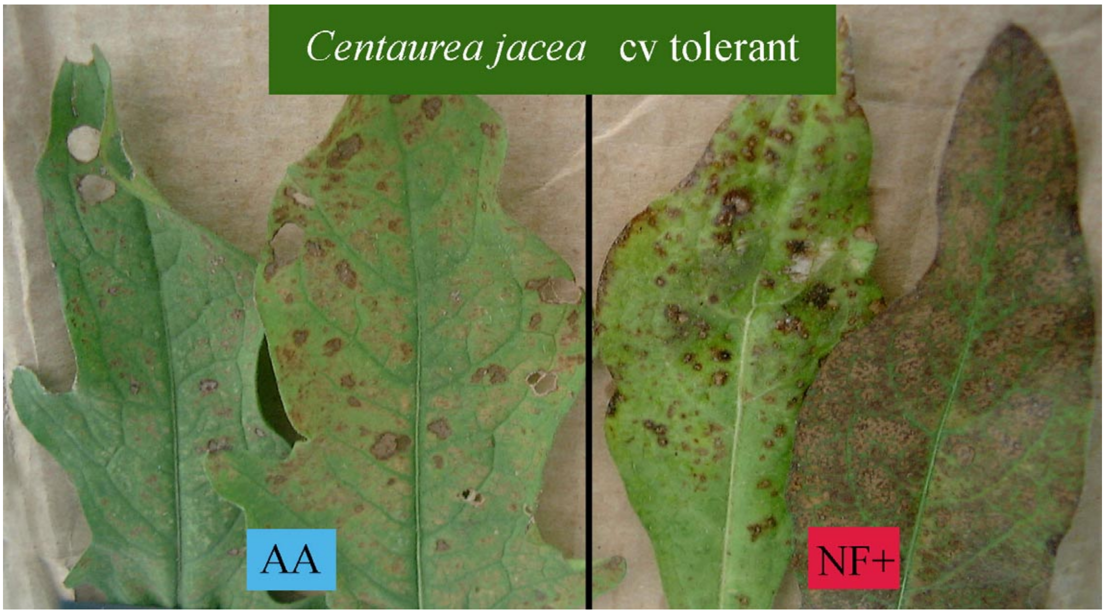


Figure 3-8 Visible leaf injury on *Centaurea jacea*, caused by ozone in the ambient air (AA) and in an open-top chamber with elevated ozone (NF+). Leaves from one open-top chamber with filtered air showed no signs of injury. Photo taken at Östad, Sweden, 2001-08-24 from an ICP-Vegetation experiment financed by the Swedish Environmental Protection Agency.

The overall conclusions for ozone are that the trends are very weak both in terms of observed ozone concentrations and observed effects and that increasing hemispheric background concentrations may counteract improvements due to emission reductions in Europe.

3.3.3 Particles

The evidence that airborne particles cause significant health effects have increased the general interest in understanding and quantifying composition and sources of these particles. The recently approved CEN measurement standard for particle mass (PM10) has made it possible to perform comparable measurements all over Europe. The EMEP measurement programme had until recently not given any priority to measurements of the particle mass. Even if data exists from national programs and research projects, comparisons and trend analyses are difficult due to differences in sampling techniques applied in individual countries and changes in instrumentation over time. However some insight can be gathered by looking at one site and to make a first rough comparison with other similar stations.

The EMEP site Aspvreten in central Sweden has a record of PM10 since 1990 which can provide some insight in trends. In this limited data set a general decreasing trend of sulphate concentrations in air is clearly recognised (Figure 3-9). The other inorganic components follow roughly the same trend.

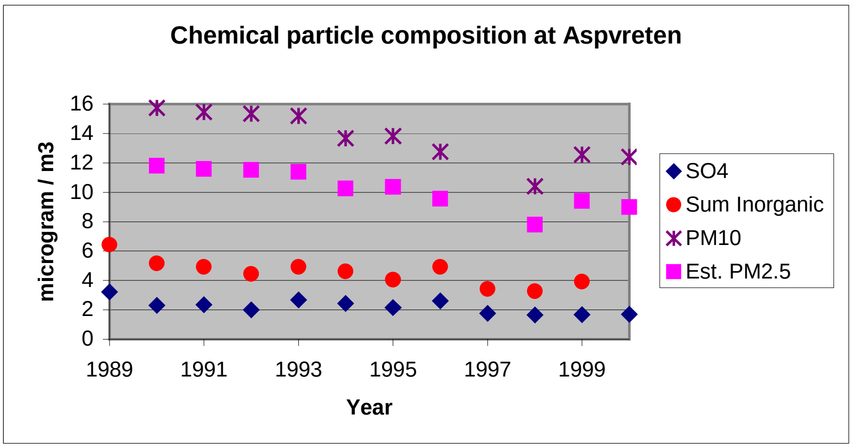


Figure 3-9 The development with time on concentrations of sulphate, sum of inorganic ions (SO_4, NO_3 and NH_4) and PM10 at Aspvreten. PM2.5 is estimated from the last two years measurements.

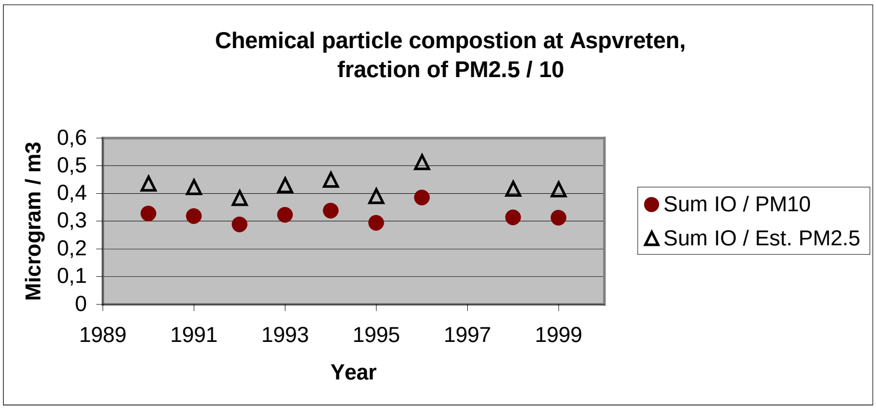


Figure 3-10 The fraction of the sum of inorganic ions (SO_4, NO_3 and NH_4) to PM10 and to an estimate of PM2.5 at Aspvreten 1990 to 1999.

When summing up the mass of the measured inorganic components sulphate, nitrate and ammonium, it is striking to see that for this site the fraction of inorganic ion mass to PM10 is roughly constant around 0,3 – 0,35 over this ten year period. This indicates that the contributions from other sources than S- and N-compounds has decreased with the same relative rate as sulphate. This includes both secondary as well as primary sources for fine as well as coarse particle.

Looking at the EMEP data, only a few stations in Switzerland, Germany, Spain, Italy and the Netherlands have long enough records to allow a trends analysis. During the last ten years all, stations show a decrease or an indication of a decrease in particle mass concentrations, i.e. the same result as was found at Aspvreten. The ratio of sulphate to total particle mass is about the same as in Aspvreten. Despite this, it is quite difficult to make any general statements on trends. At some stations roughly the same concentrations are found during the last 10 years while others show some indications of a decreasing ratio.

During the last two years, PM2.5 has been measured at Aspvreten giving a possibility to estimate previous PM2.5 mass concentrations using the measured ratio PM2.5 / PM10. The ratio of the sum of inorganic compounds to PM10 is roughly constant around 0.4. It is striking that the S and N compounds of importance in the Gothenburg protocol and the NEC directive represent less than 50% of the total fine particulate mass. Other potentially important components are organics and resuspended dust.

3.4 The situation 2010 ozone, acidification, nitrogen, particles.

The situation in 2010 will to a large extent be dependent on the implementation of the Gothenburg protocol and the NEC directive. The protocol and the directive include rather drastic reductions of SO_2 emissions and also substantial cuts in VOC and NO_x emissions (Table 1.1). For NH_3, the protocol will lead to emission cuts of 17%, which is clearly not sufficient to reduce the impacts of eutrophication in Europe to acceptable levels. Another important issue is the countries capability to reach the target emission reductions by 2010. At present, only 2-3 countries have ratified the protocol, which means that there are no legal instruments for implementation in most of the individual countries. Some countries are however well in place and have at least fulfilled the tasks for sulphur dioxide.

For ozone the protocol is expected to lead to significant reductions of peak values in Europe. At the same time, as pointed out earlier in this chapter, observations from monitoring and research activities indicate that background concentrations are unchanged or increasing. This is mainly an effect of increasing hemispherical background levels and may lead to requirements of further emission cuts in Europe in order to avoid negative effects on plants and human health (Johnson et al., 2001). The observations also points to the needs of an assessment and possibly a strategy on the reduction of northern hemispheric background concentrations in ozone.

The effects of the Gothenburg Protocol on emissions and ecosystem impact have been assessed using the RAINS model (Amann et al., 1999). The study focussed on acidification, eutrophication and ozone effects on vegetation and human health, for different emission scenarios. Here we summarise some of the results from this study for three different scenarios:

- "1990" is the reference scenario representing the emission situation in 1990;
- "Protocol" representing the emission situation after implementation of the emission ceilings of the Gothenburg protocol; and
- "MFR ult" representing a hypothetical Maximum technically Feasible Reductions.

The protocol will lead to significant improvements of the ecosystem effects in comparison to 1990. In Table 3.3, the fraction of European ecosystems where the critical load for acidification is exceeded is presented for different emission scenarios. The improvement from the scenarios "protocol" to "MFR" is on the order of 2% reduction of *ecosystem exceedance* (unprotected ecosystem area). Even if this is a low number when considering Europe as a whole, the 2% represents a significant improvement in regions such as southern Scandinavia, UK, and mountainous areas in Central Europe. In addition to this, the critical load calculations are based on static critical loads and do not take into account the recovery time. When the "protocol" scenario is implemented, a large fraction of the sensitive ecosystems where the critical load was exceeded in 1990 or earlier will still be severely acidified despite significant reductions in sulphur deposition. Reduction of emissions according to the MFR scenario will reduce the time needed for recovery from acidification significantly. To support these further emission reductions, development of integrated assessment models taking into account recovery times are a necessity. This is also the main focus of the acidification research in ASTA.

Table 3.3 Percent of ecosystems with acid deposition above their critical loads for acidification in three emission scenarios: in 1990, for the Gothenburg Protocol and the hypothetical maximum technically feasible reductions (MFR) scenarios (from Amann et al., 1999).

	Percent unprotected ecosystem area		
	1990	Protocol	MFR
Sweden	16.4	3.8	1.2
EU-15	24.7	3.6	0.6
Total	16.1	2.6	0.3

For eutrophication, the results of the scenario calculations are presented in Table 3.4 for the three scenarios. The relative difference between critical load exceedance in the scenarios protocol and MFR are larger than the difference in the case of acidification. This is partly an effect of the different nature of the ecosystem impact of nitrogen. The effect of nitrogen impact is not limited to sensitive areas in the boreal or mountainous regions but will appear over a large geographical area covering most of continental Europe. Reduction of nitrogen emissions is in this perspective a high priority for future protocols. In ASTA, the impacts on ecosystems of nitrogen deposition are also a focus or research aiming towards a new concept for critical loads. The main hypothesis of this research is that excess nitrogen deposition leads to vegetation changes and thus affects biodiversity at levels much lower than the present. This implies that even in areas where the critical loads are not exceeded at present, significant impacts on biodiversity and ecosystem structure may have already occurred. If these impacts and the basic processes involved can be described quantitatively and thus modelled, critical load concepts based on vegetation change and biodiversity can be developed and applied in the next generation protocol negotiations.

Table 3.4 Percent of ecosystems with nitrogen deposition above their critical loads for eutrophication for in three emission scenarios: in 1990, for the Gothenburg Protocol and the hypothetical maximum technically feasible reductions (MFR) scenarios (from Amann et al., 1999).

	Percent unprotected ecosystem area		
	1990	Protocol	MFR
Sweden	13.8	4.6	0.3
EU-15	55.3	39.5	16.1
Total	30.3	19.9	6.4

In Table 3.5, the cumulative vegetation exposure to ozone is presented. It is clear that significant improvements are expected in comparison to the situation in 1990. However, it is also evident that further reductions of VOC and NO_x emissions are needed to further reduce the impacts of ozone damage to vegetation. The scenario calculations are based on the AOT40 concept, which is based on the assumption that the vegetation damage is linked to the duration and extent of exposure to ozone concentrations above 40 ppb. Recent research has shown that the damage to vegetation is not correlated directly to concentration since it is the stomatal uptake of ozone that is the controlling process for the bio-availability of ozone. The stomatal uptake is, apart from concentration, dependent on other factors such as humidity in the soil and air, solar irradiance and temperature. This implies that differentiated impacts can be expected in e.g. southern and northern Europe (where climatic differences are apparent) even at the same air concentrations of ozone. The development of ozone flux models for different vegetation types is the main aim of the ozone programme within ASTA.

Table 3.5 Vegetation exposure indices for the emissions of 1990 and the Protocol, and hypothetical maximum technically feasible reductions (MFR) scenarios. Cumulative vegetation exposure index (1000 km^2 excess ppm.hours) (from Amann et al., 1999).

Cumulative vegetation exposure index (1000 km^2 excess ppm.hours).Average			
	1990	Protocol	MFR
Sweden	163	11	0
EU-15	12412	6804	1875
Total	21946	12200	2074

The future situation concerning particle exposure in more difficult to predict. Emission inventories, atmospheric transport and transformation models as well as integrated assessment tools are currently under development and this is a major focus of development work within the convention. Further research and development is here clearly warranted before a scientifically sound abatement strategy can be implemented.

4 Scientific approaches

In this chapter we will elaborate the different scientific approaches and methods used in the ASTA programme – all the way from basic research over syntheses and compartment models to integrated models and assessments with the direct aim to support policy issues. In connection with the establishment of the ASTA programme the scientific needs in different areas were evaluated and some of the most crucial were taken on board in the programme. Most of the priorities were made in relation to the needs for Integrated Assessment Modelling. For some areas, the maturity in knowledge were low and in these areas experimental research has been an important part of the programme. For other areas, where more basic knowledge was available, focus has essentially been put on the development of model concepts and criteria to support the IAM work.

We will start with a presentation of the methodologies used for studies on how scientific credibility is reached and then continue on a general discussion of the relations between scientific understanding, conceptual and computational models and how these may be used in connection with air pollution strategies. Then we will continue with the more specific presentation of the methodologies used in the different sub-programmes.

4.1 Establishing credible and legitimate abatement strategies

One central aim of the ASTA programme is to improve our understanding of the science-policy relationship and make use of it in the science-oriented parts of the programme. This means to improve communication between different groups, in theory as well as in practice, when further developing air pollution abatement strategies. Most studies on the role of science in environmental policy-making have been conducted by political scientists. To a large extent this research has taken scientific results – and the development of expertise – for granted, as “black-boxed” inputs to policy negotiations. At the same time it has proposed that scientific results have been of great importance in the policy-making process (Gehring 1994; Grünfeld 1999; Levy 1993; Wettestad 1999). For instance, the protocols in connection to the LRTAP Convention are understood as being based on scientific knowledge which have been able to influence policy making in a strong way leading to a science-based policy, politically fair and good for the environment. From the scientific side, on the other hand, there is often an understanding that scientific results as published in peer reviewed papers should be taken care of by policymakers and used for the development of policy without any further interference with science. Many scientists maintain a strong boundary to the policy process. Our assessment has found that the viewpoints held by both policy analysts and scientists partly misconceive the relationship between science and policy in the CLRTAP process. Therefore, we have proposed an alternative approach, which explains the production and distribution of expertise relevant for policy making as a social process without postulating a clear-cut dichotomy between science and policy.

The problem with the traditional view is twofold. The first is the risk of supporting a *technocratic policy approach*, while the second means to support a superficial and *clear-cut dichotomy between science and policy*.

The problem of technocracy has recently been confronted by the EU, in its White Paper on Governance (see section 2.6), with the aim of democratising expertise. This initiative should be assessed in relation to problems, which are considered science-based but where expertise is controversial or distrusted. The ambition is to overcome "the frequent opaqueness of the process of providing advice and of tracking the evidence produced and used" (European Commission 2001a).

Among the most important characteristics of democratised expertise is transparency, i.e. the visibility in how experts are recruited and how the process of development, use and diffusion of expertise is managed. Obviously, transparency is considered a key characteristic in striving for credible expertise and successful policy process. In the EU CAFE Programme one main objective is to strengthen the links between research and policy and at the same time increase transparency and stake holders involvement. In the Programme it is stated that:

> *The need to increase transparency and bring Community policy closer to the citizen is well recognised. Regular, accurate information on Community policy is essential in order to increase public trust. As well as helping citizens to feel more involved, such information also allows the public to influence policy being made in their name. Such participation is particularly important for environmental policy where the public, as opposed to economic interests, provides the key driver… regular and accurate information on the progress and priorities of environmental policy will help to motivate and guide such change (European Commission 2001b: 11-12).*

The new demands put forward in the Commission's White Paper, and followed up in the outline of the CAFE Programme, are interesting in relation to the LRTAP work, where a technocratic tendency can be identified. However, at the same time the LRTAP work has been supported and considered credible among politicians and the general public.

The second important thing that is lacking in the traditional approach described above is the understanding of how the existing boundary between science and policy and the credibility of expertise *is achieved*. A clear-cut boundary is presupposed and from this foundation recommendations are delivered on how science and policy *should be* balanced, e.g. how to balance neutrality with partisanship; make room for scientific evidence as well as scientific questioning and uncertainty in the policy process, keep the process transparent; and make the actors accountable. From the viewpoint of experts it is focused upon how to give reliable advice; communicate to policy makers; build strong relations to policy makers and stake holders; and maintain integrity.

This approach is not satisfying, since it leaves the most important issue uninvestigated. Before recommendations are formulated, a better understanding of "the social machinery used to produce, present, and defend science advice" is needed in order to “explain *in operational terms* precisely how advisory bodies achieve and defend their credibility” (Hilgartner 2000).

In the ASTA programme we have focused our interest on how to improve the alternative understanding on the science-policy relationship while basing it on the following three presumptions (Lidskog & Sundqvist 2002):

i) knowledge never moves freely, but has to be carried by social arrangements in order to be distributed in society, ii) the value of scientific knowledge, for instance the value of science for policy, is not given by its content but is negotiated by scientists in social processes where also other actors are involved, iii) science and policy are co-produced: scientific knowledge and political order are shaping each other in an interdependent process of evolution.

The objective of our studies is to find out if there are common elements in the relationships that are of importance for the further development of international environmental strategies in general and for the European air pollution control strategies in particular.

Our approach stands against technocracy, in which science as a key source for policy is taken as a presumption without understanding the social base for its importance. A conclusion is that scientific knowledge has no strength in itself but has to be given strength by different institutions, and this has to be explained by the social scientist.

One important example of the co-production of science and policy was the establishing of critical loads as a key concept in abatement strategies for transboundary air pollution (Sundqvist, Letell & Lidskog 2002). This concept has served as an important meeting place, a *boundary object,* which shapes legitimacy within science and politics as well as among other stakeholders. Boundary objects are defined as ”objects which are both plastic enough to adapt to local needs and constraints of the several parties employing them, yet robust enough to maintain a common identity across sites” (Star 1989:21). Such objects are tools for integrating different groups, for example scientists and policy-makers, while at the same time helping to create consensual attitudes and knowledge. The boundary object of critical loads could be understood as an object making science and policy more interdependent and giving stability to abatement strategies, for instance the LRTAP protocols. This means a production of mutual understanding, as well as mutual interests, among scientists and policy-makers.

Also the concept of transparency has to be understood more properly. To just propose the need of increasing transparency does not solve the problems with quality and credibility of expertise. On the contrary new problems could be created if transparency is not properly understood. More important than transparency is to improve the understanding on how expert advice as well as policy regulation achieve its credibility. In addition to transparency it is necessary to give attention to how expert-based policy regulation actually is produced. Transparency has to be connected to the social process of shaping credibility, and critical questions about how credibility is achieved must be asked. Expertise is produced by people involved in social processes as part of specialised cultures, and to just look into such processes without understanding the strategies used by the experts is perhaps a good start but has to be accompanied by an increased understanding of these cultures (Sundqvist, forthcoming).

4.2 Strengthening the scientific support: Assessment modelling in ASTA

The policies developed within the framework of CLRTAP rests on two fundaments:

1. A general scientific understanding (conceptual models) of the environmental problems, their causes and possible solutions on one hand and

2. A system of computational models from which consequences of different scenarios and control measures can be assessed or optimised strategies can be developed (compartment models and integrated assessment models).

None of these have been more important than the other. The general understanding has been important for the acceptance and legitimacy of the problem. Without scientifically derived evidence on long-range transport of air pollutants and the links to devastating environmental effects, the problem had not been placed on the agenda. The advanced theoretical modelling approaches have then been able to quantify the relations.

The LRTAP work has always had a dualistic approach between producing scientific evidence through research and monitoring and model developments and applications in order to generalise, quantify and prognosticate the knowledge on air pollution. The methodology approaches in ASTA have considered both these aspects but have through its first phase put an increasing interest in the modelling part. This means that experimental research has focused on the use of results in particular in integrated assessment modelling.

The activities within ASTA have the following structure

1. Acidification

2. Eutrophication

3. Ozone and gaseous effects

4. Particles and human health

This structure has been followed as separate lines with a number of interactivity integrating efforts. The integration emphasis will be strengthened in the second phase in ASTA.

4.3 Acidification assessments

4.3.1 General approaches for new critical load concepts

The question of recovery and how to express the dynamic aspects in recovery in terms that can be used for assessments has been the focus for ASTAs activities on acidification. The activities have consisted of experimental research, model development and active promotion of the issue in terms of presentation of the issue at the Critical loads conference in Copenhagen 1999 and the organisation of two Expert Group Meetings in 2000 and 2001.

The Expert Group concluded that several model tools for dynamic modelling are available in Europe and that activities are on-going in several countries. No final proposal on how to integrate dynamic modelling results into IAMs has yet been presented but potentially useful methods include:

- Recovery iso-lines. Recovery times are calculated for a given environmental parameter at different deposition scenarios.

- Critical load functions. Describes critical limits as a function of NO_x and SO_2 deposition

These outputs can be linked to the RAINS model either via simplified functions derived from modelling results or by using the actual results for assessing the benefit of different modelled deposition scenarios. Remaining questions are how to generalise results into grids or regions, and if target values should be used e.g. based on biological effects in aquatic ecosystems.

ASTAs approach can be summarised in the following three items: i) to initiate interest and international research collaboration on the dynamic (recovery) aspects of acidification, ii) to collect experimental and monitoring data in order to achieve a better understanding of the links between decreased atmospheric input and environmental responses and iii) to develop and apply methods for dynamic modelling for national assessments and for integrated assessment modelling (IAM).

4.3.2 Experimental data in support of model development-acidification and recovery - Roof experiment

The best way of testing and evaluating the geochemical models is to use the data from two principal sources: long term monitoring programmes and large scale manipulation experiments. The monitoring programmes have the advantage of providing the information from the real world situation. Understanding the time series of monitoring data and the ability of models to reproduce these serves as an indicator of the confidence in model predictions. The advantage of manipulation experiments is that a large degree of control can be exercised and only those parameters which are relevant to questions asked can be manipulated in a way resembling an expected future scenario. ASTA has taken advantage of both information sources. The Roof Experiment focussed on the effect of a drastically decreased deposition. The deposition reduction was designed to be even greater than expected over the next 10 years (next 20 years at the time of construction).

The roof at Gårdsjön was built from the beginning with model development and testing as one major objective (Hultberg and Skeffington 1998). The key findings, which have been used for the modelling, include:

- the rate of recovery of waters and soils in a heavily acidified catchment

- the importance of the sulphur stored in the soil for delaying the recovery
- the limits of the recovery in surface waters which could be expected without any increase of the soil base saturation
- the sensitivity of the runoff composition to sea salt episodes

Within the ASTA programme (1999 – 2002), the roof project has served two main purposes: to provide information on processes controlling recovery after the initial rapid phase; and to serve as a basis for model testing and development.

The covered catchment experiment was started in April 1991 when a plastic roof was constructed over a micro-catchment near Lake Gårdsjön, SW Sweden (Figure 4-1). The overall objective was to experimentally determine the rate of recovery of a severely acidified forest soil after a drastic cut in input of acidifying input. The main research tool was monitoring of run-off chemistry but soil water and soil content of base cations and sulphate was also monitored. The plastic roof was dismounted in the summer of 2001 after 10 full years of experimental work.

Figure 4-1 The Roof Experiment at Gårdsjön. View from under the roof.

4.3.2.1 SULPHUR ISOTOPES

Sulphur exists in the form of two dominating stable isotopes in the environment, ^{32}S and ^{34}S. Although the basic chemical characteristics of the different isotopes are similar, some discriminatory chemical and biological processes exist. These selective processes can lead to an accumulation of specific isotopes in *e.g.* different geological materials, in different soil horizons, in seawater and in any other part of the environment. This means that sulphur from different origins can have distinctly different isotope composition and can thus be separated using mass spectrometry. In the case of the roof experiment, the isotopic composition of the sulphur added in the irrigation water (originating from additions of seawater) differed from the sulphur present in the soil. By monitoring the change in sulphur isotope distribution in runoff, in the organic soil and in the mineral soil it was possible to obtain information on sulphur cycling in the catchment.

4.3.3 National surface water and catchment monitoring programmes

Just as Gårdsjön is a leading international example of an intensively studied ecosystem, Sweden's national surface water and catchment monitoring programs are a notable example of how to seek a comprehensive national picture of environmental status and human influence. This is achieved with a co-ordinated set of programmes that monitor at different spatial and temporal scales, ranging from national surveys of lakes (ca 4000) and water courses (ca 700) every five to six years which define the full spatial variability within the country (**Figure 4-2**), to a set of four integrated monitoring (IM) catchments distributed across the country that seek to capture the full temporal variability of linked biogeochemical cycles with an intensity approaching that of the Roof catchment. (In fact the reference catchment for the Roof Experiment at Gårdsjön is one of these four IM sites.) At intermediate levels of intensity are several hundred reference lakes and watercourses with monthly sampling, and some dozen "PMK" catchments monitored since the early 1980's that were the forerunners of the IM program (Table 4.1).

The ASTA programme has sought to take advantage of these sets of monitoring information (sometimes in co-operation with other research projects) to make a comprehensive assessment of the acidification status in Sweden today and in the past, but especially to predict how different abatement strategies will influence the future of the nation's aquatic resources.

The evaluation of the PMK sites, started with a statistical evaluation of the time series (between 10 and 20 years in the case of reference lakes and the PMK catchments). The developments in soil water chemistry (Fölster and Bringmark, in press), stream water chemistry (Fölster and Wilander, 2002) and catchment output fluxes (Fölster *et al.*, in press) from the PMK sites were also evaluated statistically. In the national lake inventories, the new Environmental Quality Criteria for surface waters were applied to achieve Sweden's first nationally comprehensive assessment of acidification status over a decade.

Several of these data sets were then used in dynamic modelling using MAGIC, including nine of the PMK sites from across the country (Krám *et al.*, 2001a, b). These geographically dispersed sites represent a wide range of soil and surface water responses to acid deposition over the last decades. The reference lakes were also modelled with MAGIC to achieve a still more spatially distributed picture of the acidification history of Sweden, and its potential future, depending on the development of deposition, climate and land use in the years to come (see section 5.2.1.5). In selected cases, paleoecological data were used to constrain and test the MAGIC lake modelling (e.g. Krám et al., 2001 c). The potential for predicting the development of acid episodes by "piggy-backing" the new "Episode model" to Magic predictions of changes in average annual chemistry was also demonstrated.

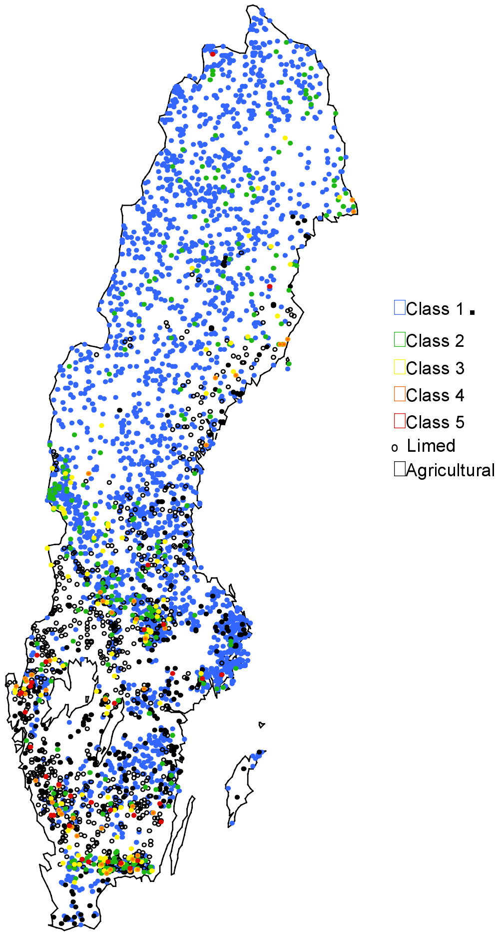


Figure 4-2 The acidification status of lakes from the 1995 National Lake survey when using the Swedish Environmental Protection Agency's acidification index to assess buffering capacity. Between 18 and 25% of Swedish lakes were acidified in this study, depending on the specific assumptions made. Most acidified lakes are found in the south-western part of the country. The classes range from no significant acidification (Class 1) to extreme acidification (Class 5) (Rapp et al., submitted to Environmental Pollution). Limed lakes (open circles) and agricultural lakes (black circles) were not assessed.

Table 4.1 Site characteristics of Swedish PMK reference sites.

Code	Area	Stream	Zone	Forest	lat.	long.	Altitude	Area /km2	Till%	Soil <0.2 m%	Peat %	Lake %	Moraine depth	Soil depth
SE5A	Ammarnäs	Lillbäcken	alp./	subalp birch	65°58´	15°58´	550	2.2	100	0	0	0		
SE5B	Ammarnäs	Raurejukke	alp./	subalp birch	65°58´	15°58´	540	10.5	86	0	10	4	2	1
SE3	Reivo	Laxtjärnsbäck	boreal	conif.	65°47´	19°05´	440	10.9	85	0	14	1	3	2
SE6	Vindeln Svart	Stora dammen	boreal	conif.	64°15´	19°48´	220	2.2	70	0	20	0		
R1	Stenbitshöjde	Höjdabäcken	boreal	conif.	16°56´	19°34´	440	4.9	39	1	40	20		
SE8	Sandnäset	Lilltjärnsbäcke	boreal	mixed	63°46´	12°26´	430	0.5	62	2	24	13	1	1
SE9	Stormyran	Stormyrbäcke	boreal	conif.	62°16´	16°16´	410	3.2	63	0	30	7		
SE10	Tandövala	Lillfämtan	boreal	conif.	60°51´	13°07´	455	5.8	57	15	25	3		
SE11	Tresticklan	Ringsmobäck	boreonemoral	conif.	59°00´	11°45´	180	1.4	1	79	15	5	1	0
SE1A	Tiveden	Lommabäcke	boreonemoral	conif.	58°41´	14°38´	190	1.0	4	84	8	4	1	0
SE1B	Tiveden	Bråtängsbäck	boreonemoral	conif.	58°41´	14°39´	100	7.5						
SE2	Berg	PipbäckenN	boreonemoral	mixed	57°04´	12°48´	75	0.9	64	1	31	4	2	1

The evaluation of these national monitoring data sets have given us a much better understanding of acidification and recovery in Sweden and its regional distribution than was available before ASTA began. It has also given us a more robust scientific basis for the development of a new generation of international pollution abatement strategies.

4.3.4 Modelling the recovery of acidification

In the ASTA programme, two different models for acidification and recovery of soils and surface waters have been used, SAFE and MAGIC. The models have several common features in that they both simulate the changes in chemistry of the soils and soil water (and surface waters in the case of MAGIC) driven by changing deposition, climate and land use. Both models are also calibrated to fit measured data of specific parameters. The time series modelled by both models can be divided into three periods: the historical period; the evaluation period (i.e. the year(s) when measurements data are available) and the forecast period.

An important aspect of both models is that they are *dynamic*. Dynamic used in this context implies that the models are capable of simulating time-dependent processes. This is a large step from *static* models such as models for traditional critical load calculations, where no time-factor is included, i.e. the models do not contain information on when the modelled outcome is expected to occur. By complementing the assessment of the final state to which ecosystems will reach (assuming unlimited time is available) with dynamic assessment, several important question concerning sustainability and final status of ecosystems at different scenarios of deposition and land-use can be answered. However, the dynamic aspect can be rather crucial too. For a number of issues it is important if the ecosystem response is expected to occur in a year or in a thousand years. The dynamic models can be used for describing and explaining a historical process and for providing policy-relevant information on the results of different scenarios of emissions in the future.

4.3.4.1 SAFE - BRIEF DESCRIPTION

SAFE is a dynamic, multi-layer, soil chemistry model, developed with the objective of studying the effects of acid deposition on soils and ground water. It can be used to study the process of acidification and recovery, as affected by e.g. deposition rates, soil parameters and hydrological variations. A detailed description of the SAFE model can be found in Alveteg (1998).

The SAFE model includes process-oriented descriptions of cation exchange reactions, chemical weathering of minerals, leaching and accumulation of dissolved chemical components and finally solution equilibrium reactions involving CO_2, organic acids and Al-species. As for nitrification, SAFE assumes all ammonium to be either taken up or nitrified in the top soil layer. SAFE, which treats calcium, magnesium and potassium as a lumped divalent base cation (Bc), also includes the effects of

- deposition,
- uptake of N and Bc from soil solution,

- nutrient cycling of N and Bc as litterfall and canopy exchange,
- net mineralisation and/or immobilisation of N, Bc and sulphate,

with fluxes specified as time-series either by the model user or by a separate model, e.g. the MAKEDEP model for deposition. A schematical sketch of the SAFE model framework is presented in Figure 4-3.

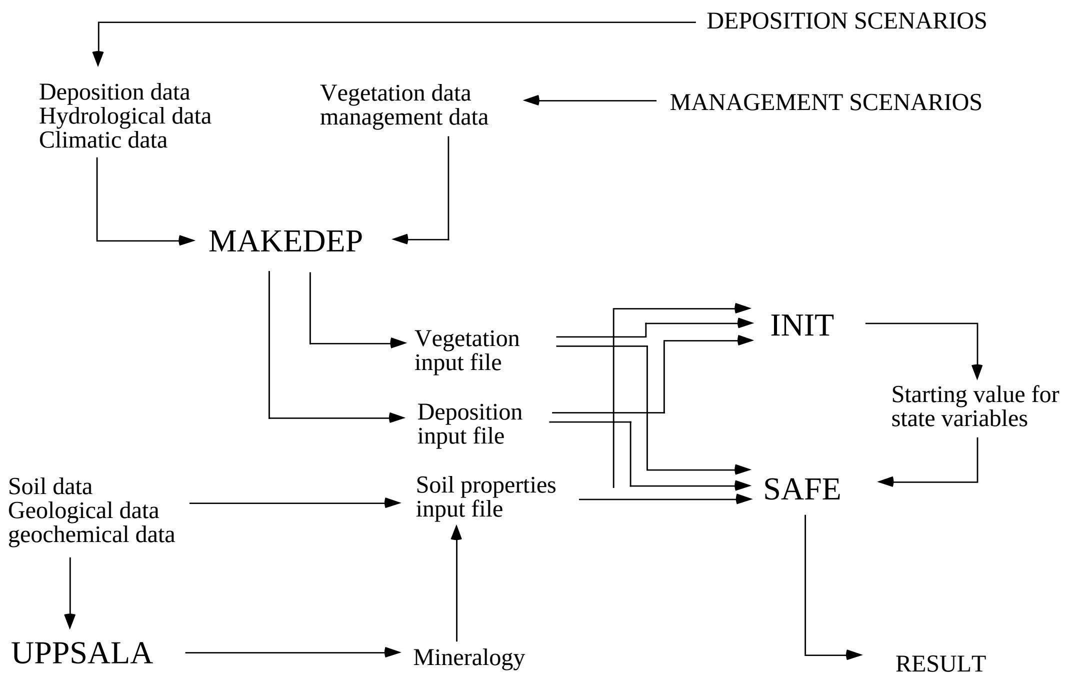


Figure 4-3 Dynamic assessment with use of MAKEDEP and SAFE models.

Of course, the included processes only represent a selection of naturally occurring processes in the soil. Among processes that have not been included are sulphate adsorption and a series of reactions that may change the cation exchange capacity (CEC) of the soil matrix, store sulphur irreversibly or affect the acid neutralising capacity (ANC) balance in certain soils. All processes included in the model have been subject to necessary simplification in some respect:

- The soil is considered to be a series of continuously stirred tank reactors (CSTR) where each tank reactor represents one soil layer. Thus, every soil horizon is assumed to be homogenous. In reality, however, the soil is heterogeneous with its many roots, stones, micro-organisms, etc.

- It is assumed that all changes with depth in physical and chemical characteristics are discrete. In reality there is often a gradual visual change in the enrichment zone.

- Every soil horizon is assumed to be perfectly mixed. This is approximately true for the uppermost part of a coniferous forest soil and for the upper part of a deciduous forest soil, where nematodes and alike move things around. For other soil horizons, this must be considered as a simplification.

- The water flow path is assumed to be downwards only. This is approximately true for areas where the potential evapotranspiration is less than the rainfall, but in areas where the potential evapotranspiration exceeds the annual rainfall, upward flow (capillary flow) may be of great importance

- The distribution of uptake with depth is assumed constant over time. In addition, the cation exchange capacity (CEC) is assumed constant.

Division into different soil layers can be done based on available soil chemistry data for different depths in the soil profile, the preferred principle being compartments that correspond to the natural soil stratification, as the soil horizon is the largest chemically isotropic element in the system. In each layer, the water flow is divided into evapotranspiration, horizontal flow and the residual which continues downward to the next layer. The hydrology is currently assumed to be constant and may be calculated using a separate hydrological model or water-flux measurements. The change in soil solution chemistry and subsequent changes in the distribution of elements on the cation exchange matrix are calculated by means of mass balance equations.

The SAFE model requires data on soil properties, such as mineralogical composition of the soil, soil texture, cation exchange capacity and base saturation. Time series of data are needed concerning precipitation, atmospheric deposition of major ions, nutrient uptake, nutrient cycling and net mineralisation.

Models generating input to the SAFE model, e.g. the MAKEDEP model, may be of any level of detail and of any temporal resolution, the commonly used resolution being yearly averages. To ensure model stability, calculations should start from stable conditions, thus well before the acidification process started. SAFE is therefore usually initiated as early as 1800-1850.

The initial base saturation is calibrated by shooting until simulated current base saturation is in agreement with measurements of base saturation. SAFE calculates weathering rate rather than calibrating weathering rate using measurements of soil chemistry. Measurements of soil chemistry can thus be used for validation of a SAFE simulation.

Many of the simplifications in the SAFE model may be refined in future versions: A decomposition submodel have been constructed that could be coupled to future versions of SAFE. For sulphate adsorption a model has been developed and tested at the roofed catchment at Gårdsjön. Furthermore, a simple hydrological model might be included in future versions of SAFE. As model refinement might affect model applicability, some future refinements might be made optional or only be included in special versions of the SAFE model.

4.3.4.2 MAGIC - BRIEF DESCRIPTION

A number of mathematical models of soil and surface water acidification in response to atmospheric deposition were developed in the early 1980's (e.g., Christopherson and Wright 1981; Christopherson et al. 1982; Schnoor et al. 1984; Booty and Kramer 1984; Goldstein et al, 1984; Cosby et al. 1985a,b,c). These models were based on process-level information about the acidification process and were built for a variety of purposes ranging from estimating transient water quality responses for individual storm events to estimating chronic acidification of soils and base flow surface water. One of these models, MAGIC (the Model of Acidification of Groundwater In Catchments; Cosby et al. 1985a,b,c), has been in use for more than 15 years. MAGIC has been applied extensively in North America and Europe to both individual sites and regional networks of sites, and has also been used in Asia, Africa and South America. The utility of MAGIC for simulating a variety of water and soil acidification responses at the laboratory, plot, hill slope and catchment scales has been tested using long-term monitoring data and experimental manipulation data

Model Description.

MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on surface water chemistry. The model simulates soil solution chemistry and surface water chemistry to predict the monthly and annual average concentrations of the major ions in these waters. MAGIC consists of:

1) a section in which the concentrations of major ions are assumed to be governed by simultaneous reactions involving sulphate adsorption, cation exchange, dissolution-precipitation-speciation of aluminium and dissolution-speciation of inorganic carbon; and

2) a mass balance section in which the flux of major ions to and from the soil is assumed to be controlled by atmospheric inputs, chemical weathering, net uptake and loss in biomass and losses to runoff.

At the heart of MAGIC is the size of the pool of exchangeable base cations in the soil. As the fluxes to and from this pool change over time owing to changes in atmospheric deposition, the chemical equilibria between soil and soil solution shift to give changes in surface water chemistry. The degree and rate of change of surface water acidity thus depend both on flux factors and the inherent characteristics of the affected soils.

Cation exchange is modelled using equilibrium (Gaines-Thomas) equations with selectivity coefficients for each base cation and aluminium. Sulphate adsorption is represented by a Langmuir isotherm. Aluminium dissolution and precipitation are assumed to be controlled by equilibrium with a solid phase of aluminium trihydroxide. Aluminium speciation is calculated by considering hydrolysis reactions as well as complexation with sulphate and fluoride. Effects of carbon dioxide on pH and on the speciation of inorganic carbon are computed from equilibrium equations. Organic acids are represented in the model as tri-protic analogues. First-order rates are used for retention (uptake) of nitrate and ammonium in the catchment. Weathering rates are assumed to be constant. A set of mass balance equations for base cations and strong acid anions are included. Given a description of the historical deposition at a site, the model equations are solved numerically to give long-term reconstructions of surface water chemistry (for complete details of the model see Cosby et al., 1985 a-c, 2001).

Magic has been used to reconstruct the history of acidification and to simulate the future trends on a regional basis and in a large number of individual catchments in both North America and Europe (e.g., Beier et al., 1995; Cosby et al., 1990, 1995, 1996; Hornberger et al., 1989; Jenkins et al., 1990; Lepisto et al., 1988; Whitehead et al., 1988, 1997; Wright et al, 1990, 1994; Norton et al., 1992).

Model implementation.

Atmospheric deposition and net uptake-release fluxes for the base cations and strong acid anions are required as inputs to the model. These inputs are generally assumed to be uniform over the catchment. Atmospheric fluxes are calculated from concentrations of the ions in precipitation and the rainfall volume into the catchment. The atmospheric fluxes of the ions must be corrected for dry deposition of gases, particulates and aerosols and for inputs in cloud/fog water. The volume lake discharge for the catchment must also be provided to the model. In general, the model is implemented using average hydrologic conditions and meteorological conditions in annual or seasonal simulations, i.e., mean annual or mean monthly deposition, precipitation and lake discharge are used to drive the model. The model is not designed to provide temporal resolution greater than one month. Values for soil and lake temperature, partial pressure of carbon dioxide in the soil and lake water and organic acid concentrations in soil water and lake water must also be provided.

As implemented in this project, the model is a two-compartment representation of a catchment. Atmospheric deposition enters the soil compartment and the equilibrium equations are used to calculate soil water chemistry. The water is then routed to the lake compartment, and the appropriate equilibrium equations are reapplied to calculate lake water chemistry.

Once initial conditions (initial values of variables in the equilibrium equations) have been established, the equilibrium equations are solved for soil water and lake water concentrations of the remaining variables. These concentrations are used to calculate the lake discharge output fluxes of the model for the first time step. The mass balance equations are (numerically) integrated over the time step, providing new values for the total amounts of base cations and strong acid anions in the system. These in turn are used to calculate new values of the remaining variables, new lake discharge fluxes, and so forth. The output from MAGIC is thus a time trace for all major chemical constituents for the period of time chosen for the integration.

Calibration Procedure.

The aggregated nature of the model requires that it be calibrated to observed data from a system before it can be used to examine potential system response. Calibration is achieved by setting the values of certain parameters within the model, which can be directly measured or observed in the system of interest (called “fixed” parameters). The model is then run (using observed atmospheric and hydrologic inputs) and the output (lake water and soil chemical variables, called “criterion” variables) are compared to observed values of these variables. If the observed and simulated values differ, the values of another set of parameters in the model (called "optimised" parameters) are adjusted to improve the fit. After a number of iterations, the simulated-minus-observed values of the criterion variables usually converge to zero (within some specified tolerance). The model is then considered calibrated. If new assumptions (or values) for any of the fixed variables or inputs to the model are subsequently adopted, the model must be re-calibrated by re-adjusting the optimised parameters until the simulated-minus-observed values of the criterion variables again fall within the specified tolerance.

Calibrations are based on annual fluxes for a given period of observation. The length of the period of observation used for is not arbitrary. Model output will be more reliable if the annual flux estimates used in calibration are based on a number of years rather than just one year. There is a lot of year-to-year variability in atmospheric deposition and catchment runoff. Averaging over a number of years reduces the likelihood that an "outlier" year (very dry, etc.) is the primary data on which model forecasts are based. On the other hand, averaging over too long a period may remove important trends in the data that need to be simulated by the model.

The estimates of the fixed parameters and deposition inputs used to calibrate the model are subject to uncertainties so a "fuzzy" optimisation procedure was implemented for calibrating the model. The fuzzy optimisation procedure consisted of multiple calibrations of each catchment using random values of the fixed parameters drawn from the observed possible range of values, and random values of deposition from the range of model estimates. Each of the multiple calibrations began with (1) a random selection of values of fixed parameters and deposition, and (2) a random selection of the starting values of the adjustable parameters. The adjustable parameters are then optimised using a numerical algorithm to achieve a minimum error fit to the target variables. This procedure is undertaken ten times for each lake. The final calibrated model is represented by the ensemble of parameter values and variable values of the successful calibrations.

The results discussed in this report are based on the **median** values of the simulated water quality variables for each lake for any given year. The use of median values for each lake assures that the simulated responses are neither over- or underestimates, but approximate the most likely behaviour of each lake (given the assumptions inherent in the model and the data used to constrain and calibrate the model).

4.3.5 Soil (and stand) inventories

Assessing the status and future dynamics of forest soil chemistry in Sweden is data intensive. So far, the main source of information regarding soil and stand properties is the Swedish Forest Inventory database (Ståndortskarteringen). The three factors currently limiting the number of sites for which the assessment can be made is availability of data on 1) mineralogy, 2) exchangeable cations and 3) current standing biomass. In a steady-state assessment such as the calculation of critical loads, data on exchangeable cations is not needed and long-term nutrient uptake rather than current biomass is needed.

Data on mineralogy is currently available for slightly less than 2000 sites and critical loads have consequently been calculated for 1883 forest sites. As for exchangeable cations, measurements have been made for a large number of sites. Many of these measurements are, however, just on the verge of being incorporated in the database.

A problem in using the information on exchangeable cations is that the relation between these measurements and the soil stratification is not clear. Information on the presence and thickness of different soil layers is unfortunately somewhat minimised in the current databases. In the critical loads calculations a rather simple scheme was used to deduce soil layer stratification from available data. In a dynamic assessment, however, the soil layer stratification must relate to the available measurement on exchangeable cations. Translating the available information into a layer stratification where each layer has a certain thickness and a certain amount of exchangeable cations has proven to be one of the major difficulties in the dynamic assessment. The database for regional dynamic assessment currently consists of data for 700 Swedish forest sites.

The current standing biomass is used in combination with a logistic growth curve to estimate how nutrient uptake has and will change over the years using the MAKEDEP model. Since site specific information on nutrient content is not available and since nutrient content changes with nutrient availability literature data on minimum and maximum nutrient contents for different species is used. In spite of the possibility for MAKEDEP to use the minimum literature values on nutrient content the reconstructed deposition is often not enough to explain the nitrogen bound in standing biomass. This indicates that there is a missing source of nitrogen in the model concept. One such source could be deposition of organic nitrogen. The fate and flow paths of organic nitrogen are unfortunately poorly understood and therefore difficult to incorporate in dynamic models.

4.4 Eutrophication

4.4.1 Introduction

Elevated levels of N deposition occur over large areas and a wealth of studies have described potential effects of this N input. Still, however, disentangling the role of N from that of other environmental changes and natural gradients in altering the biota is difficult. One difficulty is that deposition started with small doses decades ago and thereafter increased and accumulated in the ecosystem making it almost impossible to define a true pristine system. Other difficulties are that soil processes and vegetation development may respond linearly to an increased pollution pressure, or abruptly at certain threshold levels, and there may be a certain lag-time before responses occur. Also, experimental studies involving additions of N has mostly been carried out in areas already affected by N deposition. This means that the effects studied are secondary to those promoted by the initial N deposition. Rates of N input needed to produce measurable effects, and the suggested critical loads for N, may thus be overestimated.

Because N availability has such a critical role for structuring plant communities and regulating rates of biomass production in ecosystems, N inputs are bound to have severe effects. The complexity of the N cycle and the large number of processes affected by N supply, constrain our possibilities to describe and assess effects of N deposition on ecosystems. Thus, models of N impact on vegetation will always contain large uncertainties as will estimates of critical loads for N. In the end, our possibilities to estimate the levels of N producing negative effects on ecosystems will depend on our knowledge of the processes through which N deposition acts on these ecosystems. A profound knowledge of N effects on ecosystems will also help establishing new tools for surveying N deposition effects on ecosystems.

Forest ecosystems of the nemoral and boreal regions are typically N limited and N inputs into these forests have repeatedly been shown to increase tree growth. The understorey vegetation of these forests is affected by a number of human activities, including management, acid deposition and nitrogen deposition. Available information suggests the understorey of the nemoral region forests have changed towards a higher fraction of nitrophilous species (Diekmann & Falkengren-Grerup 2002), while similar information for the boreal region is largely lacking.

The above mentioned problems in understanding and quantifying the effects of nitrogen input have also implications for setting critical loads. Critical loads are based on what is meant as harmful effects but the very long period of cultivating soils limits the perspective of ecological effect in many central European nations. In these landscapes there is no patch of land that has not been affected by man's activities. In terms of nitrogen pollution, the levels experienced in central and northern Sweden during the latest decades, was surpassed more than 100 to 300 years ago. The type of ecosystems we want to protect in Sweden disappeared from these central European regions perhaps centuries ago. The people of these regions, including many scientists, cannot visualise or remember this former status of their ecosystems. The discussion on what we mean with critical loads for eutrophication will therefore depend very much on the type of landscape we want to achieve and protect. For Sweden this may mean that we must look for our own standards for setting critical loads for nitrogen.

Eutrophication cannot be seen as a problem isolated from acidification – or even climate change – and model approaches to assess eutrophication effects need to be coupled to acidification and climate. This aspect is focus for particular interest in the ASTA programme and is taken up in section 4.5.

4.4.2 Approaches taken – experimental studies and critical loads

4.4.2.1 EXPERIMENTAL NITROGEN RESEARCH

An important objective of the ASTA programme is to improve the scientific knowledge of how N deposition affects plant communities and ecosystem processes. The only reliable method to do this is to use experiments where manipulations are performed in a controlled fashion so clear cause-effect relationships can be illuminated. Fertilisation experiments are, however, unable to cope with the long term aspect of pollution and they also have difficulties in showing varying biotic and abiotic effects, as these time-consuming experiments must be limited to a few sites. In the ASTA programme results from experiments have been combined with efforts of extensive monitoring. Thereby, processes identified to be of central importance for vegetation dynamics in the experiments, have been studied and shown to be applicable at the landscape level. Also, extensive monitoring may more effectively than experiments discern the effects of acidification and eutrophication on soil and vegetation and refer the background noise to other causes.

With this scientific approach we wanted to gain new knowledge about the underlying mechanisms through which N alters the vegetation and describe how the understorey of different forest ecosystems is affected by historical and current N input. We wanted specifically to describe:

- Vegetation responses to increased N deposition
- Vegetation responses to different soil N forms (NO_3^-, NH_4^+, organic N)
- Ecosystem recovery following decreased N deposition
- Relationships between historical and present rates of N deposition, and vegetation and soil processes
- Assess the importance of the understorey for decreasing N leakage

4.4.2.2 CRITICAL LOADS

The work on nitrogen has been characterised by the acknowledgement that it is necessary to work at three levels simultaneously:

1. Empirical critical loads, based on observations and short term experiments
2. Mass balance based critical loads
3. Biodiversity related critical loads, based on quantitative relations between nutrient balances and ecological competition.

The reason for this three-partite approach is that the different approaches come to results at different speeds. It has been recognised that the existing empirically set critical loads have to a large extent been based on short-term experiments from central Europe and the British Isles, where the results have may be questioned:

1. The empirical experiments from central Europe are generally short term (1-5 years), and the extrapolation to longer periods (50 to 200 years) is scientifically questionable and in most cases unable to use in slow reacting cold systems.
2. There are no adequate tools for transferring results from central Europe or western British Isles to the Scandinavian semi-arctic conditions.
3. The pollution input in the experiments is much higher than what is of interest in Scandinavia.

The empirical method gives approximate results, but do so fairly quickly, especially if older experiments are used and reinterpreted for these purposes. In order to make a first approach to approximate results, we initiated some empirical studies and a survey, in parallel to the development of mass balance models.

There are mass balance methods available, and these methods are able to predict risks for N leaching. They have also been linked to carbon cycle modelling and forest growth predictions in the SUFOR programme. The present linking to ecological effects are weak and only indirect through effects on forest long term nutrition and on surface water quality. The earlier used model SAFE is evolving into the FORSAFE model, which is estimated to become operable during 2002.

Even if the biodiversity-related critical loads have a strong link to ecological effects of a kind high on the political agenda, there will take time before there will be applicable model tools available. The model development within ASTA will exploit the SAFE and FORSAFE models to generate the forest and soil chemistry environmental charges in the scenarios. The results will be with more details, and we expect them after proper adjustment to be particularly useful for Swedish conditions but also applicable for European conditions. In this work the empirical studies will be recycled into the parameterisation efforts, and thus be of double value.

Thus, we may conclude at the outset that the different approaches were designed to start as separate efforts, but with the plan to converge on the final integrated model tool, but being able to provide critical loads values of increasing quality during the process. The ecosystem and mass balance approaches will be further elaborated in the coming sections.

4.4.3 Mass balances

For Sweden, critical loads for nitrogen have been based on simple principles for mass balancing where limits are set to protect against eutrophication of the plant ecosystem. The important components of these calculations are uptake and removal by harvest, immobilisation in the system and denitrification. The calculation of critical loads for nitrogen is more difficult and less accurate than the corresponding calculation for sulphur. The largest problem is posed by the estimation of the relevant rate of immobilisation as well as to establish a good connection between the presence of nitrogen and adverse ecosystem response. The establishment of critical limits based on leaching, concentrations or contents of nitrogen remain unsatisfactorily solved.

Nitrogen enters mainly as nitric acid and ammonium and this has the potential to create large amounts of acidity in the soil. At the same time, the presence of a nutrient that normally is a limiting factor changes many fundamental conditions for plant competition and vegetation composition may change drastically. The purpose of critical loads for nitrogen is to set safe deposition limits in such a way that undesirable changes will not occur. The effects stated as undesired are vegetation changes, promoted forest growth beyond the sustainability potential and nitrate leaching with subsequent acidification of surface waters. The vegetation changes have been seen as very serious effects because they seem to occur over large areas, with effects on both rare and common species. The impact on forest ground vegetation is normally manifested as disappearance of lichen vegetation, *vitis-idea*/*erica* vegetation is changed to *vaccinium*, *vaccinium* is changed to grass, grass change to nitrogen favoured herbs.

Critical loads for nitrogen based on the mass balance method is put up in order to estimate the maximum input that can be allowed based on a critical N concentration in leaching water and important terms of removal. The removal terms are uptake to trees that become harvested, immobilisation in the soil into long-term stable pools and denitrification by soil micro-organisms. Other sources or sinks like forest fires, fixation or uptake in animals that subsequently leave the territory are much more ephemeral and in most cases insignificant in forest soils in Sweden. The critical N concentration leads to a critical leaching of N and makes a mass balance approach possible for calculating critical loads.

$CL(N) = N_u + N_{im} + N_{de} + N_l$

where N_u is the rate of N uptake (or removal by harvest), N_{im} is the rate of N immobilisation, N_{de} is the rate of denitrification, N_l is the rate of critical leaching of nitrogen, Uptake can be present or the critical uptake as calculated using the nutrient limiting method (Warfvinge et al, 1992). The maximum permitted N leaching is estimated from a plant/ecosystem-specific critical concentration. Denitrification and immobilisation are important components of the mass balance, however, they have been difficult to estimate. At present, no accepted method exists for estimating the denitrification and (acceptable) long-term immobilisation of N on a regional scale. Denitrification is normally calculated as a function of the net input of N to the soil, but the proposed methods to estimate the fraction denitrified in critical loads calculations give different results. Values between 0.5 and 1.0 kg N ha^{-1} yr^{-1} are recommended for immobilisation in the critical loads work under the LRTAP Convention (UNECE, 1996), but other methods to calculate more site-specific immobilisation, based on studies by Ineson et al, 1996 and Nilsson et al, 1998 have been proposed.

A general limitation with the calculation of critical load of N with steady state mass balances is that the factors on the right hand side in the equation above are assumed to be independent of the deposition, which is not the case. An increase of the deposition of N will normally influence for example the uptake in vegetation in N limited forests. This implies the need of dynamic modelling of N in the future to improve the long-term predictions of impact of N deposition on forest ecosystems.

4.4.4 Modelling effects of acid and nitrogen deposition on ground vegetation

4.4.4.1 INTRODUCTION

The decline in biodiversity is a complex ecological effect of environmental pollution, climate change and man-made landscape changes. The conditions for the biodiversity are given by geochemical and climatologic conditions, as well as the resource status of a system, and are in constant change. Preservation of the present status is less important than the preservation of the complexity and convergent system dynamics. Biodiversity is one of the most difficult environmental problems to predict and describe in a way that allows design and planning of mitigation strategies. Preserving biodiversity and practising sustainable forestry are global aims stated in many recent international resolutions. Consequently, the issue of integrating environmental values and multiple benefits of forests into practical forest management has gained wide interest.

Biodiversity management has often focused most effort on saving high-profile endangered species. The problem with this species by species approach is that it is expensive and often unsuccessful when there are no habitats left for the endangered species. An alternative approach to regional biodiversity management has been to develop a comprehensive inventory of the status of specific taxa, indicators or species in a region, and through information systems, negotiation and land planning, develop management strategies.

Man intervenes in forest ecosystems in a wide variety of ways, thus changing their structure and dynamics. This use is linked to ecological, economic and social processes and their complex interactions. In order to preserve certain characteristics of an ecosystem (e.g. its profitability or biodiversity), adequate land use strategies must be drawn up. In this connection, model simulations can serve as a sort of substitute experiment. They can be used to test which of the land use strategies examined is most sound and sustainable. Specifically, the biodiversity dynamics of forest ecosystems depends on three characteristics of the landscape:

- **Structure -** the spatial relationships among the distinctive ecosystems or "elements" present--more specifically, the distribution of energy, materials, and species in relation to the sizes, shapes, numbers, kinds, and configurations of the ecosystems.

- **Function -** the interactions among the spatial elements, that is, the flows of energy, materials, and species among the component ecosystems.

- **Change -** the alteration in the structure and function of the ecological mosaic over time.

Biodiversity refers to the different kinds of plants, animals and other living organisms in all their forms and levels of organisation that exist in an area. It includes:

- **Genetic diversity** - The distribution of genetic variation occurring in a particular population of a species.

- **Species diversity** - The number of species inhabiting a certain area and the amount of each species in that area.

- **Ecosystem diversity** - The number of *different* species inhabiting a particular area, and relates to the variety of habitats, biotic communities, and ecological structural processes, and the variety of ecological processes.

- **Landscape diversity** - The number of *different* species inhabiting *different* ecosystems in a particular geographical region, and relates to the diversity of ecosystem types in that area, their geographical extent, geographical connectedness and population stability robustness when subject to dynamic changes.

The aim of the modelling approach under development in ASTA is to integrate feedback mechanisms and cause/consequence relationships between soil acidification, nitrogen eutrophication and climate variation over the territory on ground vegetation occurrence, through further development and integration of existing mathematical models.

4.4.4.2 THEORY

Initially it is proper to present the operational definition of biodiversity, which is partly applied in our model. We propose:

1. The quantity of biodiversity is defined by species variation, population size of each species as well as their geographical structural arrangement.

2. The quality of biodiversity is determined by the coincidence of the present species as compared to the potential of species and the genetic stability within each species

A biodiversity index may include several components:

- a sum of all present species as a fraction of all species that could potentially be present,

- a relation to the geographical amount of each species times the ecological value of that species, and the number of geographical locations available,

- a relation to the difficulty of genetic communication with the next geographical occurrences of that particular species.

This is intended as a final and for all valid definition, but here it will be used as a point of departure, in order to define a modelling objective. For modelling purposes, an equation containing these components has to be defined which would be generally applicable to a single landscape element. To calculate the biodiversity index for a landscape, we would have to apply the equation in each landscape polygon and sum up over all polygons. In this modelling exercise this has not yet been tested on field data. For ground vegetation, the system is outlined in Figure 4-4. The diagram does not describe the biodiversity but rather the machinery of a single plant inside a plant community. The biodiversity can be calculated when all such systems for each plant or plant class present are interplayed in a mathematical model.

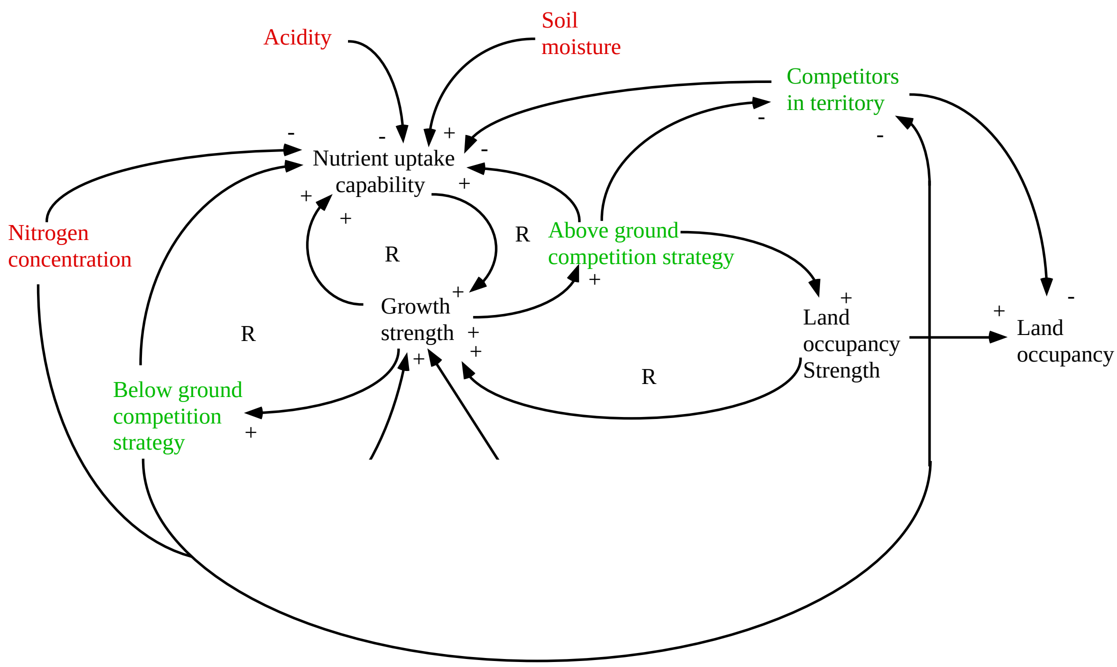


Figure 4-4 A causal loop diagram for the interactions of the ground vegetation with environmental factors. The diagram represents those factors we ultimately intend to include in a future assessment model. The figure also shows the factors that need to be entangled when going from field observations back to changes in nitrogen deposition. The model will be used with the different data sets available in order to bridge this gap.

4.4.4.3 Methods

The basic method is to conduct systems analysis for causal links and quantify these in a dynamic model. The effects on biodiversity on a spatial scale will be modelled by the combination of soil, vegetation and climatic change models including hydrology.

The tasks to be performed performed in this approach include:

- Extend and adapt a ground vegetation response model by assessing ecological response surfaces of plant species for variables related to climate change, soil acidification, eutrophication by nitrogen and desiccation for a subset of species arranged into functional groups.

- Calibrate and validate biodiversity impact assessment models and the integrated assessment tools on multiple single sites, using the Swedish Forest Inventory database, SUFOR research sites, Integrated Monitoring sites and additional regional data.

- Investigate effects of plausible scenarios for climate change and acid deposition on soil biogeochemistry, forest growth and vegetation change.

The occurrence probability of selected groups of plant species (plant functional classes) will be calculated as a result of the environmental loads and changes in the region. The species response surfaces will be used to assess ecological thresholds to climate change, acidification, desiccation and eutrophication by means of a risk assessment. Two similar model proposals exist and these will be merged to optimise the use of existing know-how. (Hansson, 1995).

Regional integration of biodiversity conservation and forestry production requires land-use allocations based on trade-offs between ecological and agricultural/forest production aspects. A number of properties need to be parameterised and coded into an internal model vegetation property database;

- Establish the response functions for acidifying pollutants (several exist already, partly in Sverdrup and Warfvinge 1993, Ellenberg et al., 1992, Falkengren-Grerup 1992, later published works from the Plant Ecology Department and other unpublished data within the Plant Ecology Department archives), nitrogen responses (several exist, others can be derived from Ellenberg et al., (1992) indices by a newly invented method, the experiments within ASTA, reinterpretation of experiments from Plant Ecology in Lund), temperature (several exist, others can be derived from existing information, data from horticultural trade is commercially available) and water availability (many exist, others can be derived from existing information, data from horticultural trade is commercially available).

- Parameterise the fundamental below ground competition strategies for nutrients, and apply it to the plant classes established. Subsequently to this, establish the fundamental aboveground competition strategies in a mixed community and apply it to the plant classes established.

- Establish a practical amount of plant classes that can be parameterised and with sufficient biodiversity relevance as indicators for a whole ecosystem (this knowledge is available in the Plant Ecology Department and at the Alterra Institute, Netherlands).

We lump plant species into a number of functional plant groups determined by their response behaviour and ecosystem function. A best-available knowledge and estimate the rest approach will be necessary (and in reality the only option). These functional groups contain both trivial plants and red-listed species.

4.4.4.4 MODEL STRUCTURE USED

A model structure was initially developed where the different filters came into play. A nitrogen response function is applied and multiplied with a root competition function, specific for the plant class. This is subsequently multiplied with a retardation function, expressing the effect of soil acidity and the counterbalancing effect of certain base cation concentrations in the soil solution. This product yields the strength of each class as they enter into the competition for light. This gives a final area weighting used to estimate the ground area occupancy fraction of the particular plant class. The existing biogeochemical model SAFE was used to generate chemical time trajectories (Jönsson et al. 1994, Sverdrup and Warfvinge 1993, 1995) and an experimental response module, VEG, was developed (Hansson 1995).

4.4.5 *Mass balance models – national applications*

Tools and models developed and applied within the ASTA programme form the basis of the national platform project aimed at providing support to national policy on sustainability and the environmental objectives. Within the national platform of the ASTA programme, specific efforts are made on assessing different effect criteria for nutrient stability and dynamic modelling of forest soil recovery. Scenarios based on possible development of the sectors energy production and forestry will be used to assess the future role of air pollutants in Sweden. Within the national programme model applications will be developed focusing on areas where both transboundary air pollution and land-use contribute significantly. The national platform will use both simple mass balance approaches and more sophisticated dynamic models such as MAGIC and SAFE. The long-term aim is to develop a toolbox to be used for integrated assessment and policy support for land-use and transboundary air pollution issues. Examples of national issues that can be evaluated by mass balance models are:

- Liming and fertilisation of acid forest soils.
- Ecological restrictions in harvest of forest fuels.
- Compensatory fertilisation (including nitrogen) after harvest of forest fuels.
- The possibilities with forest management reducing leaching of nitrogen.
- The impact of forest management on critical load in Sweden.
- The impact of forest management on recovery from acidification and eutrophication.

Several components of a Swedish integrated assessment model existed before the ASTA programme, e.g. a model for dispersion and exposure of air pollution as well as well documented databases for the status of land and water. The ASTA programme, together with the Mistra programme RESE, has contributed to a new land use map as a basis for regional model calculations. The different ASTA programmes interact to ensure that any model tools developed will be relevant for both national and international applications. Besides ASTA (e.g. SAFE and MAGIC) model development and application in the SUFOR (e.g. FORSAFE) and LUSTRA (e.g. COUP) programmes are included in the toolbox that is used for integrated assessment in Sweden.

The development of tools in ASTA for integrated assessment and predictions of future acidification and eutrophication in forests comprises three main components:

- Regional assessment with simple (steady state) mass balance models of interactions between air pollutants and forest management in Sweden. The calculations is based on national information on deposition, soil properties, forest conditions and run off with comparably high resolution (e.g. grids down to 0.5*0.5 km). The results of the mass balance calculations indicate present imbalances (or balance) with a regional resolution expressed as the flux of acidity, base cations and nitrogen in managed forests.

- Assessment of dynamic consequences (time dependent) of imbalances of acidity and nutrient flux in forests in Sweden. Different realistic scenarios of future deposition and intensity of forest management are introduced in the calculations. Existing models (e.g. SAFE, MAGIC, FORSAFE and COUP) are evaluated and the need of complementary dynamic description of soil and vegetation processes is investigated.

- Regional dynamic effects concerning acidification and eutrophication in forests impacted by air pollutants and forest management. Methods for regional up scaling of results from dynamic modelling are developed. Regional assessment of the effects of whole tree harvest on the recovery from acidification and eutrophication in Sweden is conducted.

4.5 Ozone and gaseous effects

4.5.1 General approaches for new ozone critical levels

The activities within the ozone sub-project is strongly focussed on providing basic scientific knowledge to support the development of a new generation of ozone critical levels. The new concept, so called Level II, is based on calculations of the actual ozone uptake to leaves and needles as opposed to the current AOT40 criteria which only takes into account concentrations exceeding 40 ppb. The support by the ASTA project has enabled us to use experimental data and put it into the context of constructing ozone flux – response relationships for both agricultural crops and forest trees.

The deadline for these activities is the workshop "Establishing Ozone Critical Levels II" to be organised by Asta in Gothenburg 19-22 Nov 2002, with financial support of ASTA, NMR and the National Swedish Environmental Protection Board. The workshop represents the main opportunity to introduce new Ozone Critical Levels II into the revision of the Gothenburg Protocol 2004/ 2005.

To a very large extent, the data used for the development of stomatal conductance simulation models as well as exposure-response relationships for ozone has been derived from experiments with open-top chambers performed at the field station at Östad säteri, south-west Sweden (Figure 4-5).

Figure 4-5 Open-top chambers used for crop research at Östad.

4.5.2 The scientific approach for generating new ozone critical levels II for crops

The scientific approaches for generating new ozone critical levels II for crops are:

1. to obtain stomatal conductance data from situations with variable climate etc.

2. to use these data to calibrate a multiplicative conductance model

3. to test the model against data

4. to use the model to estimate ozone uptake in different treatments in open-top chamber experiments

5. then to derive flux-response relationships using different ozone flux rate thresholds

6. to compare the flux-response relationships with relationships based on other exposure indices like AOT40

7. to adapt the chosen response relationships to modelling and mapping

8. to identify critical fluxes

4.5.3 Experimental data for generating new ozone critical levels for crops

The data sets, on which our calculations are based are taken from the following experiments:

- the field OTC experiments with cereals, clover and pasture, funded by the National Swedish environment Protection Board, the Federation of Swedish Farmers and the Swedish Council for Forestry and Agricultural Research in 1987-1993,

- the EU-funded ESPACE-wheat programme in 1994-1996,

- the EU-funded CHIP programme on potato 1998-1999 and

- a series of experiments funded by the Swedish Council for Forestry and Agricultural Research in 1997-1999.

We kindly acknowledge our colleagues in the CHIP programme who allowed us to use stomatal conductance and ozone effects data for potato from five different countries in Europe. During 2002 data from two or three other European countries will be used to similarly validate our work with wheat, which so far was based only on Swedish data. From one other country (Belgium) we have already received data covering three years of experimentation. Data from Finland are underway and hopefully data will also be available from Switzerland.

4.5.4 The scientific approach for generating new critical levels II for trees

The scientific approach for generating new critical levels II for trees includes the following steps:

1. generating stomatal conductance and ozone uptake simulation models with parameterisation for different tree species,

2. generating ozone uptake - biomass response relationships from experimental data,

3. scaling ozone exposure; juvenile vs. mature trees,

4. scaling tree response; juvenile vs. mature trees,

5. simplification; to provide the large-scale modellers with a simple, but scientifically robust, methodology.

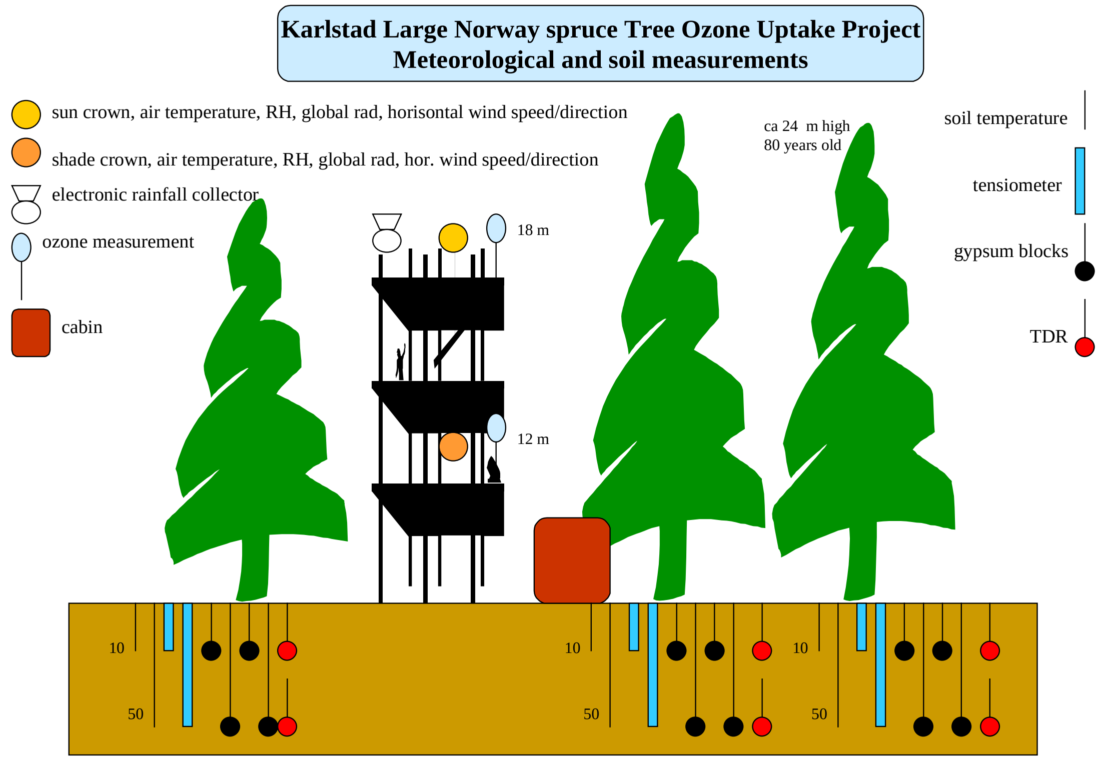
The activities within the ozone forest sub-project will focus on items 1,2,3 and 5 above. Activities within item 4 above are on-going elsewhere (e.g. Kolb and Matyssek 2001).

4.5.5 Experimental data for generating new ozone critical levels for trees

Two multiple-year experiments at Östad have generated experimental data on stomatal conductance simulation models and dose-response relationships for Norway spruce (*Picea abies*) and European silver birch (*Betula pendula*). A four-year experiment 1992-96 with Norway spruce investigated the impact of ozone, with or without a combined drought or phosphorous deficiency stress (Wallin et al., 2002, Karlsson et al, 2002a). This project was mainly supported by the National Swedish Environmental Protection Board and the IVL Foundation. A two-year experiment with Silver Birch 1997-98 (Karlsson et al., 2002d) was funded by the National

Swedish Environmental Protection Board, the IVL Foundation and the MISTRA project SUFOR.

Another experimental facility has been developed with the support from ASTA in collaboration with Karlstad's University. An 18 m high scaffolding has been raised within a mature Norway spruce stand (Figure 4-6). A meteorological station measures the climate, as well as the ozone concentrations, at two levels, at the top and at the middle of the canopy. Soil water availability is measured with several different types of humidity sensors. The stomatal conductance of the shoots is measured at regular intervals using a gas exchange system. A similar project has been initiated in collaboration with the Botanical Institute, Göteborg University, where a sky-lift is used to enable access to the crowns of mature European silver birch trees around Asa Experimental Park in Småland, south Sweden. Climate and ozone concentrations are measured at the meteorological station at Asa and soil water availability is measured close to each experimental tree. The leaf stomatal conductance is measured at regular intervals using a gas exchange system.



Meteorology and soil data are stored every 60 min, ozone data every 30 min. The tower provides access to the canopy of three different Norway spruce trees.

Figure 4-6 Schematic drawing of experimental site for mature trees operated by Karlstad University.

Data from these projects will be available during 2002 and they will enable us to model the stomatal conductance and stomatal ozone uptake to the leaves and needles of mature Norway spruce and European silver birch trees. It will give us the answer to how much ozone is taken up by mature forest trees at different levels of the canopy. The information generated with these experimental activities with mature trees will be useful to predict the ozone impact on mature forest trees. It will also be useful to validate the part of the EMEP ozone model deposition module that simulated the stomatal conductance.

4.6 Particles and human health

4.6.1 Introduction to the problem

Air pollution and specifically ambient particles are currently of great concern to environmental policymakers and scientists. This interest was founded by the discovery of a strong link between air pollution events and increased mortality. Particle mass has been used as a quantitative indicator of the air pollution levels in epidemiological studies. These studies have identified health effects occurring during events of elevated particle mass concentrations. One study claimed that about 40 000 persons in Europe die annually due to air pollution and more suffer other serious health problems (Kunzli et al., 1999). The first estimations of the cost of health problems caused by air pollution point to the particles as the major health problem in monetary units. The costs override by far monetary estimates on other environmental problems caused by air pollution on a European scale.

International and national legislation was already focused on limiting the ambient concentrations of particles when ASTA started in 1999. However, episodes of high concentrations of particles have been found to coincide with high concentrations of many other potential harmful components both in the gas phase as well as the condensed phase, i.e. in particle phase. So from toxic point of view high concentrations of particle mass might only be an indicator of high concentrations on toxic components and legislative action to reduce ambient particle concentrations may not be sufficient to reduce the health impact

Some epidemiological studies suggest that the mass of fine particles less than 2.5 μm in diameter (PM2.5) has a stronger correlation with observed health effects than the particle fraction (PM10) including coarse particles (Dockery et al, 1994). This led to initiatives to establish new measures of air pollution control focusing on limiting the fine particle emissions. PM2.5 is already established as the primary indicator of particle air pollution in the USA, mostly due to technical and traditional reasons. PM2.5 has been taken over for European use and the process of developing a standard is well under way. However, no critical review concerning its suitability from health or control strategy point of view has been undertaken.

It is important to emphasise that in critical assessment reports it is clearly shown that there are several potentially harmful components in an air pollution event and so far, no specific characteristic or type of particles has been shown to be more or less harmful (Pershagen et al., 2000, US EPA 1996).

Many sources influence the mixture of gases and particles and control the formation and characteristics of a specific aerosol type at any geographical location. , This complex nature of particle air pollution has been recognised for long. Despite this, many processes have not been investigated and described quantitatively. The knowledge about long-range transport of air pollutants very much sprung from the research on acidification and led to an understanding that some aerosol components, such as sulphate and nitrate, could be transported quite far. The magnitude of other primary sources and other condensable gases was less well known. Very little research was performed on characterisation of organic components in aerosols. The emissions strengths and transport of resuspended dust was only investigated in limited studies.

This brings us to the starting point of the particle program of ASTA. Legislative authorities were forcing the issue of limitations of ambient particle concentrations but the scientific basis for understanding sources and aerosol dynamics was, to a large extent, missing. At this point, it could not be ascertained that emission reductions would reduce the observed health effects in Europe.

4.6.2 Starting point of ASTA particle program

Fine particles have a lifetime in the atmosphere of days to weeks, depending on size and chemistry. They are thus transported thousands of kilometres before deposition to the ground. During the transport their chemical composition and size change drastically due to condensable gases that are formed and become bound to particles. Particles coagulate, i.e. collide and form aggregates. These processes were qualitatively known but their quantitative influences on different particle characteristics were not well known. However it was known from studies on large scale dispersion of sulphur and nitrogen compounds that the long-range transported part can dominate over locally emitted and formed particles even thousands of km from the source areas. It was thus logical that transboundary long distant transport was a dominating contribution of particle mass in the Nordic countries. Consequently it was necessary to support the introduction of particles in the framework of the EMEP monitoring program. In this effort, providing particle characteristics was important, not only the present focus on particle mass but also other measurements providing information related to sources and transformations of the aerosol.

The present working strategy of EMEP concerning particles was to use the present single component models to add the major components by mass to describe the total particle mass in the two size fractions PM2.5 and PM10. However the mechanisms of formation of secondary organic mass was only coarsely known and the sources of major primary components such as dust and sea salt were not at all described in the model. Consequently, measurements in background areas had to provide information that can be used for source-receptor apportionment of these components.

The dynamic processes that lead to the formation of atmospheric aerosol are often dependent on the number of particles present in different size classes, i.e. coagulation and condensation modes. The chemical composition and the phase of the particles influence absorption and heterogeneous reactions. Furthermore, particle size and chemistry determine e.g. wet deposition. In total these dynamic processes determine the size and chemistry of the particles and thus the deposition rate, human exposure and possible health impact. The available EMEP models at the time described one chemical species at a time, and most of the dynamic processes involving particle phase were described in a simple parameterisation not necessarily based on the involved basic physical and chemical processes. To further increase the accuracy in the predictions of mass the dynamic processes had to be better described.

One of the major remaining questions for EMEP is likely to be the transboundary transport of aerosols, as it is both related to the above mentioned health issue and to the influence of particles on climate. To address these issues, number, surface and chemistry of particles become important output parameters. However, to include dynamic processes in models, a parameterisations is necessary, which in turn, has to rely on basic quantitative knowledge of the involved processes as well as experimental data. Similarly these models had to be tested against measurements showing that the models describe parameter fields in a correct way. A well-developed network of measurement sites with advanced particle monitors describing size, chemistry and different types of particles is thus essential in establishing a model that properly and quantitatively describe the ambient aerosol. Conclusively the insight given by advanced monitoring data is necessary in developing the models as in the end to validate the models.

The particle models currently being developed will provide a possibility to describe the base level over all Europe. The models will give a higher spatial and temporal resolution than the measurements. The local emissions will be superimposed on the European level giving a very detailed description of the air quality including many different parameters besides mass, such as number, surface and chemistry. This very detailed high resolved air pollution description will give the best possible base both for further health studies as well in the assessment work on which sources are giving significant contribution to those parameters found to be hazardous to health.

The approach of the particle program of ASTA can then be summarised in 6 points.

1) Mapping of PM2.5 and PM10 in background to heavily polluted urban areas selected all over Sweden.

2) Detailed investigations of major primary particle sources.

3) Establish and run a network of background stations, with advanced particle instrumentation, representative of different geographical areas defined from sources and meteorology. The distance between the stations has to be large enough to observe transport related transformations. The stations should investigate parameters important both for mass related and dynamic aerosol related models.

4) Develop and test basic information such as parameterisations, both for mass related and aerosol dynamic models, based on the data collected within the network.

5) Implement a dynamic aerosol module in the EMEP-model and have the full model tested against data collected within the network.

6) Initiate and support health studies. Provide information for integrated assessment modelling.

5 Main results

5.1 Communication between science and policy

The ASTA social science sub-programme focuses mainly on the issue of how scientific consensus for international agreements is shaped. The theoretical aim is to improve the understanding of the consensual character of scientific practice, how scientists in policy-relevant situations as experts produce agreed knowledge and how they handle uncertainties, ambiguities and controversies when building credible environmental abatement strategies. Evaluations of international regimes have pointed out the importance to balance scientific credibility and political legitimacy (e.g. Young 1999). Scientific credibility indicates that policy is based on certified knowledge, that scientists have been closely involved in the policy negotiations, and that the scientific community is satisfied with the content of knowledge on which the negotiations is being based. Political legitimacy, on the other hand, indicates that all parties involved – including politicians, NGOs and citizens in the signing countries – consider the result of the negotiations to be important, transparent, understandable and fair. To achieve a result that is considered both scientifically credible and politically legitimate the two worlds of science and policy need to be balanced in a proper way.

This far, our assessment is that the relationship between science and policy is partly misconceived, not least by social scientists. Therefore, a first step has been to evaluate and criticise the dominant understanding of this relationship held by policy analysts (see Section 4.1). Our proposed alternative is based on the assumptions that knowledge never moves freely, that the value of science is the result of negotiations, and that science and policy are co-produced. The relevance and explanatory power of this approach is illustrated in a case study of the LRTAP Convention. By way of conclusion, it is stated that science has no strength in itself but is given strength by different institutions and actors, and this has to be explained by social scientists (Lidskog and Sundqvist 2002).

In the next step the proposed approach is applied in an analysis of the meaning and usage of the critical loads concept in the LRTAP context: how different actors rhetorically utilise the concept (Sundqvist, Letell & Lidskog 2002). The concept of critical loads is located in the borderland between science and policy. Divergent strategies have been identified, in which different actors, through boundary work, have tried to establish specific relationships between science and policy. The result is that the concept of critical loads is assessed differently by involved actors, but all considered it a useful science-based policy tool.

On the one hand, the narrative of critical loads is often articulated and framed as scientific, objective and politically neutral. On the other hand, it is articulated as a useful policy tool to combat transboundary air pollution. In this respect the LRTAP Convention is regarded as being successful because policy-makers have managed to steer science where they wanted it to go. In both cases a gulf is created between science and policy. However, there is also a third line of reasoning, which states that an intimate interrelation between science and policy is a prerequisite for the development of successful abatement strategies. Instead of creating a gulf, it points at an almost blurred interplay between science and policy; the success of LRTAP is explained by a common ground of understanding by all actors involved.

The overall conclusion is that within the LRTAP regime the boundary between science and policy is fluid. The ambivalent attitudes to the concept of critical loads show a flexible use of the science-policy relationship. The success of LRTAP has to do with science, as both the involved actors (natural scientists, negotiators and politicians) as well as policy analysts have stated, but it is science embedded in complex relationships with policy. There has been a lively traffic between the two worlds of science and policy within the LRTAP work. By the use of unifying boundary objects like critical loads the interests from actors with different attitudes could be connected. Thereby actors from different fields of expertise have become part of the same project, focusing on the same objectives, while at the same time being able to utilise different – sometimes competing – interpretations of the scientific status of the concept of critical loads. The concept has been plastic enough to allow different conceptions and at the same time serve as an instrument of integrating different actors.

In a historical study performed within ASTA (Letell 2002), it is shown that the concept of transboundary air pollution, in a way similar to that of critical loads, rhetorically connects the domains of facts and values. The study compares the Swedish case study presented at the UN Conference on the Human Environment (UNCHE) in Stockholm 1972 on regional acidification effects due to transboundary air pollution, with the Swedish national report, "Acidification today and tomorrow", presented at the Stockholm Conference on Acidification of the Environment, held in 1982, partly to celebrate the 10th anniversary of the UNCHE. In the 1982 report it is argued that the acidification now is internationally accepted and that international abatement strategies are necessary, but "ten years ago an appeal on these lines from Scandinavia was seen as exaggerated and largely unnecessary – indeed, almost as a demand for special favours" (SEPA 1981: 8)

It is described how the Swedish articulation of the newly discovered problem of acidification is made on the international arena in the early 1970s. When abatement strategies were advocated for internationally, the transboundary aspect of the problem was circumscribed by particular scientific descriptions, with a varying degree of certainty. However, the concept of transboundary air pollution crystallised and was encircled by discursive elements from chemistry, biology and meteorology and elements familiar within international politics. The problem of acidification was framed as a boundless threat and as a threat of national security. In this sense the concept of transboundary air pollution intervened in the international security discourse of the 1970s. A certain resemblance was assumed between the behaviour of sulphur dioxide and national states. Both "natural" and "social" discursive elements were positioned in relation to each other in such a way that made a re-articulation of the concept of security possible and desirable. This particular discursive configuration gradually gained acceptance and managed to redefine security in such a way that favoured the establishment of an international discourse of transboundary air pollution which later gained stability by an international institutionalisation.

The development of consensual science has been of great importance in scientists' strategies for influencing the negotiation processes of LRTAP. However, experts and negotiators are currently facing a situation of growing scientific and political complexity. Different social and cognitive strategies for reducing scientific complexity run the risk of producing a result that are not seen as credible by policy makers and other stakeholders. Thus, at the same time as the costs of future abatement strategies seem to increase, the possibility to deliver consensual scientific knowledge seems to decrease.

In an on-going ASTA study (Lidskog, forthcoming), the cognitive and institutional strategies for developing scientific certainty are analysed. In this study, it is found that the successfully development of LRTAP has created a new situation, where the involved experts are now pondering on how to introduce new – and costly – abatement strategies at the same time as the consensual scientific knowledge base no longer seems to exist, or at least is questioned.

This situation constitutes a challenge for the experts, and it becomes of central importance to find ways to produce credible science, where the contingency of science is made explicit without eroding the space of action for further development of abatement strategies. A way forward may be to replace the goal of scientific consensus with that of scientific credibility. By this, science is not reduced to the role of knowledge producer, but becomes also a knowledge distributor. Thus, one of the most important roles for science is to be a facilitator of social learning among stakeholders. This perspective on science seems to fit well together with the European Commission's emphasis on the need of democratising expertise (European Commission 2001a).

The study and discussion of the possibility to create scientific credibility is also applied on ASTA's own research network. In a study, the policy implications of recovery of ecosystems from effects of acidification is investigated (Sundqvist, forthcoming). This study is aimed to more closely involve the natural scientists of the ASTA programme in the social science programme. Here it is investigated how natural scientists consider uncertainties as well as policy implications in their work to further develop the concept of critical loads, including dynamic modelling. The point of departure is the ”success story” of the second generation LRTAP protocols based on the concept of critical loads. However, some critical voices have argued that this story is characterised by technocratic decision-making and closed-room expertise. In our study the establishment of the critical loads concept during the 1980s as well as the re-definition of the concept occurring today, including more dynamic aspects of recovery processes, are described. The work of different kind of experts, not at least the scientists of the ASTA programme, is analysed in relation to the newly launched initiative from the EU Commission aiming for "democratised expertise".

The focus in our study is on how scientific uncertainties and policy considerations, such as decreasing public interest, increasing abatement costs and more complicated models, are handled by researchers and experts. In understanding this process and in order to support the efforts of democratising expertise, transparency is not considered the key issue. On the contrary, a too strong focus on transparency could hide the real problem, i.e. the problem of understanding expert strategies in the work of achieving credibility. Transparency is not in itself the key resource for reaching the goal of democratised expertise but has to be combined with an increased understanding of the normally invisible back stage of expert practice. To outsiders, the process of developing shared and credible knowledge could only be transparent if it is combined with increased understanding of the strategies used in specific expert cultures. Transparency as well as credibility is complex social phenomena. To better understand expert work we have to look closer into the social strategies used by specialists in order to control information, as well as the social barriers that regulate access to information, and how front stage and back stage are established in order to manage information control. These strategies can not be made transparent by just opening the doors into the laboratories. What must be added is an increased understanding of expert cultures and expert strategies. Detailed social case studies of expert cultures could play an important role in a unified effort to make the work of experts more transparent and increase the public understanding of expert strategies.

5.2 Acidification

5.2.1 Recovery of ecosystems

5.2.1.1 GÅRDSJÖN ROOF EXPERIMENT

Europe has made great progress during the 1980's and 1990's in reducing acid deposition to levels approaching the "critical load" at which acidification of sensitive ecosystems is not expected to occur (Posch et al., 1997). Sulphur deposition in Sweden probably reached its maximum around 1965. Deposition stayed near these maximum levels for 15 years, until 1980, when the first signs of decline were observed (Bernes 1993). Over the last decade, Sweden and Southern Sweden in particular, have experienced a strong decrease in sulphur deposition (EMEP/MSC-W, 1998). These improvements owe much to the abatement strategies on emissions of SO_2 in Europe that were undertaken in conjunction with the 1979 Geneva Convention on Long-range Transboundary Air Pollution administered by the United Nations Economic Commission for Europe. The international treaties on acid deposition, however, have not explicitly addressed the issue of recovery in areas where soils and surface waters are already acidified.

One effort to better understand the mechanisms and time-scale of ecological recovery when acid deposition is reduced is the Gårdsjön Roof Experiment. In 1988 a small forested headwater catchment was selected, the catchment outlet was fitted with a dam, and runoff sampling commenced. In winter 1990, the catchment was covered with a 7000 m^2 plastic roof (Hultberg and Skeffington, 1998). By preventing ambient precipitation from reaching the soil surface, the roof excluded atmospheric deposition from the headwater catchment G1 ROOF. In parallel to the sampling in G1 ROOF, measurements were also undertaken in a reference catchment F1 CONTROL, located near the experimental area. The G1 ROOF catchment has a closed stand of 80 to 120 year old Norway spruce (Picea abies (L.) Karst.), with some Scots pine (Pinus sylvestris). The bedrock is gneissic with an intermediate to acid chemical composition. The soils are generally thin podzols, and the mean soil depth for both catchments is about 50 cm. The F1 catchment has approximately the same type of soils and forest cover as G1 ROOF.

The deposition in the Gårdsjön area is dominated by sea salt which forms about two thirds of the solutes in ambient throughfall on an equivalent basis. The annual throughfall input of Cl is about 100 kg/ha. The S and N deposition in the area is intermediate relative to that in other areas of Europe. During the 6-year period from 1989 - 1994, F1 CONTROL received an average of 19 kg/ha/yr of SO_4-S in throughfall. About 5 kg of this is of marine origin (calculated from Na^+/ SO_4^{2-} sea salt ratio, assuming all Na^+ is of marine origin), as well as 7 kg of NO_3-N and 4 kg of NH_4-N (Ferm and Hultberg 1998).

The ambient precipitation intercepted by the roof was replaced by sprinkling with the same amount of water that simulates clean throughfall. The roof at Gårdsjön is a physical model. It simulated a situation where deposition of acidifying compounds was reduced to pre-industrial levels. Before the start of the experiment, the G1 ROOF catchment runoff had an annual mean pH of 4.2, an ANC of –260 µeq l^{-1} and an inorganic aluminium concentration (Al^{3+}) of 230µeq l^{-1}. In 1990 the F1CONTROL runoff had a pH of 4.1, an ANC of –140 µeq l^{-1} and an Al^{3+} concentration of 76 µeq l^{-1}. Estimates of weathering at the two catchments (Sverdrup et al., 1998) indicate about a 30% higher rate at F1 CONTROL. This helps to explain the initially lower degree of acidification at F1 CONTROL at the start of the experiment in 1990.

The roof was dismantled after 10 completed years of the clean precipitation treatment, in the summer of 2001.

Following the commencement of the clean throughfall treatment at G1 ROOF, the runoff composition started to change. The first and the most pronounced response was the decline of runoff sulphate, which has continued to decline during the experimental period (Figure 5-1). The rate of SO_4^{2-} decline has been slowing down with additional years of treatment and forms a very smooth curve (Figure 5-1), which can be modelled relatively easily with several common types of isotherm (Mörth et al., 1995).

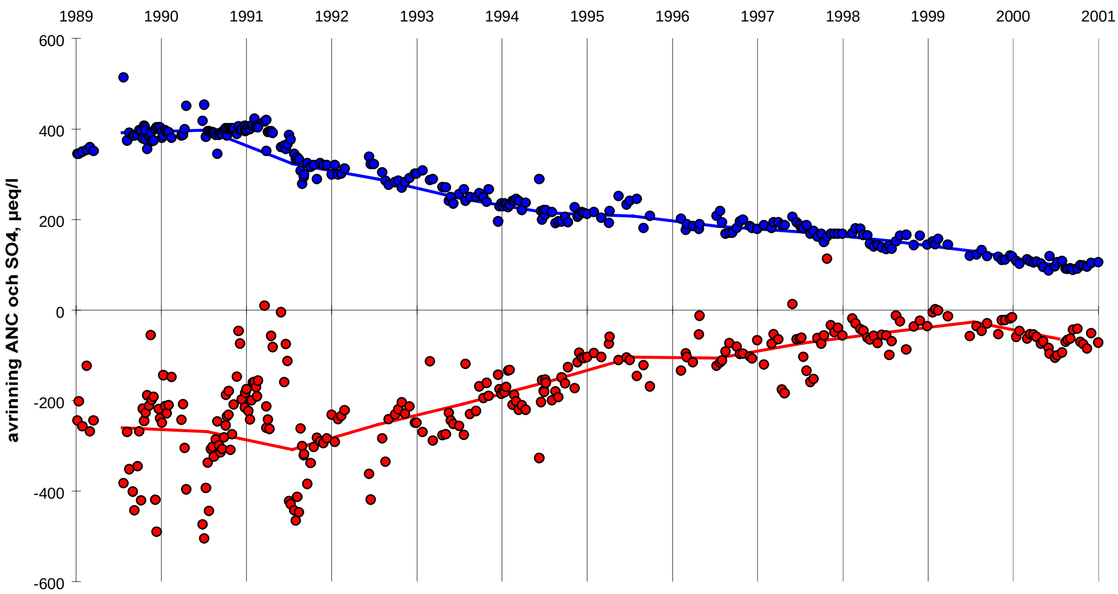


Figure 5-1 Development of SO_4 (top) and ANC (bottom) in the experimental catchment G1. The roof was constructed in spring 1991.

However, the roof project provided evidence that there are processes other than just sulphate desorption that are quantitatively important for the release of sulphur from the soil. The experimental treatment includes addition of seawaterto the sprinkling water to simulate natural sea-salt deposition in the area. The isotopic composition of sea-salt SO_4-S is sufficiently different from both ambient deposition and the isotopic composition of S stored in different forms in the soils that "new" and the "old" SO_4-S can be distinguished in the soil and runoff (Torssander and Mörth, 1998). As the experiment proceeded, it became increasingly clear that there must be another source of SO_4^{2-} within the catchment besides desorption that was also contributing to the SO_4 leaching. Torssander and Mörth (1998) concluded, that net mineralisation of organically bound S was probably this other source.

A number of other major solutes in runoff changed apart from the SO_4^{2-}. The trends of declining SO_4^{2-}, Mg^{2+} and inorganic Al^{3+} and increasing ANC identified after the first years of the experiment (Moldan et al 1995) were confirmed by additional years of treatment. Calcium concentrations continued to decrease. However, the decrease was parallel to the decrease of Ca^{2+} in the runoff at the control site, and therefore it was not identified as a statistically significant change relative to the control. The pH of the runoff also increased slightly at the both the roof and the control sites (Figure 5-2).

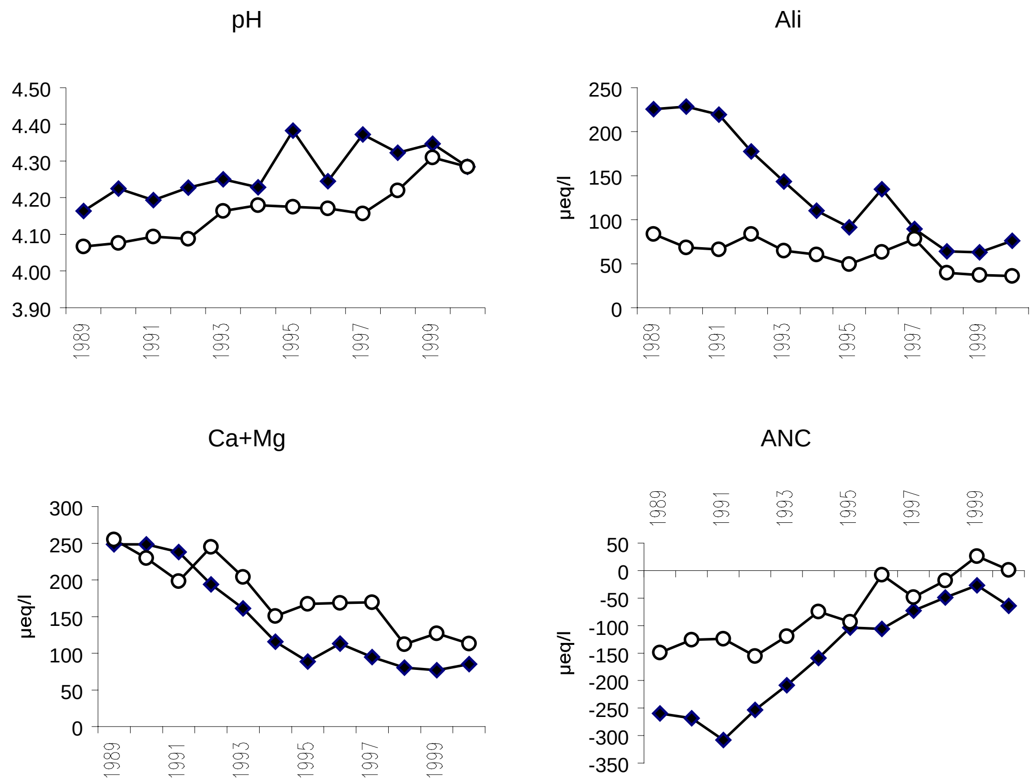


Figure 5-2 Changes in annual volume-weighted runoff concentrations at F1 CONTROL (open symbols) and G1 ROOF (filled symbols). The roof treatment at G1 was started in 1991.

The positive runoff response to the treatment at G1 ROOF and to the decline of ambient deposition at F1 CONTROL is, however, not a consequence of improving base saturation of the soils in the two catchments. The soils of both catchments were extensively surveyed in 1990 and then re-surveyed in 1995, four years after the roof began removing acid inputs to the G1 catchment. One of the most striking features of the re-sampling is that many soil parameters at G1 roof have acidified in one or more soil horizons, despite the total removal of anthropogenic S and N inputs via throughfall and a concomitant improvement in runoff ANC. That result, however, is not necessarily a surprising one, since a number of other studies have argued on theoretical grounds (Reuss and Johnson, 1986) and through modelling predictions that recovery of surface water quality can start before soils become less acid (Matschonat and Vogt, 1998). That initial, rapid recovery of runoff quality is probably, to a large extent, a result of changes in the soil-soil water equilibrium occasioned by declining ionic strength as the flux of the mobile SO_4^{2-} ion decreases. This rapid improvement of runoff quality will only progress as long as the ionic strength continues to decline. The effects of other soil properties such as soil base saturation are not as apparent at this point. It is because the relative change in soil base saturation which could be expected over such a short period of time as four or eight years, apparently has a smaller effect on runoff quality than a decrease of 50% to 75% in ionic strength (Figure 5-3) However at the same time, when the base saturation at the G1 ROOF decreased, the runoff ANC increased from –260 to –160 over the first 4 years of the experiment and from –160 to –50 over the next four years. It is also consistent with the results from F1 control. There, although the soils have not acidified further, they have not improved markedly with regards to soil base saturation, while the F1 runoff improved very significantly (Figure 5-2). At both the roof and control catchments there seems to be a close relationship between ionic strength and ANC in runoff (Figure 5-3).

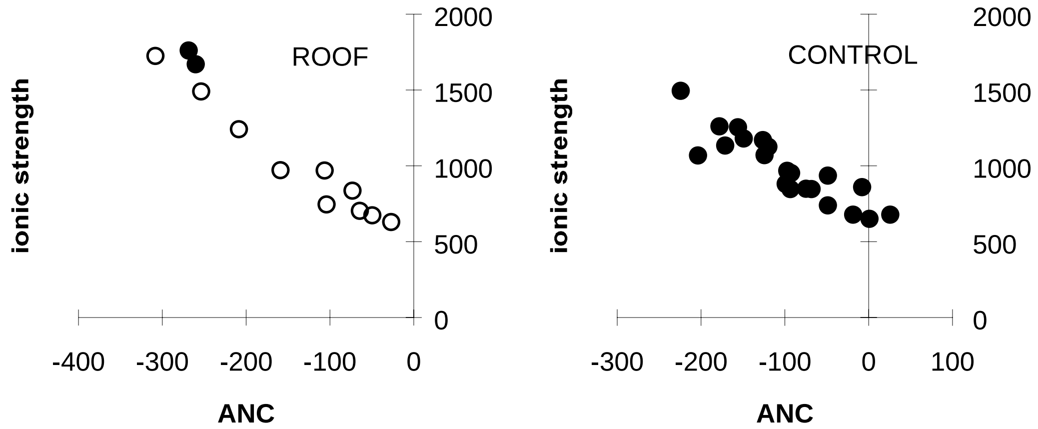


Figure 5-3 Relationship between acid neutralising capacity (ANC) and ionic strength, catchments G1 ROOF and F1 CONTROL, µeq/l. Annual volume weighted averages, at G1 ROOF two years before the roof was constructed (1989 – 1990, filled symbols) and eight years of the clean-precipitation treatment (1991 – 2000, open symbols), as well as at F1 CONTROL during 20 years from 1980 – 2000.

5.2.1.2 SULPHUR ISOTOPES

The studies of sulphur dynamics by using sulphur isotopes started at Gårdsjön in 1990 (catchment G1 and F1). Under the roof over the G1 catchment the natural precipitation was replaced by a sprinkler system, which also enabled addition of chemical and isotope tracers. The sea-water sulphate from the addition of sea-water to a clean precipitation sprinkled under the roof was practically the only sulphate which entered the catchment after the roof construction was completed. It was an excellent tracer since it had a significantly higher $\delta^{34}S$ value than runoff (+19.5‰ in comparison to +5.5‰).

The reduction of sulphate in the sprinkler water was almost immediately followed by a 50% decrease of sulphate concentration in the runoff (during the first five years).

Today the sulphate concentration is in the order of 30-35% of the initial concentration of about 100 μeq (see the section 5.2.1.1). The isotopic sulphur signature of the runoff sulphate showed that adsorbed sulphate was the dominant source in the initial phase, but during the first 6 years no change in the runoff $\delta^{34}S_{SO4}$ value was observed. Since 1996 the $\delta^{34}S_{SO4}$ value has increased by about 1-1.5‰ (unpub. data). This is surprising when considering that the artificial deposition beneath the roof has a very different sulphur isotope composition than the soil sulphur (+19.5‰ compared to about +5.5‰).

Investigation of the soil bound sulphur showed that the spatial distribution of adsorbed sulphate in the soil was large and that most sulphate was found in the B-horizon. In total it can be stated that more than 90% of the adsorbed sulphate is found in the B and C-horizons and less than 10% in the O-horizon (Torssander and Mörth, 1998). For organic forms of sulphur, the distribution is about 70% stored in the B-horizon and 30% in the O-horizon. Mass balance budgets for sulphate showed that the mass of sulphate transported out from the catchment was much larger than the input. In other words, the catchment G1 ROOF acted as a net source of sulphate from the start of the experiment in 1991. Based on isotopical composition of the runoff, this excess sulphate most likely came from the adsorbed sulphate in the B-horizon in the beginning of the experiment. However, when investigating soil samples from 1990 and 1995 a smaller decrease than expected is found of the adsorbed pool of sulphate (unpub. data), which cannot account for the mass flow of sulphate found in runoff. Sulphur isotope data helps to explain these findings. Modelling of the isotope composition in runoff sulphate shows that the $\delta^{34}S_{SO4}$ should have been about 9‰, considering only the adsorbed sulphate as reactive. The measured $\delta^{34}S_{SO4}$ value today is about +7‰. Assuming no fractionation for oxidation of organic sulphur (Krouse and Grinenko, 1991), the B-horizon organic sulphur can be ruled out as a source, since it has a $\delta^{34}S_{OrgS}$ value of about +13‰, in contrast to the O-horizon organic sulphur which is about +6‰. Contributions of sulphur from the O-horizon sulphur will be seen as an increase in the sulphate concentration and no change in the $\delta^{34}S_{SO4}$ value. Even small contributions from the B-horizon sulphur to sulphate in runoff from the B-horizon will be detected since this will increase the $\delta^{34}S_{SO4}$ value and the sulphate concentration.

Several studies have revealed that sulphur is quickly turned over in the upper soil horizons (Van Stempvoort et al, 1990) and that there is evidence for sulphur accumulation in the B-horizon. In view of this, the high $\delta^{34}S_{SO4}$ value in the B-horizon could be interpreted as a reminiscence of sea-water sulphate ($\delta^{34}S_{SO4}$ of about +21‰) mixed with anthropogenic sulphur ($\delta^{34}S_{SO4}$ of about +4‰ today). The much lower $\delta^{34}S_{SO4}$ in the O-horizon therefore reflect deposition, and can be treated as a mean value for the turnover time period.

It can be concluded that adsorbed sulphate was most important in the beginning of the roof experiment but with time data suggest that organic sulphur is mineralised and significantly contributes to the runoff sulphate. The mineralisation is an acidity generating process (Gustavsson 1995). Considering the relative sizes of the adsorbed sulphur and organically bound sulphur stores, where the latter is typically several times bigger than the former, the mineralisation of the organic sulphur is a process with potential to slow down recovery of both soils and surface waters for a substantial period of time. However, the origin and fate of the organic sulphur in the catchment and the rate for immobilisation and mineralisation is not well understood and therefore it is at present difficult to make a correct assessment of the importance of these processes for specific sites and ultimately for whole regions.

Large scale ecosystem manipulations such as the Gårdsjön Roof Experiment generates data specific to the particular experiment. During the initiation of the study in 1990, the experimental data in itself constituted important results and provided basic understanding about the temporal aspects of recovery from acidification as well as visualising the impacts of acid rain. During the later years of the experiment, when a conceptual understanding of acidification and recovery had been established among scientists as well as policy-makers, it became more important to answer questions concerning the real world outside the experimental site, *i.e.* to use the results from the roof experiment to predict the historical and future impact of acid rain in a larger geographical region. Such generalisations are best done by mathematical models, which are basically mathematical formulations of relevant processes.

Mathematical models cannot be validated in the sense of being proved to be correct descriptions of nature. However, models that are repeatedly passing tests of their predictive capabilities can be given some credibility. There are at least three major sources of uncertainties involved in the modelling of acidification and recovery. The first is uncertainty of input parameters, i.e. parameters describing the nature of the plot, catchment, lake or any other modelled system. The second uncertainty is that of driving variables (deposition sequences, climate, etc), which are also uncertain to some extent, even when measurements are available. In both these cases, the uncertainty can be dealt with. For instance, if the uncertainty of any given parameter or variable can be estimated, then the effect of that uncertainty can be tested by the model. However, there is a third type of uncertainty, which stems from the fact that models reduce the vast complexity of nature to a few key processes, which are then described in more or less simplified ways. Thus there always remains a possibility that when applying the models outside the spatial and temporal frame of the data-set used for testing and evaluation, there might be processes whose importance was not previously recognised, and which are thus not included in the model.

Large scale manipulations generate data suitable for testing and developing models. The experimental sites are usually very intensely mapped and monitored providing insights into the within-ecosystem processes. Manipulations cause effects, which can then be used for testing the models. All the gathered background information serves as a basis for comparison of the model outcomes from a number of perspectives. To be able to model correctly the observed changes in the experiment, without being in conflict with any additional information available at the site, poses the best available test of a model. After repeatedly passing all tests satisfactorily, the models can be used to generalise the experimental result. This generalisation is possible because of the relative ease with which they can be transferred from an experimental site to other situations, both in time and in space, simply by changing model parameters. Naturally, the issue of uncertainty must not be forgotten. Model predictions should never be interpreted without bearing in mind that they involve a degree of uncertainty, which can only be estimated to a certain extent. On the other hand, as stated above, there is no absolutely certain way to find out about the future in advance; it can only be estimated. A combination of physical and mathematical models such as the Covered Catchment Experiment at Gårdsjön and the MAGIC model, are valuable tools for making such estimates.

Before leaving the roof experiment at Gårdsjön, one other aspect of this large scale manipulation should be mentioned. The acidification related research driven by IVL has been going on at Gårdsjön for 20 years even before the roof was built. The roof experiment backed up by the data, knowledge and tradition gained before it, became a symbol for both acidification problem and acidification research, which is known and recognised well beyond the national borders. It has served as a visualising tool to a several thousands of visitors who came to see the experiment and has helped to get the message across how the long-range transported acidifying deposition affects the soils, forests, streams and lakes in Sweden and what are the options for the future. The experiment was presented at countless occasions to an audience ranging anywhere from both national and international level politicians, royalties, students and their teachers, scientists, journalists, TV crews, environmental activist, people from industry and to general public. The scientific results and achievements from the roof experiment are certainly of interest to a hundred or so people directly involved in the experiment and to experts who learned about it through publications. The experiment might even have affected the decision-makers at various levels, not the least during the negotiations preceding the signing of the Gothenburg protocol. However, it could be argued, that the media attention and the contact with visitors from the outside of the research community were equally important. It might have significantly contributed to form a public awareness and public understanding of the acidification problem, which is one of the key factors for achieving changes in democratic society.

5.2.1.3 MAGIC MODEL CALIBRATION AND TESTING

The calibration procedure for MAGIC involves the estimation of historical deposition, uptake and immobilisation sequences (140 yr.). Rates of historical deposition used for the period prior to start of the measurements are estimates given by Mylona (1996) for sulphur and Simpson et al. (1997) for nitrogen based on historical emissions of SO_2 and NO_x in Europe. For the experimental years the measured atmospheric input at each site was used.

Calibrations were done to the base line represented at Gårdsjön by two pre-treatment years, and the model was calibrated to the average values of these years. The calibration procedure is further described in Chapter 4.3.4.2 and in Cosby et al. (1995).

After calibration of MAGIC the model performance was tested by applying the model to the roof covered catchment where the inputs of acidity, SO_4, NO_3 and NH_4 were strongly reduced. For the evaluation procedure, the actual inputs by irrigation under the roof was used as input. The input under the roofs included additional base cations, especially K and to a lesser extent of Ca, to compensate for the canopy leaching of those elements which was removed by removing the throughfall (Gundersen et al., 1995). At all sites the roof also excluded deposition of non-marine base cations (for discussion see Moldan et al., 1998). The modelling and roof validation procedure adjusted for those potential disruptions of the element cycling.

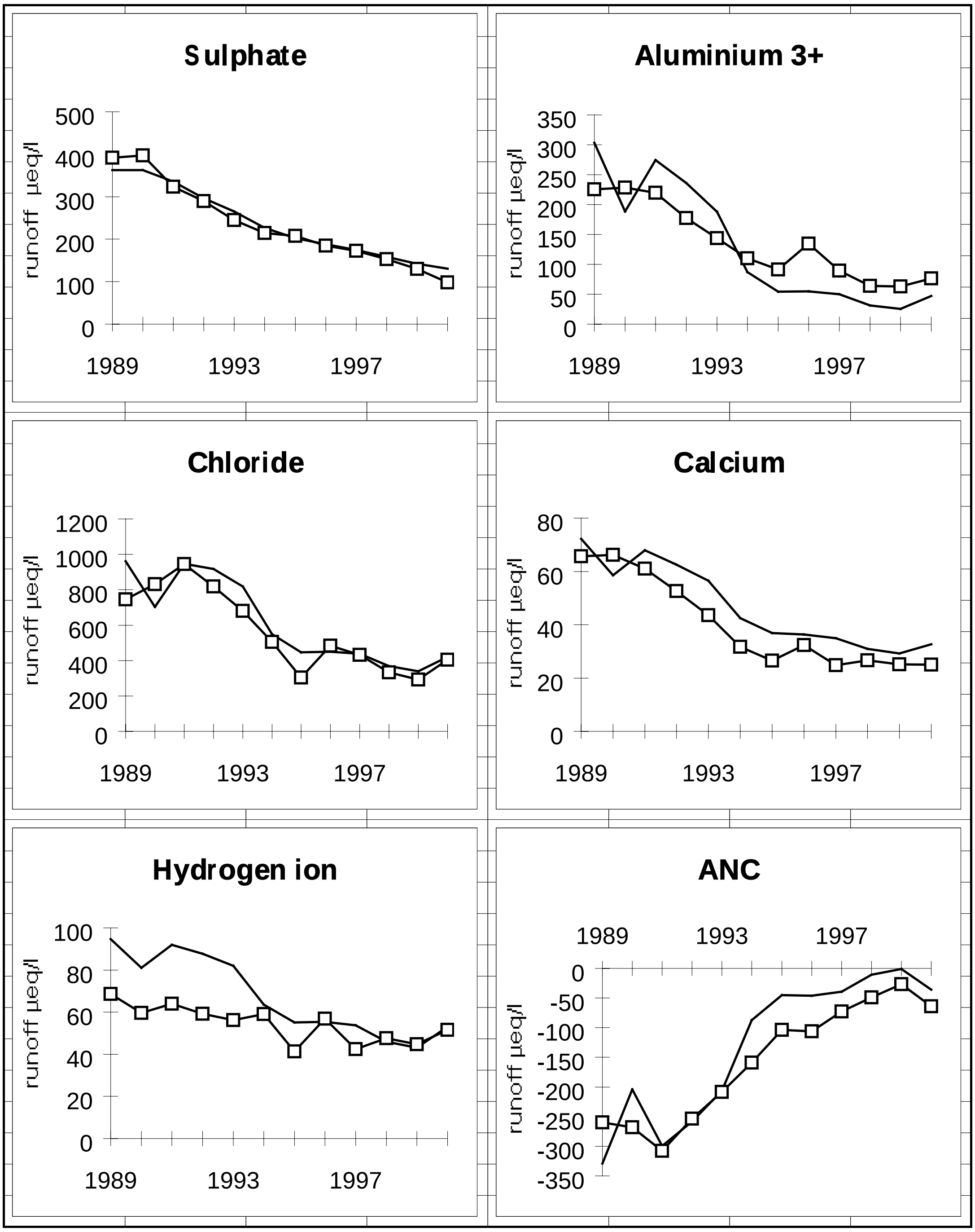


Figure 5-4 Measured annual volume weighted runoff concentrations at the G1 ROOF catchment (square symbols) and MAGIC simulation (lines).

The model output and the measurements for the “roof experiment” show relatively good agreement (Figure 5-4). MAGIC thus well predicts the observed changes in acidification of soil and water induced by the large and sudden experimental decrease in acid deposition under the roofs. The results from the application to the roof experiment is a test of the model with measurements at realistic time and spatial scales.

The calibrated model was used to test the effects of the future changes in atmospheric deposition on ecosystem acidification and recovery by applying 2 different scenarios for 2000-2030. The historic deposition sequence until the start of the measurements at each site was the same for all three scenarios, and the scenarios at each site only differed for the treatment and forecast years as follows:

Gothenburg scenario (ambient): This scenario consists of the basic historic deposition up till the start of the experiments and the measured deposition for the experimental period until 2000. For the future deposition the prescribed change in sulphur and nitrogen according to the Gothenburg protocol was used (UN/ECE 1999). This entails a reduction by 60% in non-marine SO_4 input by 2010 relative to 1999 and a reduction of 50% in NO_3 and NH_4 inputs by 2010 compared to 1996. The deposition change up until 2010 was done by calculating the target deposition by 2010 and interpolating from 2000-2010. The future input of all other elements were kept at present day levels for the whole period 2000-2030.

Roof scenario: The roof scenario was similar to the Gothenburg scenario except from the deposition during the period of the roof experiments. The actual reduced inputs of H^+, SO_4, NO_3 and NH_4 during the duration of the roof experiment was used as input. Compared to the Gothenburg scenario this means an advancement in both timing and magnitude of the reduction in acidic input. This scenario was adjusted compared to the experimental roof, so that the roof effect was idealised to be only the exclusion of non-marine S, N and H^+.

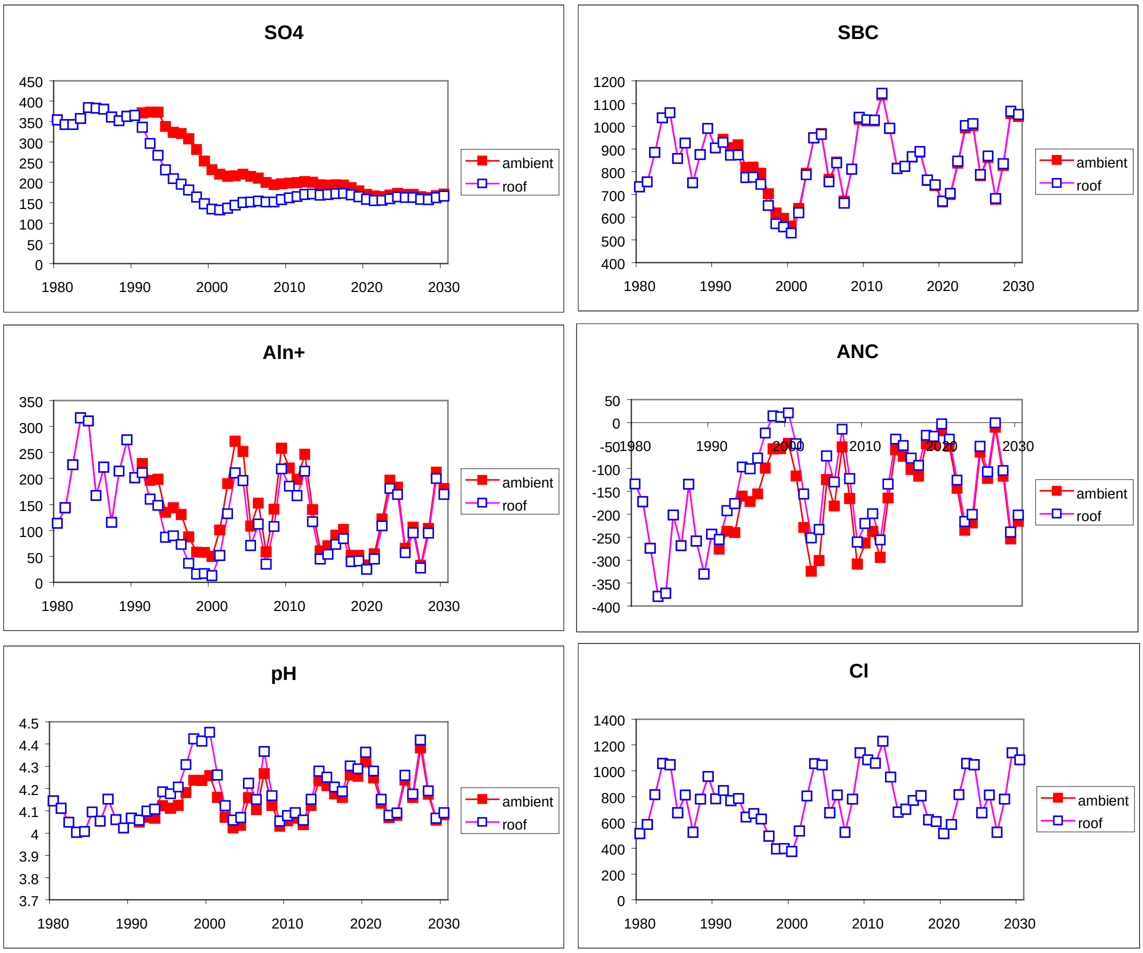


Figure 5-5 The effect of the acid deposition reduction caused by the roof and by the present and projected decrease in emissions of acidifying deposition at the Gårdsjön G1 ROOF catchment.

The effects of different strategies for reduction of the acidic input (reductions of H^+, SO_4, NO_3 and NH_4) are illustrated by comparing the model outputs for the "Gothenburg-scenario (ambient)" and the "Roof scenario" (Figure 5-5). The reduced inputs of acidifying N and S under the roof have a strong and immediate effect on the water chemistry resulting in reduced concentrations of sulphate and aluminium and increased pH and ANC. The effect of the "roofs" on the water chemistry lasts for a number of years after the roof was removed (both in model and in the field). At Gårdsjön, the difference in ANC was up to 78 µeq/l and decreased to 52 µeq/l five years after the roof was removed and even after 30 years, there is still a visible difference of 14 µeq/l. This relatively long lag time is due to the large pool of adsorbed sulphate in the soil and therefore the large absolute amount of the SO_4^{2-} leached from the soil during the roof experiment.

With respect to soil chemistry, however, the effect of the roof last even much longer. This is best shown by the development of soil base saturation under the roof scenario relative to the Gothenburg (ambient) scenario (Figure 5-6).

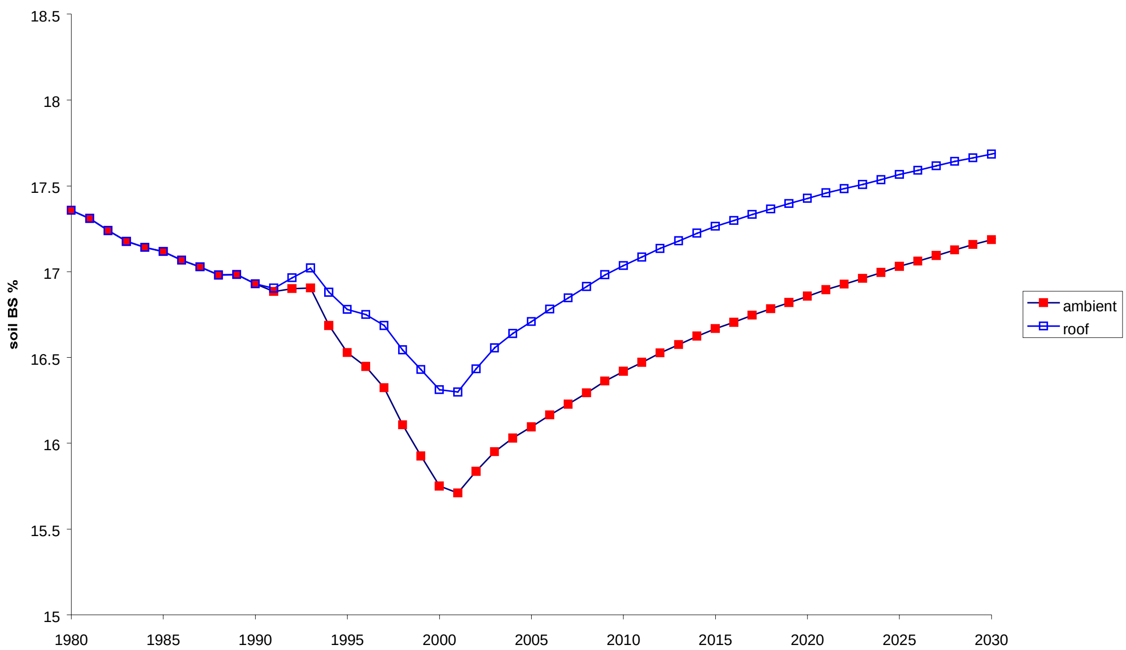


Figure 5-6 The effect of the reduced deposition by the roof and outside the roof on the soil base saturation. The roof scenario was idealised to involve only the 100% reduction of the acidifying deposition.

The base saturation increased by 1.5% and the difference persisted for several decades into the future after the roof was removed. That could be interpreted as an effect of earlier and more complete reduction in acid deposition, than what was achieved at the same time outside the roof by the emissions abatement.

5.2.1.4 Recovery of soils

As discussed in chapter 3, there are currently 1883 forest soils for which critical loads have been calculated in Sweden (Figure 3-3). These are forests in the Swedish national forest inventory Ståndortskarteringen. Most of the data needed for the critical loads calculations are available in the Ståndortskarteringen database except deposition which has been estimated by SMHI and IVL using the MATCH model and mineralogy which has been estimated through total elemental analysis on all 1883 soils, mineralogical analysis on a subset of the 1883 soils and the UPPSALA back calculation model.

Information on mineralogy is currently the limiting factor for calculating critical loads of acidity for forest soils in Sweden. This might change in the near future, however, as an extensive data set with more than 10 000 total mineralogies in southern Sweden is being developed in co-operation with the SUFOR programme.

An extensive update of the critical loads calculation model PROFILE was carried out with aid from the ASTA programme in 2001 and the resulting new critical loads maps (chapter 3) have been reported to the CCE. The new features include both improved routines for processes such as nitrogen immobilisation, root uptake efficiency, interdependence between nitrogen and base cation uptake as well as improvements in the critical loads iteration procedure. As a result of the improvements it turned out that the negative critical loads calculated for some of the sites in an earlier uncertainty assessment (Barkman 1998) was due to unrealistic combinations of base cation and nitrogen uptake rates. Changing the critical loads iteration procedure and introducing an interdependency between base cation and nitrogen uptake totally eliminated these negative critical load estimates.

Several of the improvements made in the critical loads calculation model PROFILE are unfortunately difficult to implement in a dynamic model: Currently the SAFE model calculates the soil chemistry and calibrates the initial base saturation for one layer at a time, using the result from one soil layer as input to the next soil layer. To implement the improvements discussed above it is necessary to calculate the chemistry in all soil layers simultaneously, something which makes the calibration of initial base saturations a lot more difficult. A new version of the SAFE model which calculates all soil layers simultaneously has been developed within the ASTA programme, but that version is currently not robust enough for regional assessments. It has been used, however, together with the sulphate adsorption model developed within ASTA to estimate the effect of sulphate adsorption on the recovery from acidification at lake Gårdsjön (Figure 5-7).

While information on mineralogy currently is the limiting factor for calculating critical loads for acidity for forest soils in Sweden, the limiting factor for dynamic calculations of soil chemistry is measurements of exchangeable cations. According to recent information from Ståndortskateringen, exchangeable cations have been measured to some extent in all 1883 soils for which data on mineralogy exist. This data is, however, currently being processed and all the data is therefore not yet available.

The model set-up used in the dynamic assessment of forest soil chemistry consists of the MAKEDEP model which creates internally consistent time series of atmospheric deposition and nutrient uptake, the initSAFE model which calculates the initial, pre-acidification, soil chemistry, and the dynamic soil chemistry model SAFE. The dynamic assessment was carried out using the new version of the MAKEDEP model developed in a co-operation between the ASTA and SUFOR programmes, EKG-GeoScience, Switzerland and ÖKODATA, Germany. The new MAKEDEP model includes, among other things, nutrient content elasticity, i.e. that the nutrient content of a tree varies with nutrient availability.

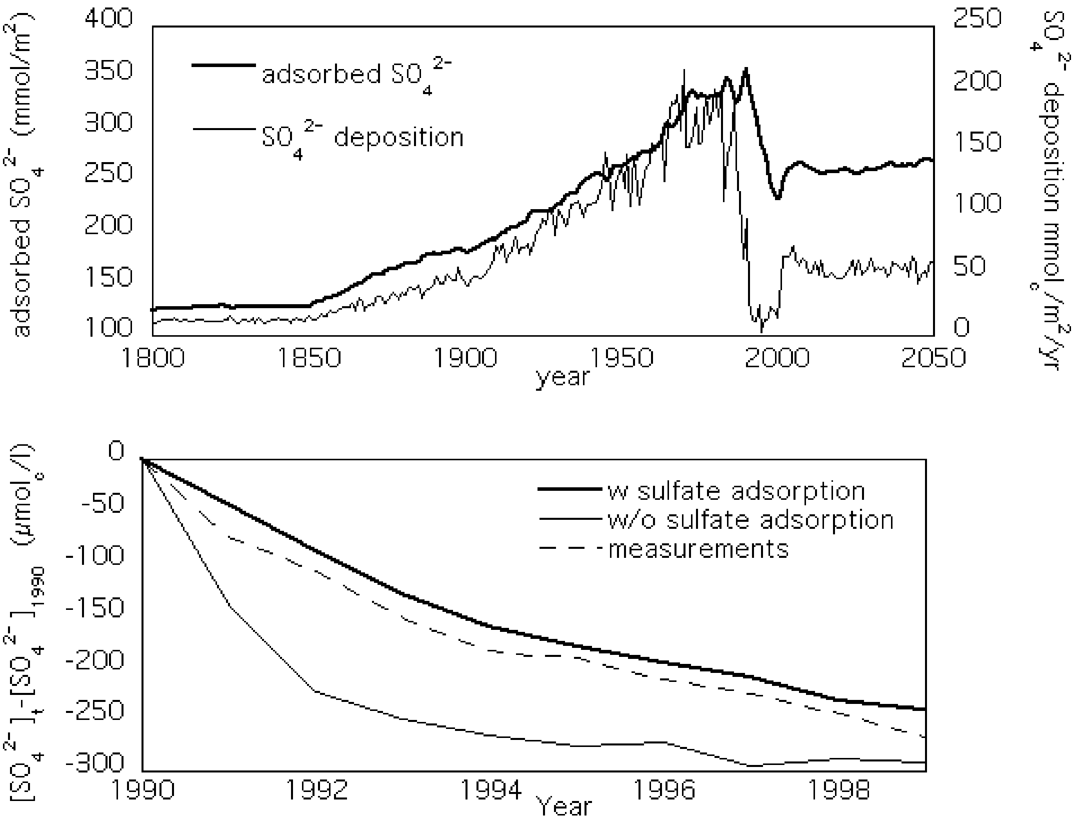


Figure 5-7 The upper graph above show simulated adsorbed sulphate at the roofed G1 catchment at lake Gårdsjön using the SAFE model with sulphate adsorption included. To highlight the change in response the relative change in sulphate concentration with and without sulphate adsorption is displayed in the lower graph.

There are currently 710 sites for which all data needed is available. Out of these 710 sites, MAKEDEP currently succeeds in reconstructing the estimated vegetation biomass at 644 sites. The number of sites for which the MAKEDEP calculations are successful is to some extent dependent on the estimated land use history.

The land use history influences the results, especially the hindcasts of soil chemistry. In the current regional assessment, little information was available. It was assumed that the rotation period is longer in the northern part of Sweden than in the southern. In order for the rotation period to be consistent with the information on stand age where the mean age of some stands are up to 175 years old, it was assumed that the rotation period may be as long as 200 years. Forest thinnings where 10% of the vegetation is cut down were assumed to be carried out when the forest is10 20 and 30 years old. The MAKEDEP model was calibration to 1997 deposition and to the biomass present at the time of the Forest Inventory at each site. Care must be taken to avoid a potential inconsistency between MAKEDEP and the deposition data, as the deposition data available uses type of forest only and do not consider that the filtering efficiency varies with stand age, something which MAKEDEP considers. The land use history was therefore tailored to make sure that the simulated forests were at least 15 years old in 1997.

The future deposition scenario used in the dynamic assessment was provided by Gun Lövblad, IVL and is based on RAINS model calculations by IIASA using the agreements in the Gothenburg protocol. The current scenario divides Sweden into 6 different regions. In upcoming regional assessments new data from IIASA with EMEP-grid specific deposition scenarios is likely to be used.

There is a clear spatial trend in simulated soil chemistry (e.g. Figure 5-8). The soils with very low pH (3.8<pH<4) in 1990 are mainly situated in the southernmost part of Sweden, whereas the soils with pH above 4.6 are mainly found in the northern half of Sweden. Some exceptions exist, and this might be due to local variations, e.g. in mineralogy. The simulated pH starts to decrease already prior to 1900 (Figure 5-9), although the decrease is fastest around 1950-1970. Soil solution pH is only predicted to recover slightly (Figure 5-8 & Figure 5-9). The predicted increase in pH from 1990 to 2010 is similar for all percentiles (Figure 5-9).

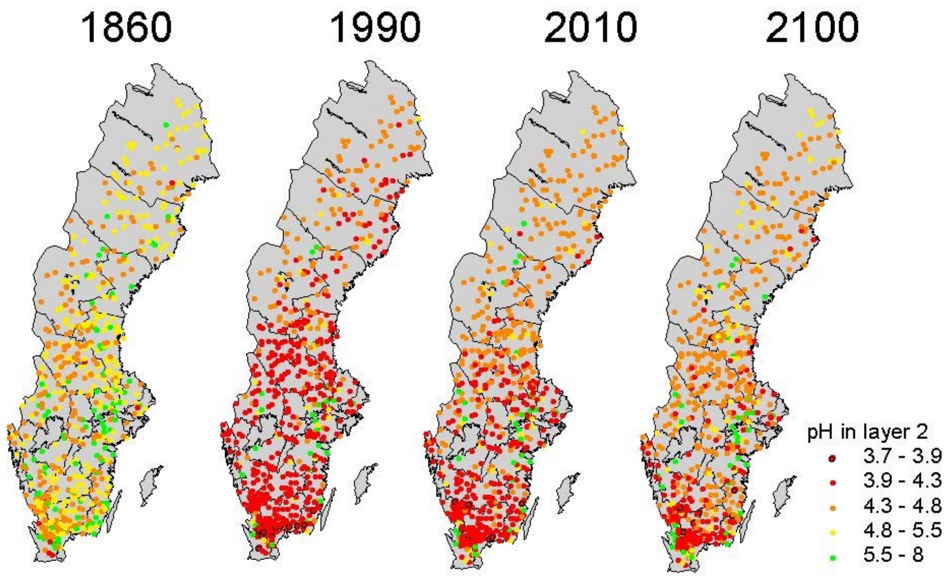


Figure 5-8 Map of Sweden showing sites in the regional dynamic assessment of soil chemistry. The colour of the dots represent the simulated pH in layer 2 at different years using the Gothenburg protocol as the future deposition scenario, assuming constant deposition between 2010 and 2100.

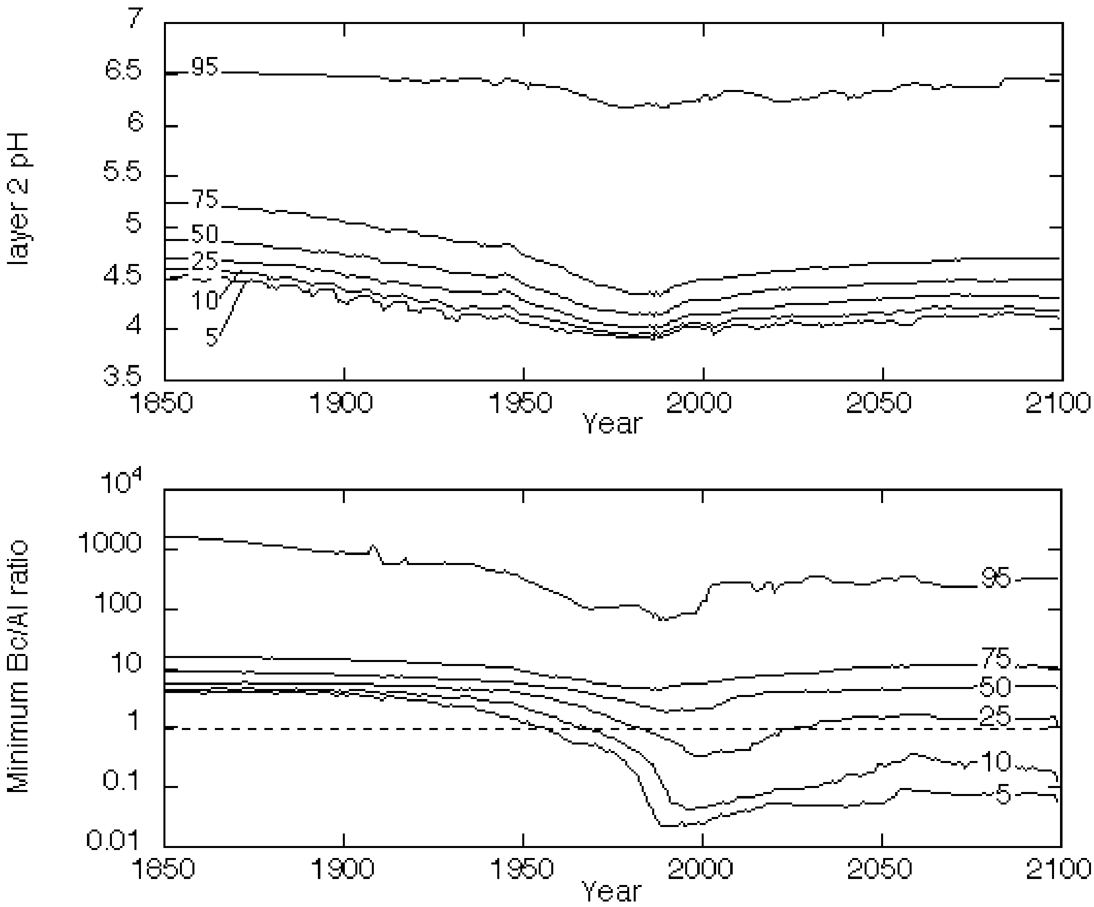


Figure 5-9 Percentiles of simulated pH in layer 2 and minimum BC/Al-ratios in the 644 forest soils.

In calculating the critical load of acidity using a multilayer soil model, the minimum BC/Al ratio in the soil profile is used as the chemical criteria. By comparing the simulated minimum Bc/Al ratio in 1860 with that of 1990 it is obvious that there has been a tremendous decrease in Bc/Al ratio for very many sites (Figure 5-10). As with pH, the increase in minimum Bc/Al ratio from 1990 to 2010 is similar for all percentiles. It is important to realise, however, that a similar increase for all percentiles does not mean that the Bc/Al ratio increases for all sites. From 1860 to 1990 the Bc/Al ratio decreased considerably for most of the sites (Figure 5-10) but between 1990 and 2010 some sites recover slightly while others continue to acidify.

Due to the time-lags involved it will take a few decades before the full potential of the measures taken in the Gothenburg protocol is translated into recovery from acidification. It is obvious, however, from the model simulations that the Gothenburg protocol is not enough if we want 95% of our forest ecosystems to fully recover from acidification. In total approximately 35 % of the sites are projected to have a remaining violation of the Bc/Al criteria in 2010 (Figure 5-11). If no emission reductions except those already agreed upon in the Gothenburg protocol are realised, approximately 20 % of the sites have remaining violation of the criteria in 2100 .

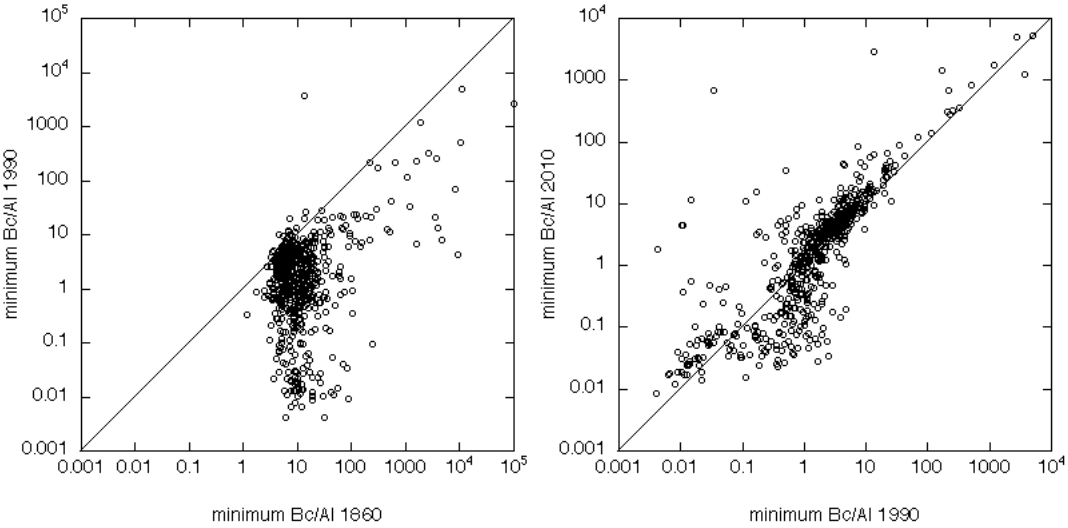


Figure 5-10 Simulated minimum BC/Al in the soils for 1990 compared with simulated pre-industrial BC/Al (1860). Dots below the 1:1 line have acidified since 1860.

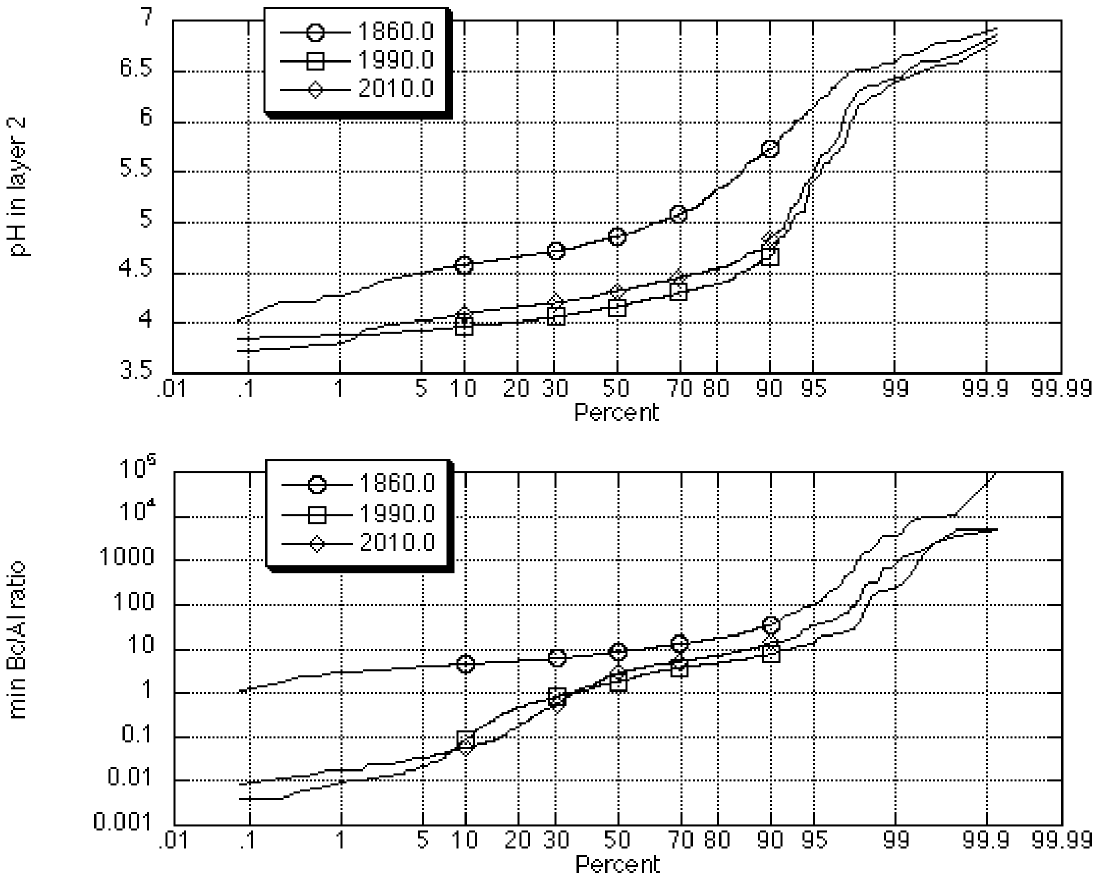


Figure 5-11 Cumulative diagrams of layer 2 pH and minimum BC/Al ratio in the 644 forest soils at 1860, 1990 and 2010.

A tool for uncertainty assessments in dynamic simulations using the model set-up described above has been developed in co-operation with EKG Geo-Science, Bern, Switzerland. Performing an uncertainty assessment is, however, labour intensive since the uncertainty in all input parameters have to be estimated and since the number of simulations needed increase tremendously. Within the first phase of the ASTA programme only qualitative estimates of uncertainty are therefore made.

The regional assessment of recovery from acidification underestimates the recovery time since sulphate adsorption is not yet included in the assessment. To assess the extent of this underestimation and to facilitate future inclusion of sulphate adsorption in regional assessment a simple sulphate adsorption submodel was added to the SAFE model. The sulphate adsorption model was applied to the previously roofed catchment G1 at lake Gårdsjön. The three most important aspects of these simulations are that 1) the amount adsorbed sulphate largely follows the deposition, 2) model output using the sulphate adsorption submodel shows a smoother simulated soil solution chemistry that is delayed as compared to model output without sulphate adsorption, and finally 3) that sulphate adsorption is most important in the short time perspective, approximately 10 years, and has no influence on the long term endpoint.

Another important source of uncertainty is land use. The data used in this regional assessment is questionable and the aim is to improve these data during 2002. The land use history affects not only the hindcasts of soil chemistry, but also indirectly the future projections since all dynamic models have to be started at some point in time and since the estimated initial conditions to some extent affect the entire simulation. The effect of future land use highlights the emerging potential conflict between forestry and the environmental goal within LRTAP: The amount of nutrients removed through harvest influences the critical load of acidity. Thus, the recovery rate of a forest soil is dependent not only on the load of acidity but also on the forest management.

5.2.1.5 RECOVERY OF THE LAKES MODELLED WITH THE MAGIC

To make a regional assessment of recovery, a model should be run on many representative sites (e.g. lakes, streams, soil sampling plots) in a region and the results of the modelling then interpolated for the whole region. This method is often referred to as the multiple site approach. The multiple site approach requires data from each modelled site, which can be a limitation. The advantage of this approach is that the model outcome allows both a statistical overview of how the region responds to acid deposition, and, since real sites with known locations have been modelled, the model results can be summarised in the form of maps. As an alternative approach, the Monte Carlo method can be applied. In this approach, data from several known sites within a region is used to generate a population of hypothetical sites which are considered to be representative for this region. The model can then be run on those hypothetical sites and the statistics of the results presented. This method is often used for the regions where enough site-specific observations are not available. In Sweden, there are several monitoring programs, which provided basic data for a multiple-site MAGIC application to a relatively large number of well investigated lakes. This modelling effort has been on-going intensively since fall 2001, and at the time of writing is neither finished nor finalised. However, some indication of the results to date is presented below.

As a part of the regional assessment of recovery from acidification within the ASTA programme, 143 lakes across Sweden have been modelled using the dynamic model MAGIC (see model description in the section 4.3.4.2). MAGIC has been previously used in regional modelling of lake acidification and recovery in Norway (Cosby and Wright, 1998) and in Great Britain (Evans et al., 2001) and is currently applied to a several other regions in Europe within the major EU project on recovery from acidification, RECOVER:2010. In Norway, data from three national lake surveys in the southern part of the country were used both for Monte-Carlo and site specific modelling of the recovery from acidification. The results from both methods agreed well with the observations from the surveys, showing that >65% of the lakes were acidic between 1974 and 1995 and that during the 1990's the average ANC of the lakes was increasing. After 10 years of sulphate deposition at 20% of the 1974 deposition, >65% of the lakes had positive ANC. In Great Britain, lakes in six acid sensitive regions have been modelled using the site specific approach and three different future scenarios. The results differ between the regions and the future scenarios, so that one region would continue to acidify under one of the scenarios, while another region started recovering under the same scenario.

The Swedish lakes chosen for modelling within the ASTA programme are part of a national monitoring programme carried out at the Swedish Agricultural University. The lakes were sampled with regard to water chemistry several times per year since 1984. The lakes are situated in areas with no significant agricultural activities, are generally not limed or polluted by e.g. waste water and are well suited for studies of acidification through atmospheric deposition. It should be noted that the most acid sensitive lakes are underrepresented among the 143, since many very sensitive lakes in Sweden have been limed before the commencement of this lake-monitoring programme.

The other key data needed for the modelling are the atmospheric deposition on the lakes and their catchments and characteristics of the soil in the lake catchments. Deposition data used has been estimated by SMHI and IVL using the MATCH model. The future deposition scenario used was provided by Gun Lövblad, IVL and is based on RAINS model calculations by IIASA using the predicted emissions according to agreements in the Gothenburg protocol. Data from Ståndortskarteringen was used to estimate soil properties (provided by Erik Karltun, SLU), using averages of parameters from sites from within and near the lake catchments. Base cation uptake by vegetation was estimated from data in the Swedish National Atlas. As a first approximation, uptake has been assumed constant over time, i.e. the forest in the catchments were modelled as extensively managed so that the average age of the stand is constant. The deposition of sulphate, nitrate, ammonium and H^+ were the input-data that have varied with time (Figure 5-13) (Mylona, 1996) and the observed changes in lake chemistry are driven by these.

Large lakes with retention time longer than 5 years were excluded from the data set and these were not modelled. This was because higher retention time might mean that in-lake processes (as opposed to the processes in the terrestrial parts of the catchments) could have very important role in mediating the lake chemistry response to changing deposition. The model was calibrated for the year 1997. Lake and soil chemistry of each individual lake was simulated from 1857 to 2050. The way in which all necessary parameters were derived from the deposition, soil and lake data was the same for all lakes. In the first round of model calibration, in 122 cases the model predicted correctly both lake water and soil chemistry for the calibration year. Twenty-one of the 143 lakes had inconsistencies in the data that made them impossible to calibrate without site specific adjustments of the data that could not be justified by our present knowledge of the lakes.

The modelling of the lakes and evaluation of the results is currently in progress and hence the results presented here are preliminary to an extent.

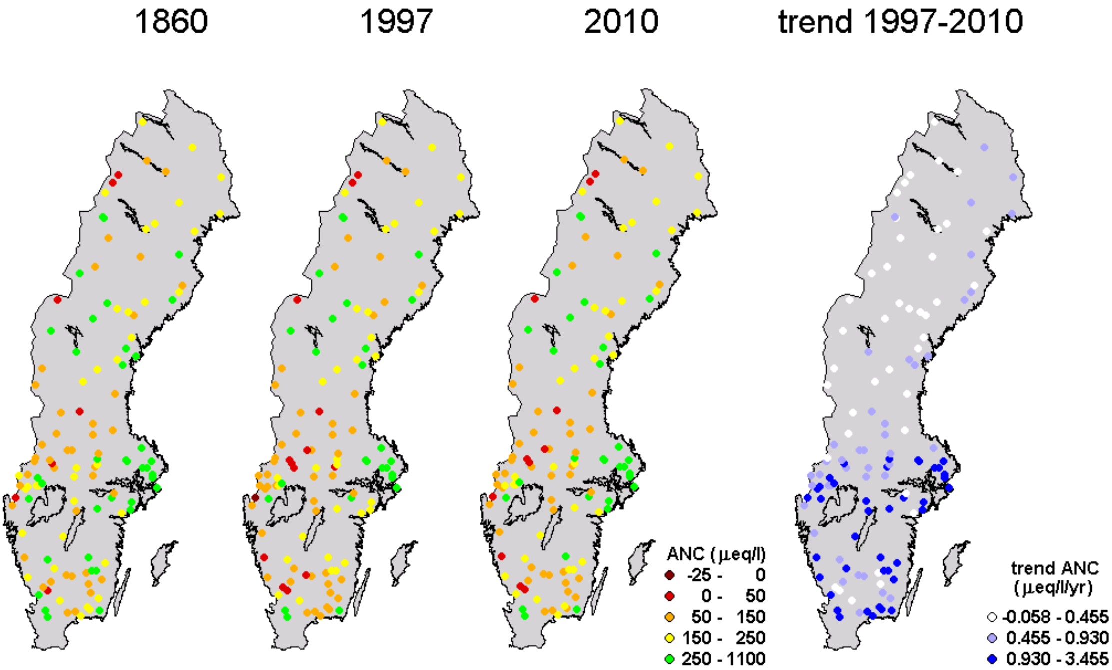


Figure 5-12 Map of Sweden showing the 122 calibrated lakes. The colour of the dots represents the modelled ANC in the lakes at 1860, 1997 and 2010 and the trend of ANC between 1997 and 2010.

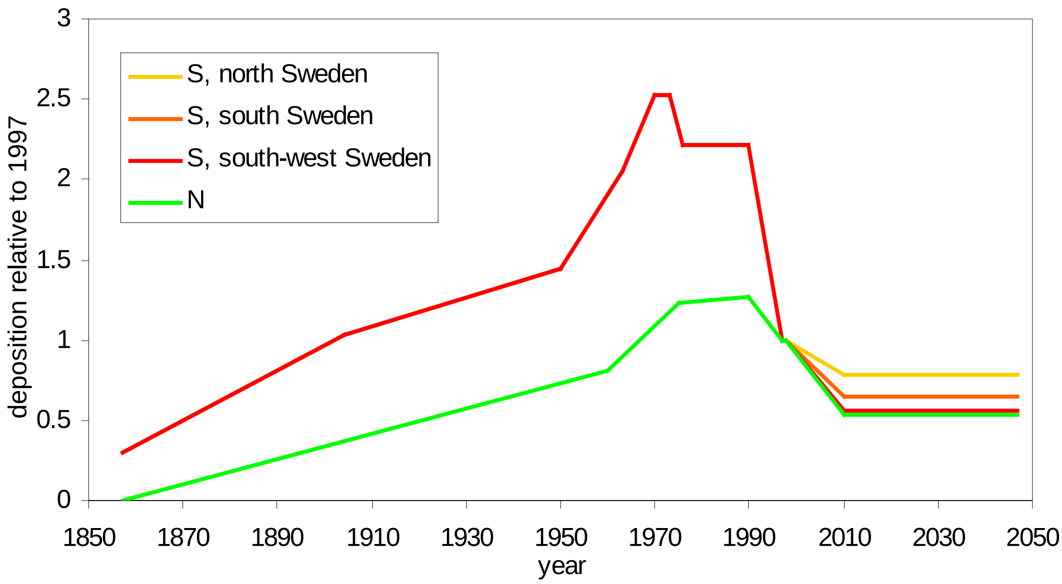


Figure 5-13 Deposition sequences of sulphur and nitrogen.

All of the 122 lakes have been affected by the acid deposition, but many have never become heavily acidified (Figure 5-12). That is not surprising considering the selection of the lakes. The recovery of the acidified lakes, whether fully or partly, is much faster than the recovery of the catchment soils. The soils in most cases did not recover and at a number of the sites continued losing base cations despite the decrease in acid deposition from 1997 to 2010 (Figure 5-17).

The model calibration to the measured characteristics of the soils and lake water in 1997 succeeded relatively well (Figure 5-14). Given that no site-specific adjustments were done at any of the 122 successfully calibrated lakes, this points towards a considerable internal consistency of the data set used for the modelling and it also increases confidence in the ways in which the soils and deposition data were aggregated.

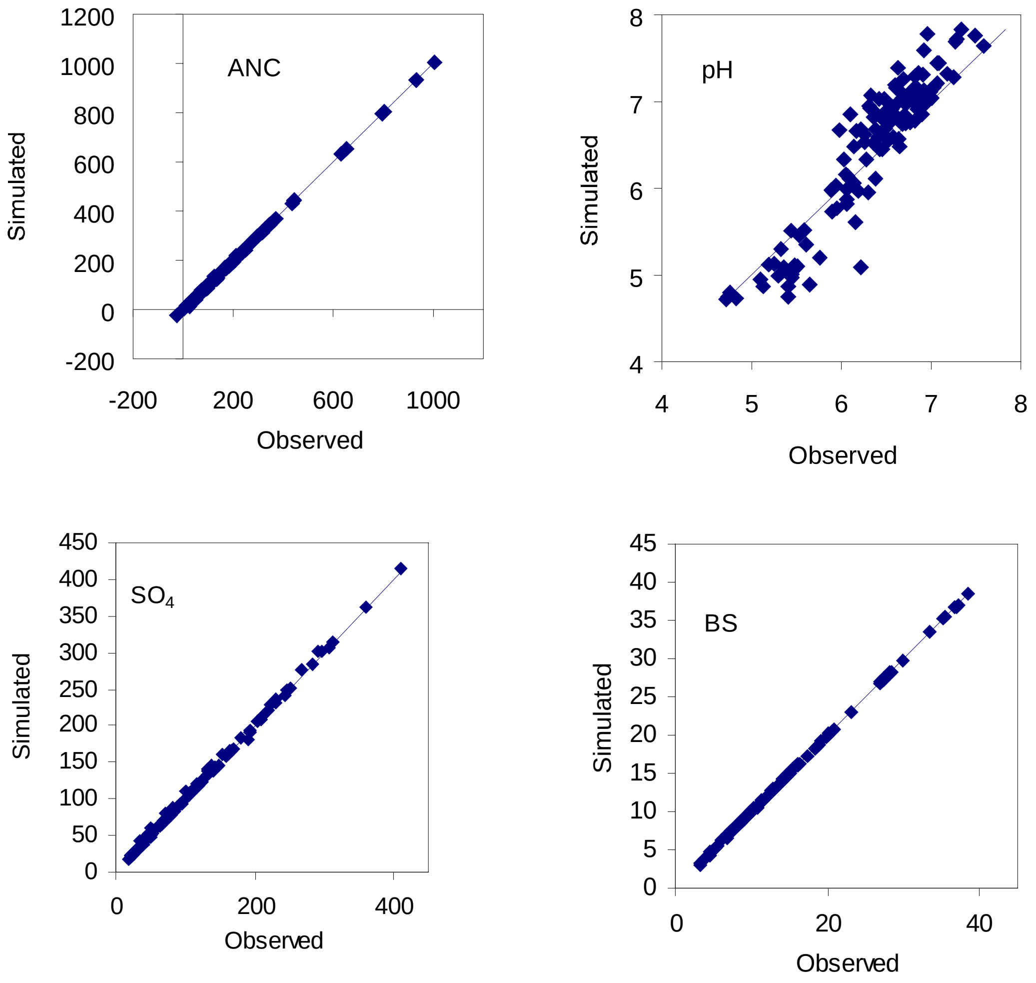


Figure 5-14 Simulated vs. observed values for 1997 of ANC, pH and SO_4 in the lakes and soil base saturation (SB) in the lake catchments.

The calibration of the model was done to a single year 1997. Apart from comparing the modelled and observed data in 1997, another test of the calibration is to compare the modelled and measured lake water chemistry trends over time. The lake water chemistry trends in Scandinavia have been recently published by Skjelkvåle et al., (2001) who reported measured trends in lake water chemistry in Sweden, Norway and Finland. The Swedish lakes included in the analysis by Skjelkvåle et al., were in 58 cases the same lakes as the ones included in the MAGIC modelling. Note that the comparison of the observed and modelled trends in lake water chemistry on the 58 lakes presented below is without the model being calibrated to the trends.

The modelled and observed trends for 1990 to 1999 in lake water ANC (Figure 5-15) and SO_4 were in average nearly the same. Therefore both testing the model against a single year and against a trends over a decade confirmed the model's performance. A number of valid management questions about the average behaviour of the modelled lakes can be answered by projecting the modelled lakes into the future. Since the lakes are distributed over the whole country, conclusions about the regional assessment can be drawn.

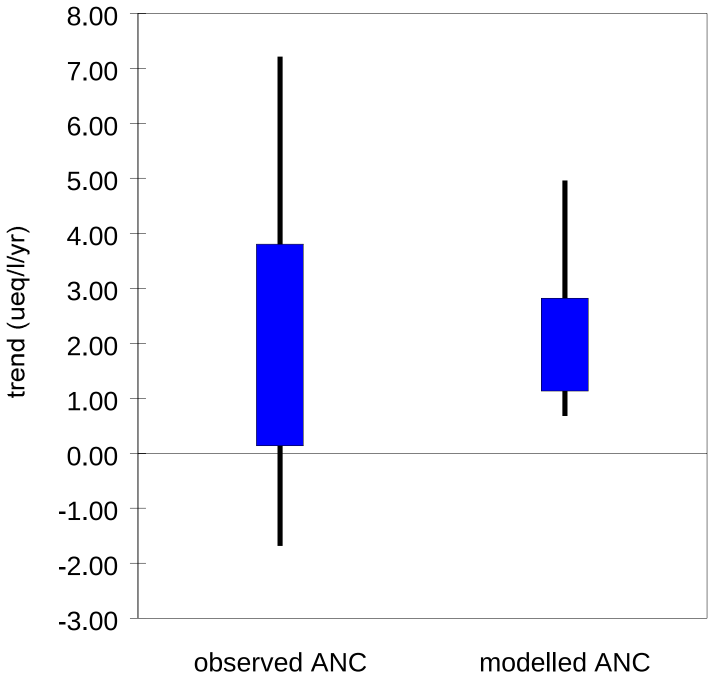


Figure 5-15 5, 25, 75 and 95 percentiles of trends of ANC for the lakes common to the modelling and the Skjelkvåle et al (2001) investigation.

As an example the Gothenburg protocol deposition scenario was modelled (Figure 5-13, Figure 5-16, Figure 5-17, Figure 5-18). The historical lake ANC (as simulated by MAGIC) has decreased regionally from 1860 to 1990. In 1990 there were decreases in sulphate deposition as a result of the early European emissions reductions protocols. The Gothenburg protocol requires additional emissions reductions until the year 2010. The model simulations of the effects of these deposition reductions was a countrywide increase in ANC from 1990 to 2010. These reductions, however, did not bring the ANC to pre-industrial conditions on any of the modelled lakes (Figure 5-16. The main explanation is to be found in the lack of recovery of the catchment soils (Figure 5-17)

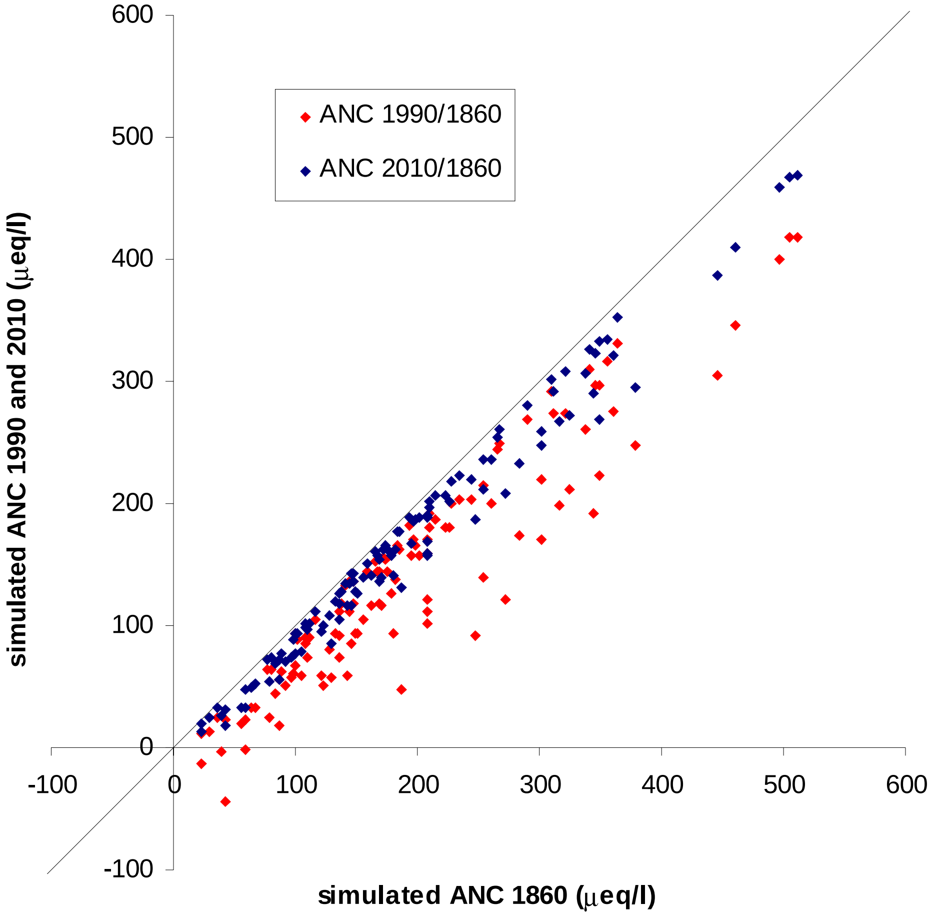


Figure 5-16 Simulated ANC in the lakes for 1997 and 2010 compared with simulated pre-industrial ANC (1860). Dots below the 1:1 line have acidified since 1860.

Although the average regional response of the lake chemistry was modelled well, the observed trends exhibited larger extremes than the modelled ones (Figure 5-18). The procedure in which the regional data are used for the modelling involves smoothing of extremes in several ways. It is essentially a question of the scale in which the data are available. Real lake systems are subject to a great deal of spatial heterogeneity. Small-scale differences in geology, land-use, deposition exposure, etc. can result in a large effects on the chemistry of a particular lake. However, the spatial distribution of the data used for the modelling may not always capture that small-scale variability. The maps and surveys from which the modelling data are drawn capture the means and large-scale trends in variables, but small-scale differences are frequently not observable in the data sets used to drive the model simulations. Therefore it is to be expected that the observed lake chemistry values and trends will exhibit larger extremes than model predictions, as is the case here.

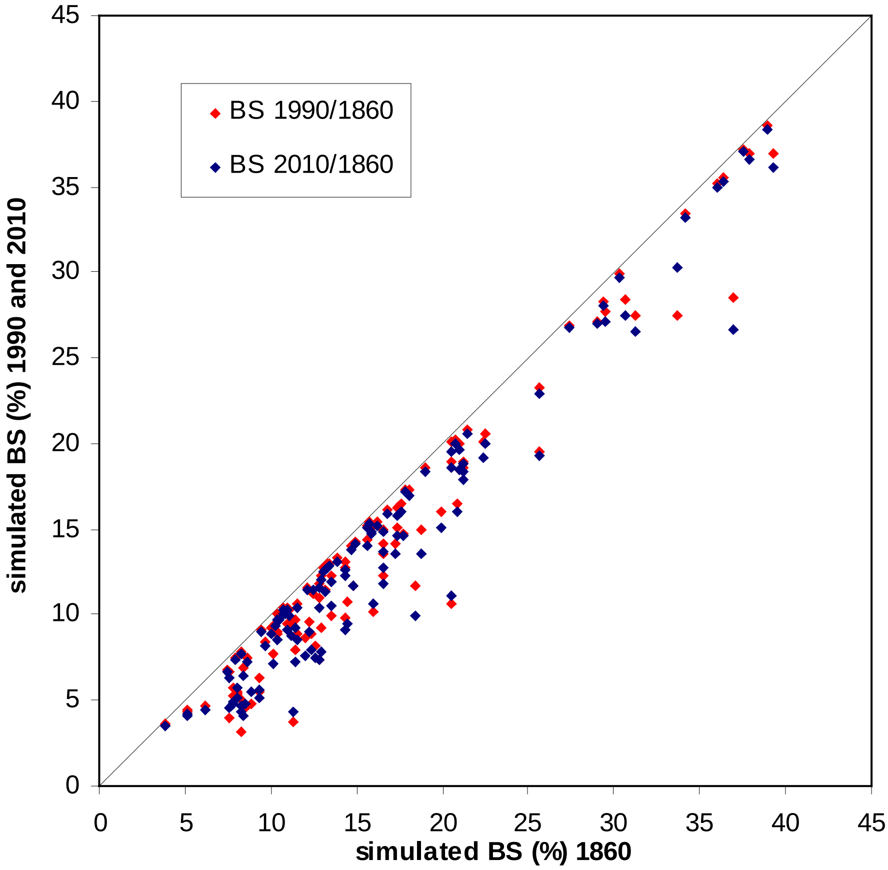


Figure 5-17 Simulated soil base saturation (BS) in the lake catchment soils for 1997 and 2010 compared with simulated pre-industrial BS (1860). Dots below the 1:1 line have acidified since 1860.

This may raise a need to refine the regional prediction, because there are other valid management questions focussed specifically to a subset of the modelled lakes, e.g. the most sensitive lakes. If so, then further effort must be focussed on modelling these lakes in a greater detail, because their future development may not be correctly captured even if the mean of the modelled and observed lake population is predicted correctly. This could be done by stratification of the data set and refinement of the input data specifically for the different categories.

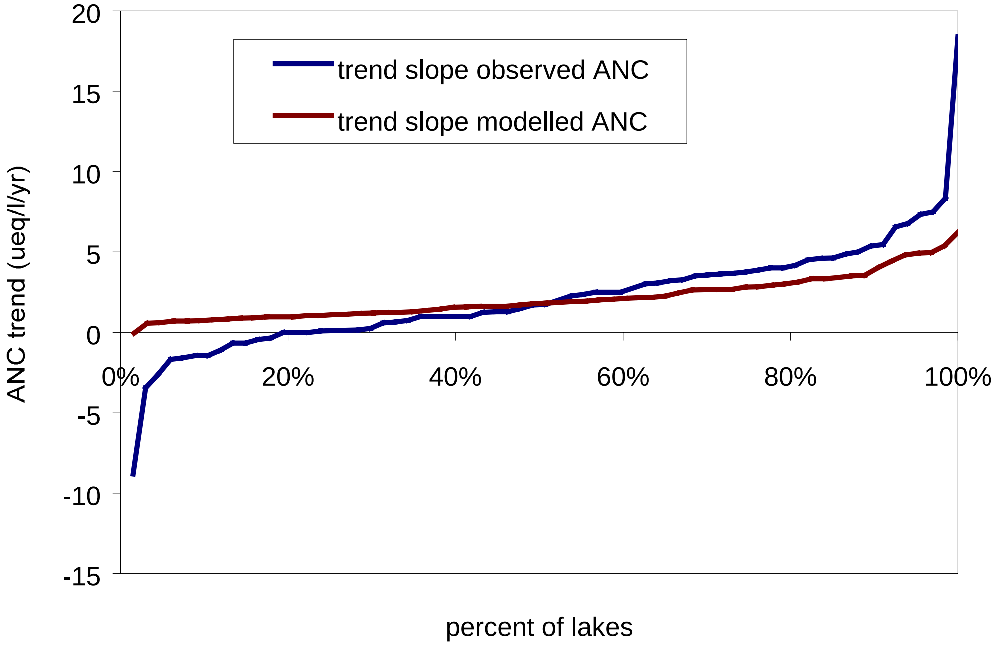


Figure 5-18 Ranked slopes of the trends in ANC (μeq/l/yr) for the lakes common to the modelling and Skjelkvåle's investigation.

The main conclusion from the modelling of the lakes presented above is the relatively high degree of internal consistency of the monitoring data used for the modelling. The data on soils, deposition and lakes were collected with different time and spatial resolution, with different aims and over different periods of time. However, the results obtained using this data set proved that they are suitable as input to modelling applications. The second conclusion is the performance of the MAGIC model. The model was successfully applied to a relatively large number of sites without exaggerated demands on manpower. This opens possibilities of applying the model to a larger number of lakes, for refining the model calibration and testing multiple scenarios. The MAGIC contains also several features, which were not exploited in this calibration as yet such as a fish toxicity module or a module for estimating the future acid episodes and a module to calculate dynamic critical loads functions as exemplified in section 5.3.2. This opens possibilities to broaden the questions which can be answered by the regional MAGIC model calibration. Thirdly, some preliminary conclusions could be drawn about the results of the calibration to the 122 lakes. The chemistry of the lakes improved as the deposition decreased. This demonstrates the benefits of the deposition reduction which has occurred from 1990 to 1997 and which are expected to occur by 2010 under the Gothenburg protocol. It also showed, that in particular for the recovery of the catchment soils, there will not be much of general improvement by the year 2010. Recovery of the soils will require either much longer time or even more reduced deposition and/or different land use or both.

5.2.1.6 TIME SERIES ANALYSIS OF NATIONAL MONITORING DATA

Analysis of over 100 Swedish Reference Lakes has shown an average recovery in alkalinity of ca 1 ueq/L/yr between 1983 and 1997 (Wilander and Lundin, 2000). In the more intensively studied PMK catchments (Figure 5-19), stream water concentration data shows a recovery in buffering capacity in the south of Sweden, with less consistent change in other parts of the country. (Fölster and Wilander, 2001) The declines in sulphate are compensated to some extent by declines in base cations.

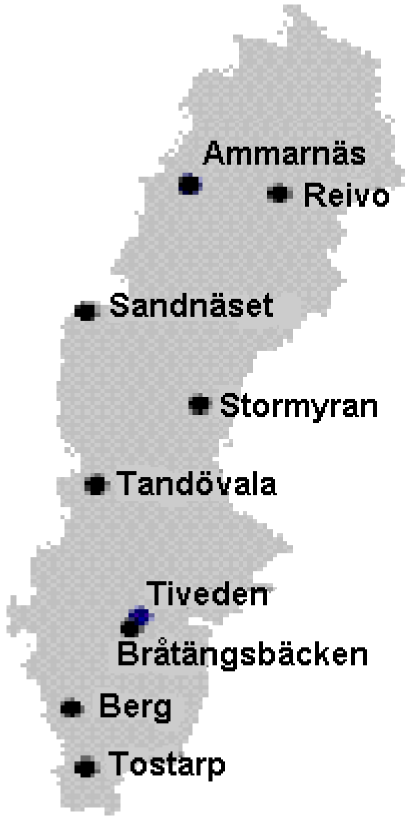


Figure 5-19 Map of Sweden with locations of study sites.

Time series of fluxes from the catchments show a similar, but less distinct pattern. (Fölster, et al.; in press) The recent decline in S deposition has been accompanied by a reduced transport of SO_4^* (* = non marine) from most of the investigated catchments. In chronically acidified sites in southern Sweden, the SO_4^* decline was partly balanced by decreases in BC^*, and only a small recovery from acidification, in terms of increasing ANC, was found (Figure 5-20). In northern Sweden, runoff acidity status is mainly controlled by natural variations in climate, and no response to the decline in SO_4^* was found. A summary of observed changes in annual fluxes in Swedish reference streams is presented in Table 5.1.

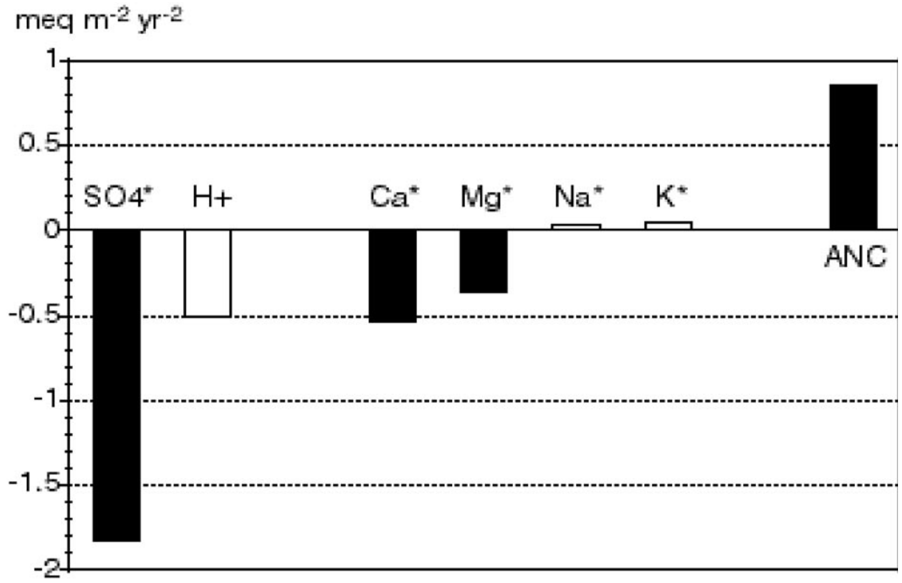


Figure 5-20 Average trends in stream transports (annual fluxes 1986-1999) from the four chronically acidified PMK sites Tandövala, Tiveden, Bråtängsbäcken and Berg in Southern Sweden. Filled bars indicate statistically significant changes. The recent decline in SO_4 deposition has been accompanied by a reduced transport of SO_4^* (* = non-marine) from most of the investigated catchments. In chronically acidified sites in southern Sweden, the SO_4^* decline was partly balanced by decreases in BC^*, and only a small recovery from acidification, in terms of increasing ANC, was found. In northern Sweden, runoff acidity status is mainly controlled by natural variations in climate, and no response to the decline in SO_4^* was found (From Fölster et al., in press).

Table 5.1 Annual changes in transport of Swedish reference streams from 1986-1989 until 1999. Bold figures indicate statistically significant changes. Underlined sites are chronically acidic.

Site	SO4*	H+	ANC	BC*	Q
	meq m-2 yr-2	meq m-2 yr-2	meq m-2 yr-2	meq m-2 yr-2	mm yr-2
Ammarnäs	-0.85	0	-3.74	-4.49	-2.1
Reivo	-0.18	0	-0.74	-0.98	-2.2
Sandnäset	**-0.78**	0	-6.07	**-6.85**	-3.1
Stormyran	0.49	-0.03	2.17	3.1	21.0
Tandövala	**-1.16**	**-0.83**	**0.89**	-0.47	-1.6
Tiveden	**-2.93**	-0.78	**1.35**	-1.47	4.0
Bråtängsbäcken	**-2.84**	-0.31	**1.11**	-1.62	4.0
Berg	-0.39	-0.09	0.04	0.15	12.1
Tostarp	6.55	0.05	1.76	9.8	25.1

The low rate of recovery at the acidified sites means that recovery will be prolonged over several decades. This implies that the deposition has to be reduced further, to levels well below the critical load that an *unacidified* site can tolerate without damage, if a recovery is to take place more quickly, or at all.

One factor prolonging the rate of recovery is the mobilisation of sulphate that had earlier accumulated in catchments. This mobilisation is evident from the sulphur input/output budgets from the PMK catchments (Figure 5-21) where SO_4 is being lost all across the country, but with the greatest net annual losses in the south.

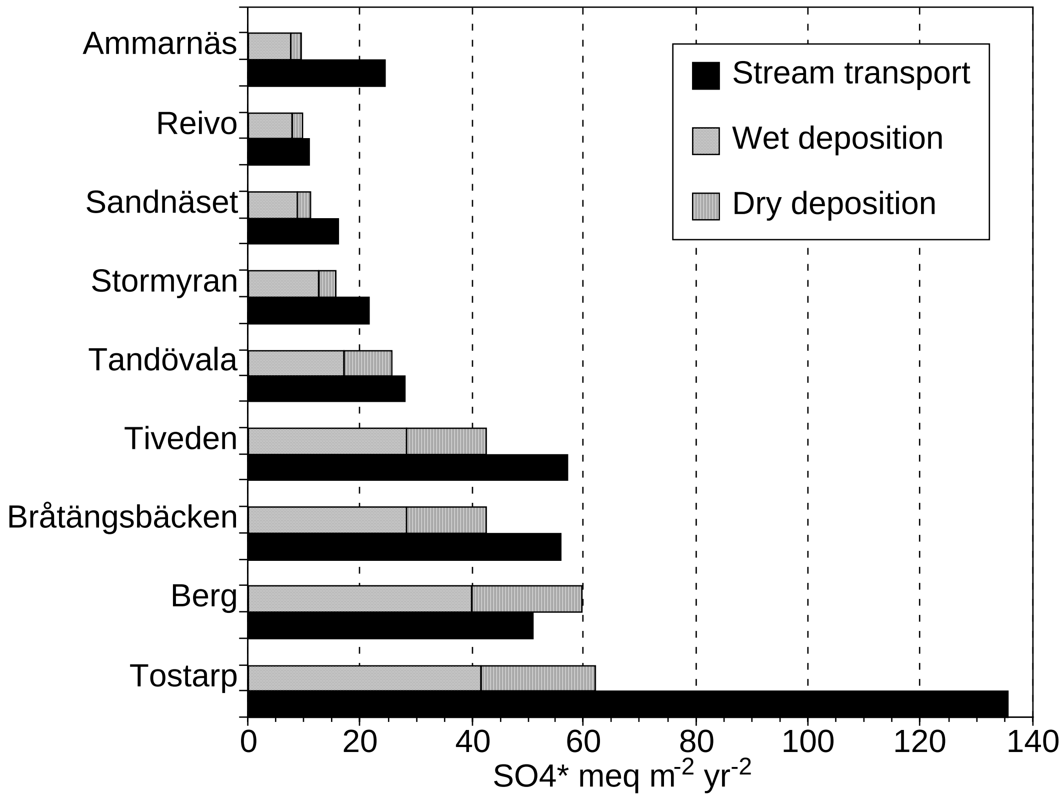


Figure 5-21 Budgets of non marine sulphate, SO_4*, 1995-1999. Bulk deposition from MISU/IVL. Although dry deposition estimates are preliminary, the budgets clearly show the loss of SO_4 from Swedish catchments across the country.

Soil solution data from PMK catchments also show a decline in sulphate and an increase in ANC. (Fölster and Bringmark, in press). Soil water concentrations of SO_4 have decreased at all sites as a response to the decreasing SO_4 deposition. The changes were much higher at the southern sites compared to the north. The SO_4 decreases in E-horizons were smaller than in deposition, which was an indication of mineralisation of organic sulphur in the O-horizon. The negative trends in soil water SO_4 were largely balanced by decreases in base cations but there were also tendencies of recovery from acidification in soil solution in the south with increasing pH and ANC. This was however contradicted by increasing Al concentrations. A high influence of marine salts in the early 1990-ies may have delayed the recovery but did not change the long term trend. Decreasing trends in the ratio $Ca/(H^+)^2$, are most pronounced in Tiveden, suggested that the soils were getting more acid although soil solution tended to recover.

An interesting analysis of the PMK data was to employ a technique proposed by Kirschner and Lydersen (1996) that interprets the relative changes in different anions in terms of the changes in soil chemistry. This analysis (which is being worked on in the final year of ASTA) suggested that soils in the sensitive catchments from southern Sweden were still losing base cations (Fölster, pers. comm.)

Our evaluation of stream trends depends on a correct understanding of soil processes, and recent studies show the need for a better understanding of these. Findings from the experimental "roof" catchment at Gårdsjön (Torssander and Mörth, 1998) and from the present program for Integrated Monitoring (Löfgren et al., 2001) have shown the need for a quantification of the mineralisation of organic sulphur, a factor that was earlier neglected. The relevance of a hypothesised surface water recovery accompanied by continued soil acidification (Matschonat and Vogt, 1998) for acidified regions in Sweden should be further investigated. See also discussion in Ch 5.2.1.2.

5.2.1.7 MAGIC MODELING OF PMK CATCHMENTS

The geochemical model MAGIC (5.01) was applied in detail to the Berg catchment in South-western Sweden (Krám *et al.,* 2001a). This catchment and another five PMK-catchments along the length of the country were modelled using two scenarios of atmospheric deposition (Krám *et al*., 2001b). Since these were the first applications of MAGIC in the north of Sweden, this model was also used to model one lake in N. Sweden where paleoecological reconstructions of pH history going back several hundred years were available to constrain/test the MAGIC hindcast (Krám *et al.*, 2001c). A key question for this application of MAGIC was whether the relatively stable lake pH indicated by paleolimnological methods could be consistent with the MAGIC model of how acid deposition has affected the forest ecosystem at a time when forest harvesting also contributes to the acidification pressure. It appears that anthropogenic atmospheric deposition and forest growth can be consistent with the slight changes in lake water pH indicated by the diatom record (Krám et al., 2001).

Figure 5-22 The location of the six PMK sites where the MAGIC model was applied. The lake application with calibration against paleolimnological data was in the vicinity of the Vindeln PMK site.

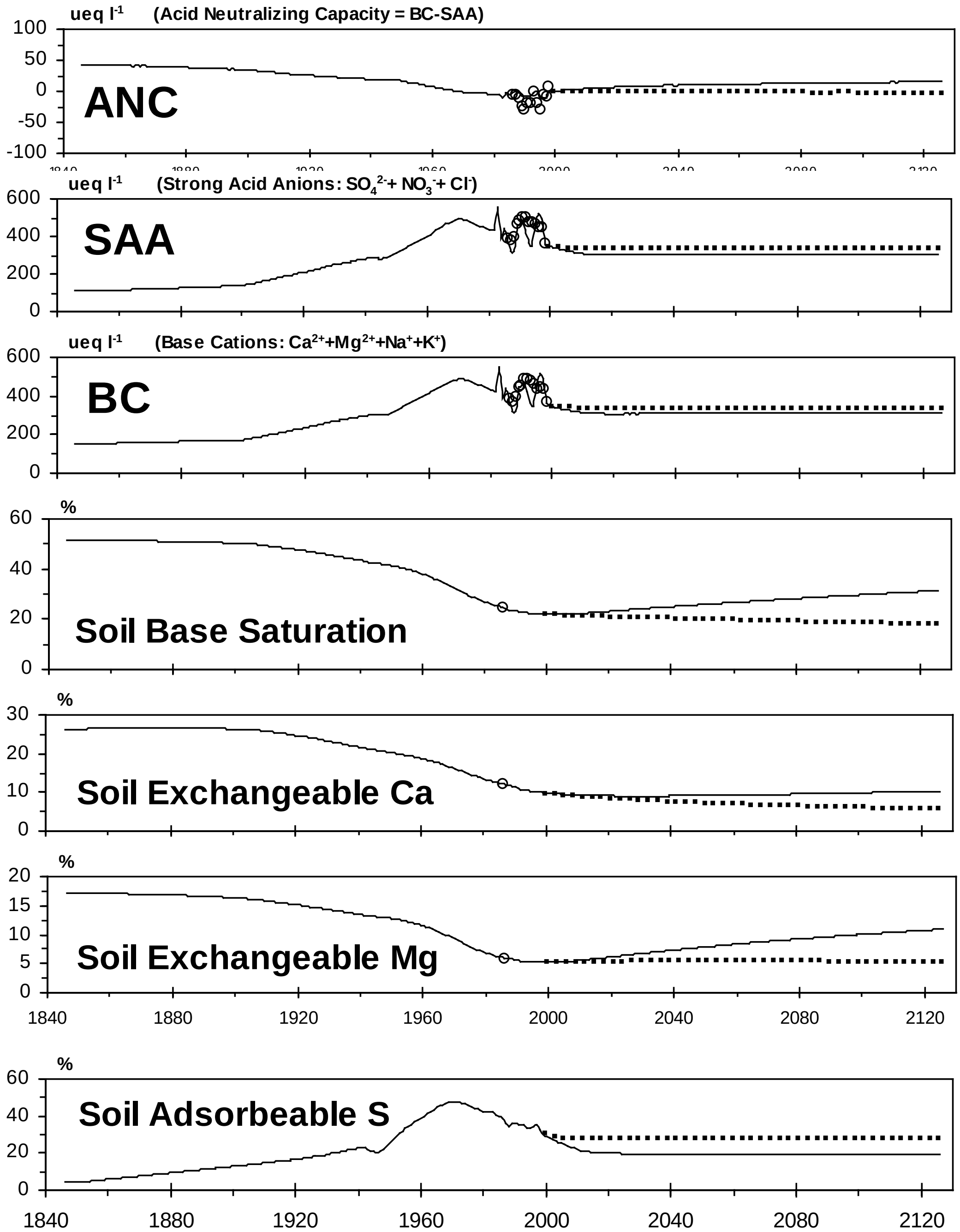


Figure 5-23 MAGIC model simulation results of the annual mean stream water and soil concentrations at Berg from 1846 to 2126. The impacts of implementing the Gothenburg Protocol on atmospheric deposition reductions of sulphate, nitrate and ammonium in 1999-2010 are shown in full lines. Results of the "business as usual" scenario are in dashed lines. Observed annual mean concentrations are shown in circles.

MAGIC was applied to the PMK sites (Figure 5-22) using two simulation forecasts for 1999-2010. One simulated implementation of the “Gothenburg Protocol” (decline in deposition of sulphate, nitrate and ammonium). Using 1995 as the Baseline year and 2010 as the Target year, this gave a decline of non-marine sulphate by 31-54%, a decline of nitrate by 35-41%, and a decline of ammonium by 0-2%. The other scenario used was "Business as Usual" with the 1995 Deposition continuing from 1999-2010. The simulations of the PMK catchments benefited from the up to two decades of data on runoff, deposition and soil chemistry data. One example of the results is shown for the Berg Catchment in Southern Sweden (Figure 5-23).

The experience of applying MAGIC to catchments across Sweden (Figure 5-24) confirmed the extensive acidification of sensitive catchments in southern Sweden, while also indicating the much lesser degree of chronic acidification in northern Sweden where deposition has been significantly lower. Anthropogenic deposition was the dominant factor causing stream water acidification in the southern catchments (Tresticklan, Tiveden, Berg). At the two catchments with extremely shallow soil (Tresticklan, Tiveden) also the tree uptake of nutrients contributed significantly to the acidification. Only slight long-term acidification was found in the three northern catchments (Reivo, Sandnaset, Vindeln). It should be noted though, that episodic acidification during spring flood, when much of the buffering capacity of the soil was bypassed, is not reflected in these MAGIC simulations of mean annual chemistry. Separate episode studies have simulated the increase in this episodic acidification and the subsequent recovery concomitant with declining deposition (Laudon and Hemond, 2002; Laudon and Bishop (in press)).

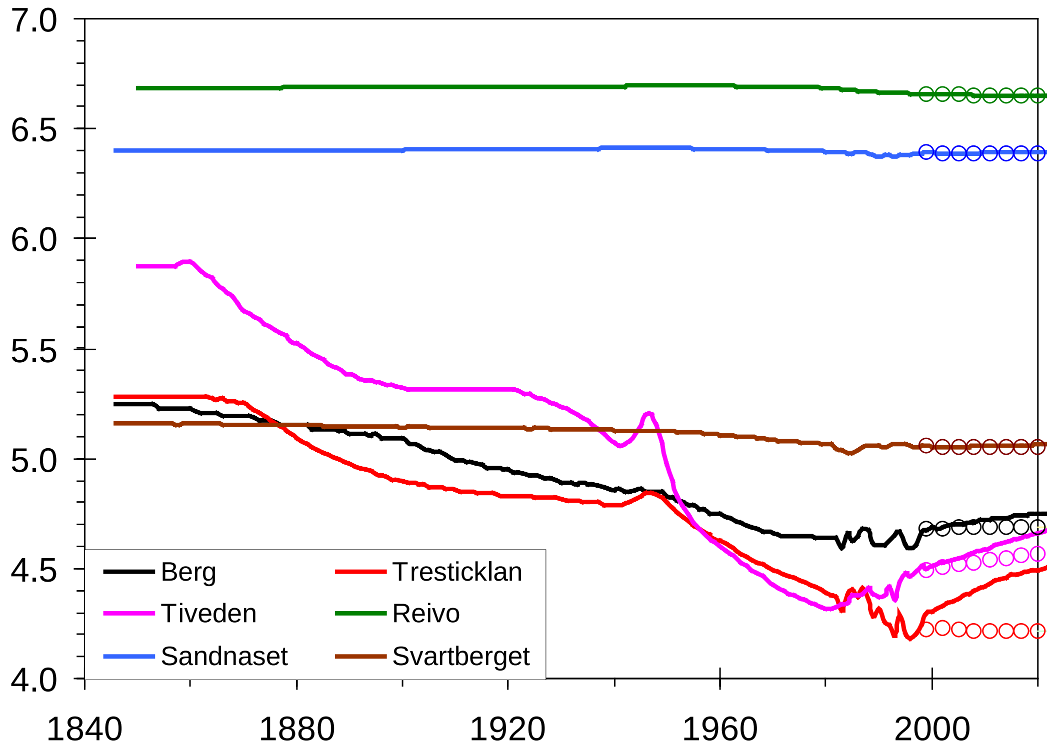


Figure 5-24 Hindcasts and forecasts of stream pH at the PMK sites. The Gothenburg Scenario is the solid line from 1995 to 2010. The "business as usual" scenario with stable deposition form 1995 onwards is represented by the open circles.

The difference in acidification recovery resulting from the “Gothenburg Protocol” as opposed to “Business as Usual” scenario showed the most significant increase in acidification recovery rate in the two southern catchments with very shallow soils.

5.2.2 Couplings between nitrogen deposition and acidification

Since nitrogen is a nutrient, the role of nitrogen deposition, i.e. nitrate and ammonium deposition, in soil acidification is much more complex than that of sulphate. Low levels of nitrogen deposition do not cause acidification and slightly increased deposition levels do not directly increase soil acidification as long as the vegetation can take up the extra nitrogen. There is, however, an indirect effect, since an increase in nitrogen uptake also leads to an increase in base cation uptake. At high nitrogen deposition levels the ecosystem can not absorb the additional nitrogen and the nitrate concentration in the water leaving the rooting zone increases which in turn leads to soil acidification.

In the long-term perspective the ecosystem can not make use of more nitrogen than what is physiologically possible to store in the vegetation. Temporarily however, it has been shown, e.g. with the new MakeDep model, that it is likely that a forest ecosystem can withstand a nitrogen deposition above the long-term acceptable level for many years. At first the extra nitrogen is used to increase the nitrogen content of the vegetation and secondly due to litter fall the carbon to nitrogen ratio in the soil decreases.

The time lag, however, between an increase above the long-term acceptable level of nitrogen deposition and effects on soil water chemistry is unfortunately to a large extent uncertain. Furthermore, the time lag between a change in soil water chemistry and effect on vegetation is even more uncertain. However, the absence of short term effects of elevated nitrogen deposition on forest ecosystem health can not be taken as an evidence that the critical loads are not exceeded.

Since the sulphate deposition has decreased regionally to a large extent (≈50%), the relative importance of nitrate has increased. In the NITREX experiment at Gårdsjön, 40 kg of NH_4NO_3-N per year has been added for 11 years to the catchment G2 NITREX. The primary purpose was to study the risk of nitrogen saturation indicated by leaching of NO_3 to the groundwater and runoff. At the beginning, the catchment was one of the two most nitrogen-limited sites across the European gradient investigated within the EU project NITREX. Over the same time period as the N was added, the ambient deposition of S decreased by more than 50%. The SO_4 runoff concentrations decreased at the G2 NITREX from 350 to less than 100µeq/l, while NO_3 increased from 0 to over 50 µeq/l. Provided that about 50µeq/l of SO_4 in the runoff come from the sea salt, the nitrate became over 10 years with experimentally increased N deposition equally important from the acidification point of view.

5.3 Application of new concepts for critical loads of acidification

There are many problems to overcome in order to use dynamic aspects rather than only steady-state calculations of critical loads and exceedances in integrated assessment modelling. The three major problems are most likely 1) the additional need for input data 2) the additional computation time needed and 3) the problems in defining unique and consistent targets. All of these problems have been addressed by the ASTA programme.

The problem with the additional need for data was addressed by supporting national efforts in Europe to perform regional dynamic assessments. The support has been in the form of 1) joint projects with local scientists, as with EKG-GeoScience in Switzerland to further develop the model set-up and investigate possibilities to estimate the uncertainty in dynamic assessments and with OEKODATA to perform a regional dynamic assessment of German forest soil chemistry, 2) co-organising of training workshops for local scientists, as in Hungary, Croatia and Slovenia, and by 3) extensive contacts with local scientists to help investigating the potential for a national regional assessment and to help finding funds to organise a local project, as in the Czech Republic and Poland.

5.3.1 Dynamical models in soil critical load calculations

The critical loads that are currently used include the assumption that the ecosystems are in a steady state condition where temporary storages of acidifying substances or base cations do not change or affect the critical load. This is true at a larger time scale and good enough for a first estimate of what acid deposition levels the ecosystems can tolerate. But when the deposition decreases and approaches the critical load, dynamic modelling becomes important to estimate the relation between the time it takes for an already acidified ecosystem to recover and the decrease in deposition below the critical load. As long as the deposition is at the critical load, by definition, no recovery or additional acidification occurs. To reach recovery the deposition must be further decreased and the time to recovery to an acceptable condition is strongly dependent of the extent of the decrease in deposition below the critical load. The Joint Expert Group on Dynamic Modelling (JEG) recommends that dynamical modelling should be carried out at all areas where critical loads have been exceeded. One of the conclusions from the JEG was that it would not be possible to directly incorporate dynamical models into the integrated modelling framework. This is mainly due to a large increase in complexity which would require significant modification of the whole model system and, if successful, very large computer resources.

A solution for the problem with time-consuming dynamic model calculations was presented at the first expert meeting on dynamic modelling in Ystad and demonstrated at the Acid Rain conference in Japan. Rather than incorporating the entire dynamic model into the integrated assessment model system, something which is very unlikely to be successful, it was suggested that dynamic models are to be used to calculate isolines of ecosystem recovery and that these isolines, e.g. in the form of look-up tables, are to be used in the integrated assessment modelling. Such isolines could for example include the recovery time as a function of sulphate and nitrogen deposition reductions or the recovery time as a function of a deposition reduction and the timing of such a reduction. It was noted that non-linear effects are to be expected in these isolines and that for example a fast drastic reduction in deposition might lead to slightly longer recovery times than a somewhat less drastic deposition reduction. These non-linear effects are dependent on the definition of ecosystem recovery as the point in time where the chosen chemical criteria is no longer violated and that the recovery endpoint is not considered. A drastic deposition reduction may temporarily lead to a worse situation as the top soil layers start to recover and increase their base saturation thus reducing the flow of base cations to the lower soil layers. In the long run and within reasonable limits a larger reduction in acidifying deposition will always result in a better soil chemistry than a smaller reduction.

The non-linearity effect is one of the aspects of the problem in defining consistent targets with dynamic models. Another aspect is the problem in defining a recovery time. If the recovery time is the time it takes until the BC/Al ratio is no longer exceeded, it is often implicitly understood that the BC/Al ratio will remain above the critical limit. This might not be the case, however, as the management practice influences the soil solution chemistry through its influence on dry deposition and nutrient uptake. Events in the management practice might temporarily drive the BC/Al ratio above the critical limit after which it decreases below the critical limit again. It is e.g. obvious that a forest clear-cut will temporarily change the soil chemistry dramatically. The chemical criteria may thus fluctuate around the critical limit. Consequently, the non-violation of the chemical criteria in the near future does not preclude a violation of the criteria at a later date. One possibility would then be to demand that the chemical criteria are not violated during an entire rotation period.

A third aspect is that if it takes too long for sites to recover there is a risk that the recovery times can not be used in the policy process. A solution to this problem may be to use an analogue to "gap-closure" as used in the negotiation of earlier protocols. The recovery can then be expressed in relation to a reference year, e.g. time to increase the chemical criteria to a value halfway between the reference year and the wished target (called half-recovery time).

A problem in calculating the recovery time for a site is to find a unique definition of such a time. In simulations where the BC/Al ratio fluctuates a possible definition of recovery is the point in time after which the BC/Al ratio will not be violated again.

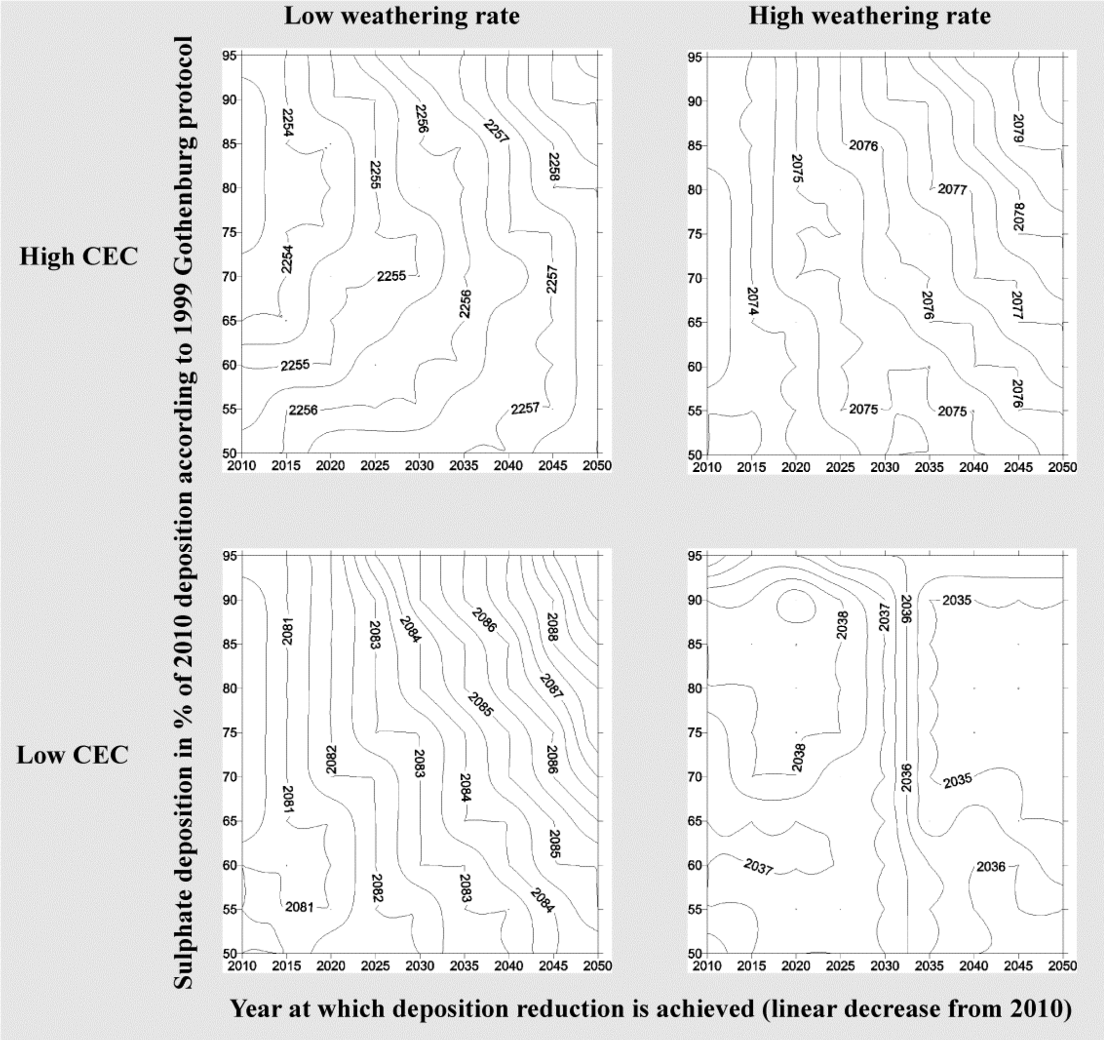


Figure 5-25 Recovery isolines showing at what year recovery (BC/Al-ratio equals one) takes place, given deposition reduction (on y-axis) and the year this reduction is implemented (x-axis).

5.3.2 Dynamic recovery in lake critical load calculations

For lakes an alternative approach for critical load calculations has been explored. The MAGIC model can be used to calculate dynamic critical loads functions for lakes. The model output can be set to present the maximum tolerable deposition of sulphur or nitrogen for different chemical criteria in a specified year. If recovery is defined as an ANC at or above a certain level and a year when this recovery should have taken place is defined, MAGIC will give a function showing the highest allowed deposition of sulphate and nitrogen at that specific lake. By performing model simulations for a large number of lakes in a region, a large set of critical load functions can be generated. These functions can then be aggregated into percentiles showing the highest allowed deposition at which e.g. 5% of the lakes in the region will exceed the specified ANC level in the specified year. In Figure 5-26, an example is given for a set of 122 Swedish lakes which were modelled using MAGIC (see section 5.2.1.5). The model was set to calculate the maximum allowable deposition of sulphur and nitrogen with the criteria that ANC does not decrease below 50 µeq/l in the year 2050.

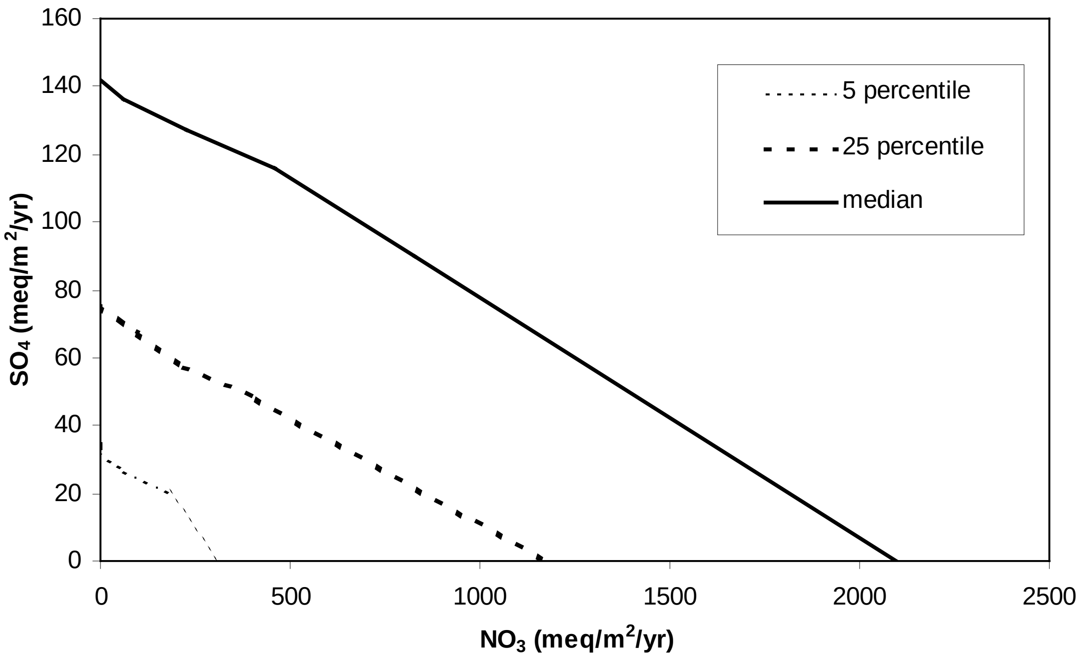


Figure 5-26 Critical loads function from MAGIC showing the highest allowed deposition of sulphate and nitrate if ANC > 50 μeq/l should be violated in 2050 in 5, 25 and 50% of the lakes.

Regional assessments of present day and future recovery are being done at a number of acid sensitive regions in Europe, using the dynamic acidification models. The currently available modelling tools are also capable of transforming these results into dynamic critical load functions, which are suitable for use within integrated assessment.

5.4 Eutrophication

5.4.1 Effects of nitrogen deposition on vegetation from coniferous forest ecosystems

The boreal forest is the largest terrestrial biome. We have studied the effect of N on only a few species present in boreal forest. These species are by no means threatened by extinction, e.g. they are not red-listed. Instead the species studied are all very common in understorey vegetation, and changes in their abundance are therefore likely to have large effects on central ecosystem processes. Unlike earlier experiments we added relatively small amounts of N to forest ecosystems formerly not affected by N deposition. With this approach we wanted to follow the early responses to increased N. The main experiment used additions of NH_4NO_3 at rates of 12.5 and 50 kg N ha^{-1} yr^{-1} to mimic N deposition. In an additional study, the effects of NO_3^- and NH_4^+ were studied separately.

In ecosystems like the boreal forest where the N supply is irregular and limited the vegetation is well adapted to take advantage of available N. When plants are exposed to N deposition the initial response is thus an increased N uptake. This increased uptake is accompanied by various changes of both the N and C biochemistry of the plants. Plants taking up N in excess of their basic need accumulate N as free amino acids, notably glutamine, asparagine and arginine. The bottom-layer vegetation in boreal forests consists of various mosses. Bryophytes depend on wet and dry deposition of N. They are therefore considered to be highly sensitive even to small changes in N supply. For example, the addition of 12.5 kg N ha^{-1} for three consecutive years caused arginine concentrations of *Pleurozium schreberi* to increase significantly (p=0.03, Student's t-test) (Figure 5-27). This indicates that the moss was not able to respond to N additions by increased growth, and instead N was accumulated in the form of arginine. High amino acid concentrations may be harmful to bryophytes, and has been shown to correlate with reductions in length growth of *Sphagnum* (Nordin and Gunnarsson 2000). Nitrogen induced changes in species composition of the bottom-layer vegetation may persist long after the N input has been terminated. In a forest fertilisation experiment terminated 50 years ago we found that *Hylocomium splendens* was less abundant in formerly fertilised plots (Strengbom et al. 2001).

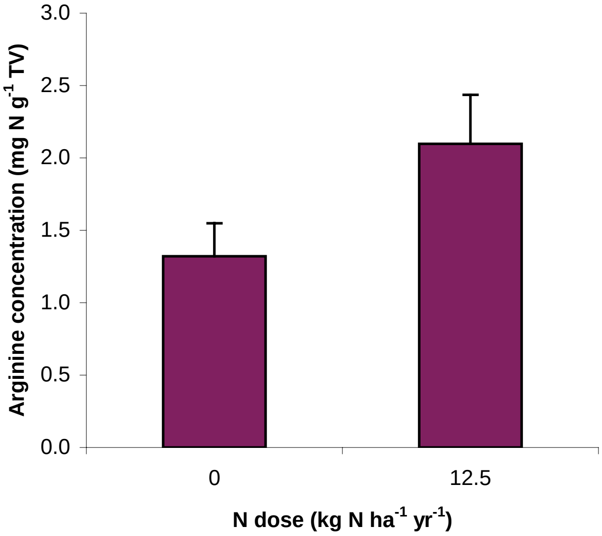


Figure 5-27 Arginine concentrations in shoots of *Pleurozium schreberi* after three years of N addition with 12.5 kg N ha^{-1}. Means (n=6)±SE.

Ericaceous dwarf-shrubs like *Vaccinium myrtillus* and *Vaccinium vitis-idaea* normally dominate the field-layer vegetation in boreal forests. N induced biochemical changes in these species are thus important since related processes may have large impact on vegetation structure. In response to N addition amino acid concentrations of *V. myrtillus* and *V. vitis-idaea* increase (Nordin et al. 1998, Strengbom et al. 2002). At the same time there is a decrease in levels of carbon based defence substances (Witzell et al. accepted ms). These biochemical changes predispose plants, in particular ericaceous plants, to damage by biotic and abiotic stresses. Our experiments show that *V. myrtillus* becomes heavily infected by fungal pathogens when exposed to N (Strengbom et al. 2002). These infections will decrease the viability of *V. myrtillus*, thereby giving the opportunity for the competing grass *Deschampsia flexuosa* to expand (Strengbom et al. 2002). The sequence of events following N deposition to the field-layer vegetation of a forest formerly not exposed to N is thus:

N-deposition ⇒ Increased uptake of N by the vegetation ⇒ Increased levels of free amino acids and decreased levels of carbon based defence substances in *V. myrtillus*

⇒ Increased damage by stresses, notably fungal pathogens ⇒ Increased growth of competing species such as *D. flexuosa*.

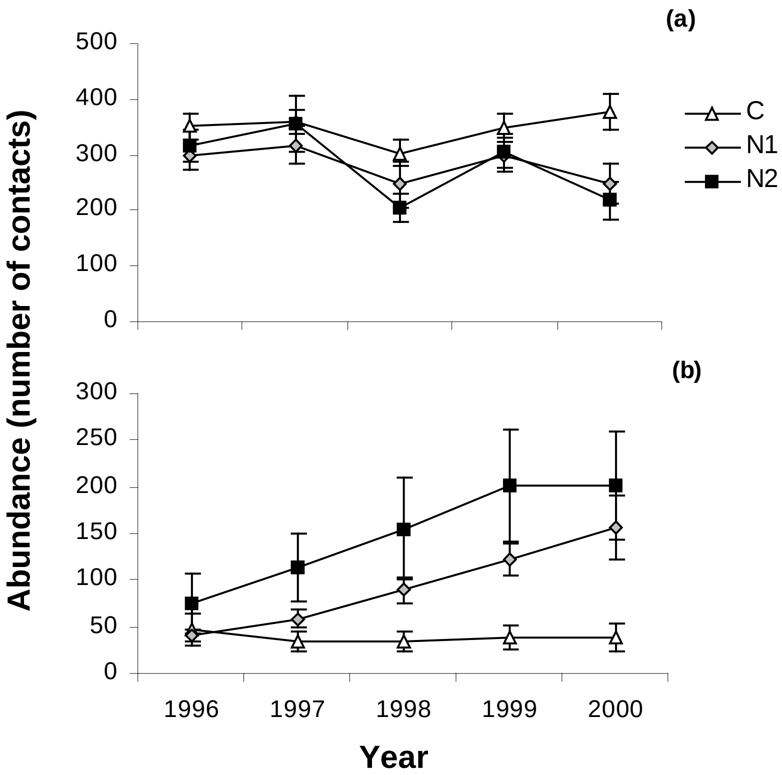


Figure 5-28 Vegetation responses to N additions (C = control, N1 = 12.5 kg N ha^{-1} year^{-1}, N2 = 50 kg N ha^{-1} year^{-1}) over five years in terms of abundance *of V. myrtillus* (a) and *D. flexuosa* (b). Means (n=6)±SE.

The resulting changes in field-layer species composition can persist for a considerable time after N input has been terminated. We found an increased abundance of *D. flexuosa* and a decreased abundance of *V. myrtillus* nine years after N fertilisation had stopped (Strengbom et al. 2001). Nearly fifty years following N fertilisation we, however, found no significant differences between control plots and formerly fertilised plots in the abundance of *V. myrtillus* and *D. flexuosa*, but disease incidence of *V. heterodoxa* on *V. myrtillus* leaves was still higher on formerly fertilised plots (Strengbom et al. 2001).

The rates of N deposition needed to initiate the process of vegetation change are difficult to exactly determine. In our study, significant effects on the cover of *V. myrtillus* and *D. flexuosa* occurred after three years in the high N treatment and after five years in the low N treatment. Moreover, the level of impact of high N and low N treatments were similar after five years of treatments. These results have led us to suggest that vegetation changes will occur at least at N deposition rates above *c.* 10 kg N ha^{-1}, yr^{-1} (Figure 5-28). Consequently the understorey vegetation of large areas of boreal coniferous forests may already be significantly altered by N deposition. A survey of the understorey vegetation of Swedish coniferous forests shows large differences in the abundance of ericaceous species between regions with different levels of N deposition, i.e. the abundance of e.g. *V. myrtillus* and *V. vitis-idaea* are much lower in regions with N deposition above 6 kg N ha^{-1} and year^{-1} (Figure 5-29, Strengbom et al. 2002). Furthermore, the frequency of fungal infection on *V. myrtillus* leaves is much higher in regions with high N deposition (Figure 5-29, Strengbom et al. 2002). This indicates that the processes shown to be responsible for the decrease of ericaceous species in experiments are active also in regions with high N deposition. These results clearly illustrate the potential benefit of combining experimental studies with large scale monitoring studies.

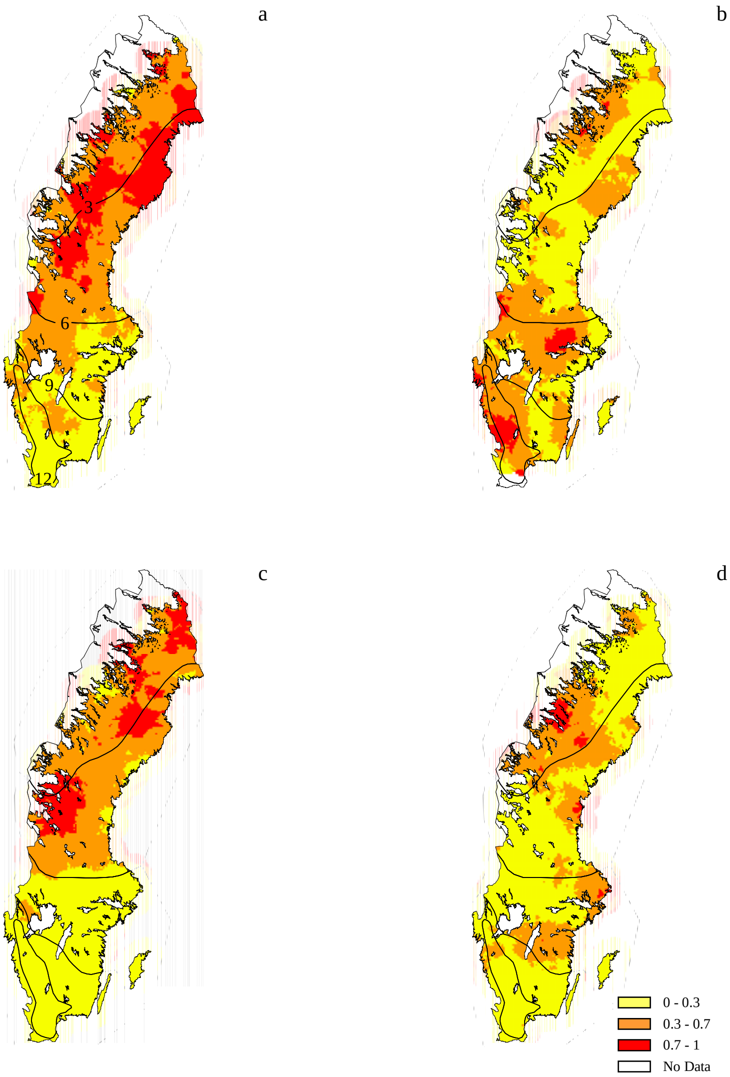


Figure 5-29 Frequency of subplots where the following were present: (a) *V. myrtillus*, (c) *V. vitis-idaea*, (d) *D. flexuosa*. Figure (b) shows the proportion of subplots in which *V. myrtillus* was infected by the fungal leaf pathogen *Valdensia heterodoxa*. The isolines represents from north to south N deposition of 3, 6, 9 and 12 kg N ha^{-1} year^{-1}.

A number of studies have indicated that not only the amount, but also the chemical form of N available to plants affects species composition of plants in different ecosystems. In boreal forests, soil solution N is dominated by organic N followed by ammonium while very low levels of nitrate are present (Näsholm et al 1998, Nordin et al. 2001). Thus, input of N from deposition will not only increase the amount of plant available N but also qualitatively change the pool of plant available N. Recent studies indicate that species differences in acquisition of N occur in the field resulting in a partial separation of N niches between species (McCane et al 2002). We have studied

N uptake from different chemical forms both in short- and long term experiments and both in laboratory and field settings. From these studies we conclude that:

- the capacity to absorb amino acids is widespread among plant species of both deciduous and coniferous forests (Figure 5-30, Persson and Näsholm 2001, Falkengren-Grerup et al. 2000)
- amino acid uptake is regulated differently than inorganic N uptake. In contrast to the uptake of inorganic N, uptake of amino acids is induced by the substrates (Figure 5-31, Persson and Näsholm, accepted ms)
- large differences between species in their capacity to utilise nitrate suggests that this N form has a special role for promoting vegetation changes (Olsson 2002, Persson et al. manuscript)

Together, these studies suggest that the composition of the plant available N pool has a direct effect on species composition in both coniferous and deciduous forests.

The quantitative role of the different N forms for nutrition of forest plants is still under debate. Our studies point to a more or less ubiquitous capacity to absorb such N forms and suggest soil N dynamics to be decisive for the N uptake of plants. This appreciation has led us to test different amino acids as N sources in conifer nurseries (Öhlund & Näsholm 2001, Öhlund & Näsholm submitted). The results from these studies have been utilised in the formulation of an alternative fertiliser to be used e.g. in conifer nurseries. This new fertiliser results in as good or better growth of seedlings while simultaneously minimising losses of N to the environment.

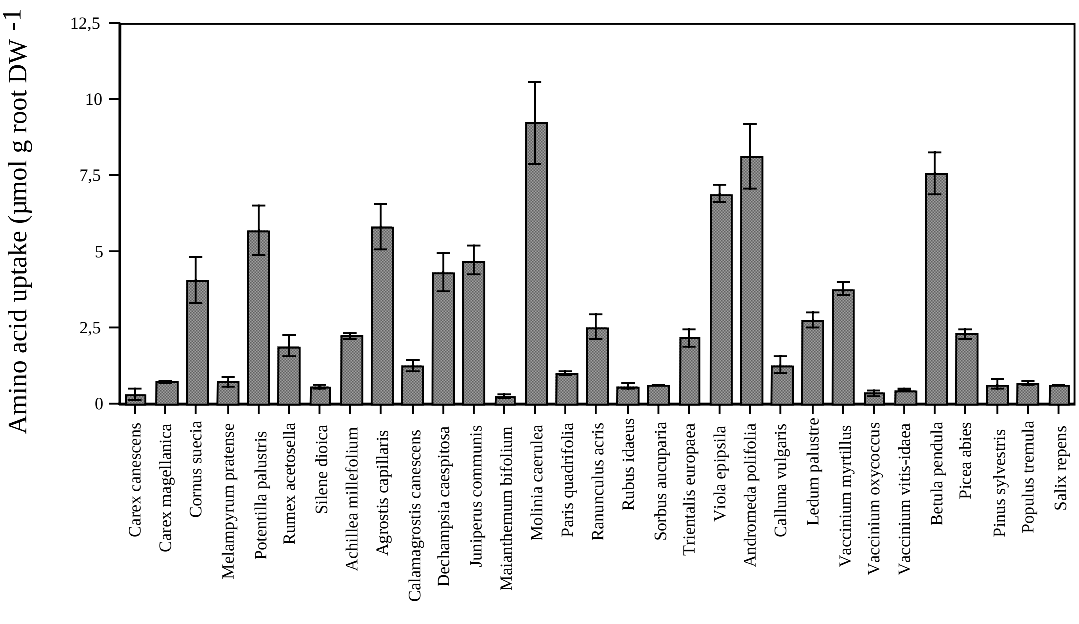


Figure 5-30 Uptake of amino acids by a range of different boreal forest species. Field-collected roots were incubated in solutions containing a mixture of amino acids and uptake assessed by GC-MS.

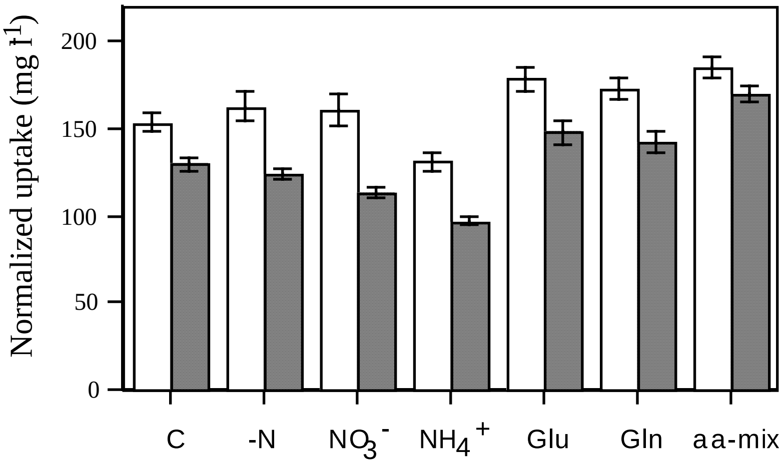


Figure 5-31 Uptake of amino acids by Scots pine seedlings grown either at high (grey bars) or low (white bars) N levels. Seedlings were pre-treated with different N sources before uptake measurements. Values refer to total aa uptake from a solution containing a mixture of amino acids from as assessed by GC-MS.

N deposition is generally in the form of NH_4^+ and NO_3^- and both inorganic N forms are deposited in approximately equal amounts. A central question when predicting future vegetation responses to N deposition is thus, the relative contribution of these two N forms to the observed effects. Scenarios of N deposition points to decreased emissions of NO_x but more or less constant emissions of NH_3. Thus, the relative rates of deposition may, according to current scenarios, change in favour of NH_3/NH_4^+. We studied the separate effects of NH_4^+ and NO_3^- on vegetation. After four years of N addition the results point out a critical role of NO_3^- in the increase of grass. *Deschampsia flexuosa* biomass and flowering was significantly higher on plots treated with NO_3^- than on plots treated with NH_4^+ (Figure 5-32), whereas *V. myrtillus* growth was unaffected by both N treatments (data not shown). Thus, the form of N deposited may be critical to the vegetation effects, and a decrease in NO_3^- deposition may give larger effects on the understorey vegetation than what could be expected from just the reduction in total N deposition.

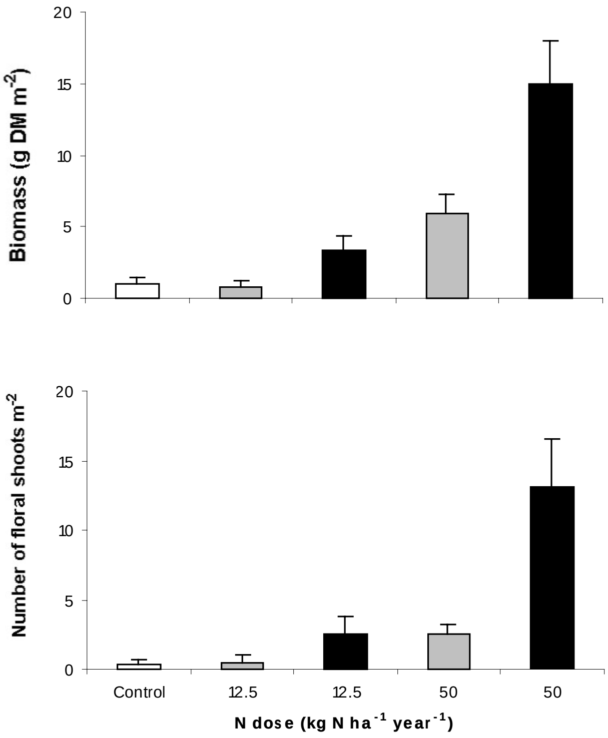


Figure 5-32 Effects of three years of addition of N as ammonium (grey bars) or nitrate (black bars) on the biomass (top) and flowering (bottom) of the grass *Deschampsia flexuosa* in an old-growth, coniferous forest. Means (n=8)±SE.

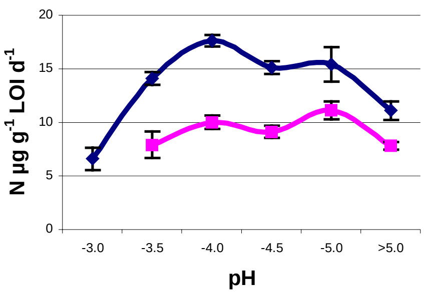
5.4.1.1 RESULTS FROM NEMORAL FOREST ECOSYSTEMS

Deciduous forests cover a substantial part of southernmost Sweden south of Limes Norrlandicus. These forests, unlike coniferous forests, usually lack a distinct humus layer. This is partly because deciduous trees demand a more fertile and less acid soil and partly because the properties of the litter are less acidifying than that of coniferous trees. Results from boreal and nemoral forests are therefore not directly interchangeable.

The purpose of the monitoring project was to calculate the effects of soil acidity and eutrophication in deciduous forests, which had relatively similar climate but varied in historic and modern deposition. This is a considerable achievement in comparison to earlier studies of deposition gradients, which have included a limited amount of sites spread over Europe or North America. The present-day deposition varies with more than 10 kg ha^{-1} y^{-1} among the studied regions which makes it possible to calculate effects of varying doses of nitrogen on soil chemistry and biological processes and vegetation. We have focused on effects on nitrogen mineralisation in the soil and the microbial community, the effects of ammonium and nitrate availability that changes both in absolute and relative amounts, the risk of nitrate leaching that may be affected by the understorey and how plants can be characterised in plant functional groups and as indicator species that respond to soil acidification and eutrophication.

It is evident that nitrogen mineralisation has increased considerably with nitrogen deposition, being twice as high in the regions with 17 as compared to 8 kg N ha^{-1} y^{-1} (Figure 5-33A). It is, however, most important to consider the acidity of the soil as the largest differences were found in the soils with low pH which indicates that soils with low pH are strongly responding to the nitrogen deposition. The accumulated nitrogen in the soil is probably the reason why nitrification too was enhanced in the most exposed regions. The nitrification process is probably substrate limited, and when ammonium/ammonia increase in the soil then more nitrate will be formed. Another hypothesis is that the nitrifiers are acid-sensitive and nitrification therefore should cease when soils were acidified, which is contrary to our findings. We found that the rate of nitrification was higher in regions with the highest deposition and that nitrification occurred in all but the extremely acid soils in these regions (Figure 5-33B). It is obvious that the higher the amount of accumulated nitrogen deposition the higher is the availability of both nitrate and ammonium. The increased availability of nitrate is important, as nitrate seems to have a selective impact on plant competition and species composition.

A



B

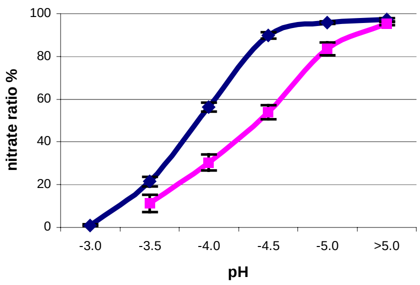


Figure 5-33 Potential net nitrogen mineralisation related to soil pH (0.2 M KCl) in south Swedish regions exposed to a modelled deposition of 17 kg N ha^{-1} y^{-1} (blue) and 8 kg N ha^{-1} y^{-1} (red). Means ± SE. A. Amount of mineralised nitrogen in a 15 week incubation experiment in the laboratory calculated per gram loss of ignition (LOI) and day. B. Degree (%) of mineralised nitrogen as nitrate (Falkengren-Grerup et al. 1998 and unpublished results).

Several soil and plant parameters seem to be related to nitrogen deposition. The most exposed areas not only have higher soil nitrogen mineralisation and nitrification rates

(Figure 5-33) but also a lower soil C:N-ratio and lower number of species whose anatomy is more broad-leafed than sclerophytic (Figure 5-34). The higher nitrogen deposition is also reflected in higher Ellenberg N-values of the vegetation and higher nitrogen concentrations of the leaves and growth rates of the understorey species (Figure 5-34). We thus have several soil and plant parameters that change in response to the south Swedish deposition levels in spite of these being relatively low in comparison to European conditions

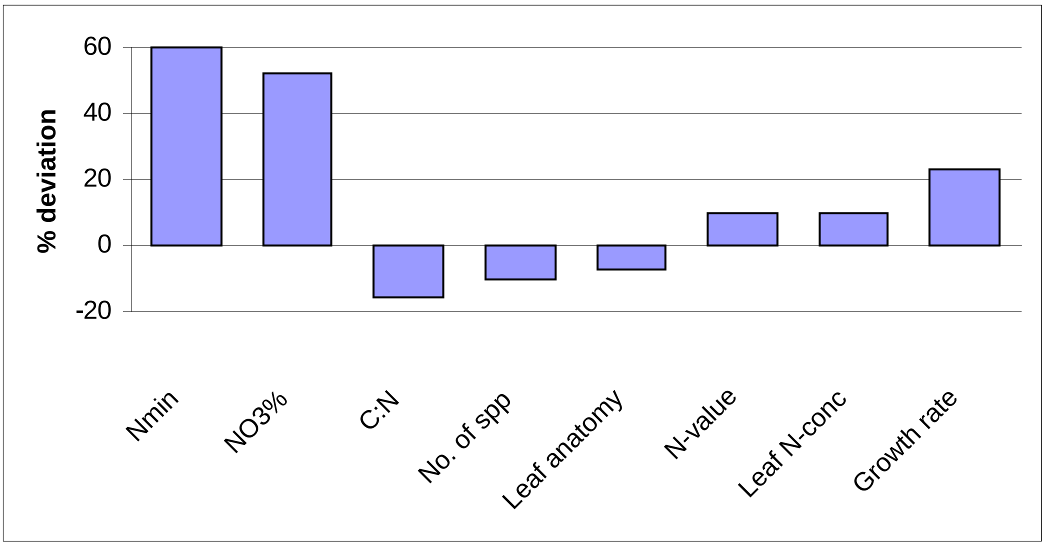


Figure 5-34 Differences (%) between regions exposed to 17 kg ha^{-1} y^{-1} and 8 kg N ha^{-1} y^{-1} in the soil processes nitrogen mineralisation (Nmin), degree of nitrification (NO_3 %) and C:N ratio, number of understorey species (spp) and the plant functional types leaf anatomy (increasing scores with degree of scleromorphy), Ellenberg N-values, nitrogen concentration in the leaf and maximum growth rate (Diekmann & Falkengren-Grerup 2002, Falkengren-Grerup & Diekmann, subm.).

The question is why the vegetation changes towards fewer but more nitrogen demanding species of taller stature. Several of our studies show that the form of nitrogen that is available for the plants is highly relevant. Some species are favoured when nitrate constitutes part of the uptake of nitrogen whereas other grow as well when most of the nitrogen is taken up as ammonium (organic nitrogen may also be of importance but is not discussed here). We have shown this dependency in our FNIS-index (functional nitrogen index for species) that is based on a function of species abundance related to a negative term of ammonium and a positive term of nitrate availability in a site (Diekmann & Falkengren-Grerup 1998). A species that grows on soils with a high mineralisation of ammonium and no nitrate will thus get a low index-value whereas a species that grows on highly nitrifying soils will get a high index-value. As the FNIS-index is significantly correlated with the Ellenberg N-values we show that the interpretation of increased Ellenberg N-values of the vegetation should be interpreted as an increased availability of nitrogen but rather that a substantial part of the available nitrogen is in the form of nitrate.

We have found that the degree of the mineralised nitrogen that has been nitrified (the nitrification ratio) is a good measure to estimate plant responses to nitrogen deposition

(Diekmann & Falkengren-Grerup 2002). As stressed before, it is most important to relate species occurrence to soil pH and nitrogen mineralisation simultaneously as most species have a clear relation to both. Out of species that tolerate acid soils we find that two ferns (*Dryopteris carthusiana*, *Athyrium filix-femina*) and *Rubus idaeus* have a higher observed than expected nitrification ratio and thus seem to be favoured by high deposition levels whereas the *Vaccinium* species seem to be disfavoured (Figure 5-35). A well-known species that grows on less acid soils and seems to be favoured is *Urtica dioica*.

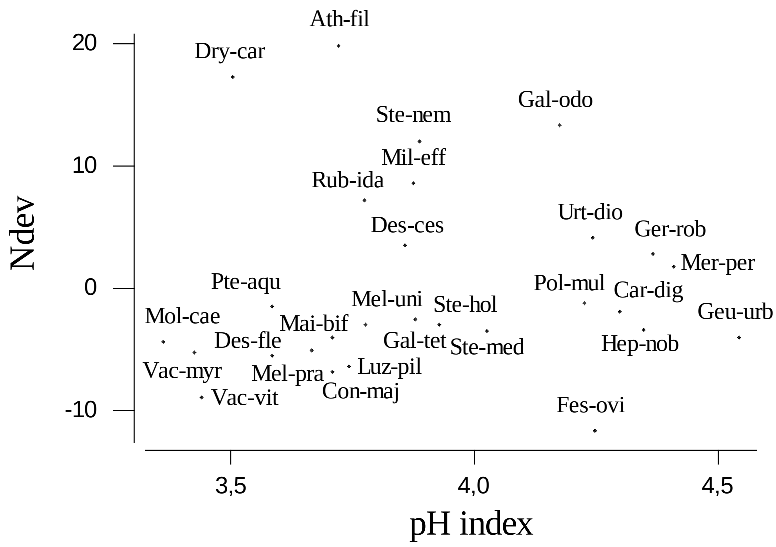


Figure 5-35 Difference (N_{dev}) between observed and expected values of the nitrification ratio (values between 0 and 100) for forest vascular plants in Skåne plotted against their corresponding pH values. Species above zero have a higher observed than expected nitrification ratio, those below a lower observed than expected value. See Diekmann & Falkengren-Grerup 2002 for explanations of abbreviations.

When nitrogen increases in the soil, and especially in highly nitrifying soils, the risk for nitrate leaching is enhanced. The microbes are active already early in the spring when the trees are still unleafed and the only source for plant uptake is the understorey. We studied three oak forests in Skåne and demonstrated that the nitrate production was high in the spring compared with the rest of the vegetative period (early and late summer). The nitrate leaching was three times higher in the spring than in the summer in spite of the high uptake by the understorey in both absolute and relative numbers. Out of the uptake of nitrate by trees and herbs, ninety percent was taken up by the understorey in the spring and as large numbers as 30-40% during the summer. The understorey vegetation is therefore important to prevent nitrate leaching during the spring and during the summer in nitrate-rich soils (Olsson & Falkengren-Grerup subm.).

We have also addressed the question of why there is a positive relationship between nitrogen deposition and potential net nitrogen mineralisation and nitrification in oak forest soils in south Sweden. A comparison of soils from regions exposed to 17 and 10 kg N ha^{-1} y^{-1} demonstrates that the soil microbes are more active in the nitrogen enriched soils and that they are not limited by carbon (Månsson & Falkengren-Grerup, subm.). The C:N ratio of oak litter and fresh leaves of *Deschampsia flexuosa* was also lower in the more nitrogen exposed sites which indicates an increase in litter quality, which in turn may result in higher carbon and nitrogen mineralisation rates in the more exposed soils. Thus, the increased microbial activity seems to increase net nitrogen mineralisation that allows nitrifiers to adapt to acid soils. Our results show that oak forest soils respond differently than coniferous forest soils, which often show a decreased respiration in response to nitrogen additions.

5.4.2 Tests with models for nitrogen effects on biodiversity

5.4.2.1 INTRODUCTION

This work describes an initial attempt to integrate feedback mechanisms and cause/consequence relationships between soil acidification, nitrogen eutrophication and climate variation over the territory on ground vegetation occurrence, through further development and integration of existing mathematical models. For this purpose the model concept VEG has been developed.

5.4.2.2 PARAMETERISATION

Parameterisation for six plant classes was determined by a numerical adaptation of Ellenbergs indices, combined with some additional information from the literature and model back-calculation of nitrogen conditions at such sites. The parameterisation of the plant classes on acidity was taken from response function determined by Sverdrup and Warfvinge (1993). So far only acidity and nitrogen responses have been estimated. The effect of soil water was all set to unity, no temperature effect was considered at this point. The competition functions (roots for nutrients, canopy for light) were also set to unity at this stage. The response functions of 6 plant classes, represented by names for species which are typically included in the class, for nitrogen concentrations in soil water are presented in Figure 5-36. The figures have been derived from Ellenberg indices through a back calculation of the nitrogen concentration at several synthetically generated sites. The response diagrams are at this point preliminary and employed for experimental purposes. Final parameterisation will eventually have to be supported by further use of existing experimental data and available Swedish regional surveys.

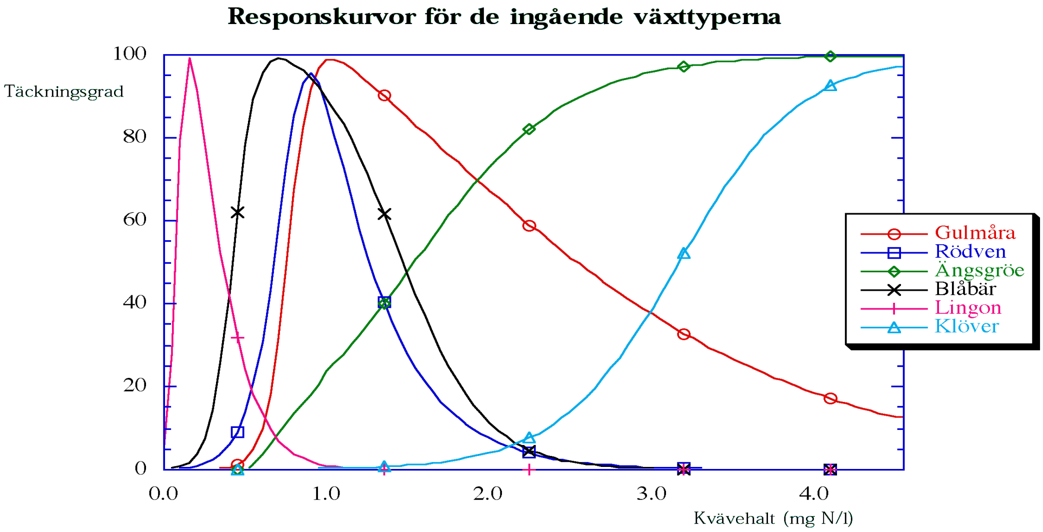


Figure 5-36 The response functions adopted for the six plant classes used in this work.

5.4.2.3 RESULTS

The experimental model was applied to a hypothetical site with synthetic site data and a research site at Fårahall, at Hallandsåsen in southern Sweden. The purpose was to test the behaviour and dynamics of the preliminary model before further development is undertaken. In Figure 5-37 the calculated response of the blueberry plant class is shown with and without competition from six other plant classes in the same plot.

At present, competition for nutrients was indirectly incorporated into the nitrogen response functions. Aboveground competition was set to unity for all classes, but later this will be elaborated into three different basic strategies.

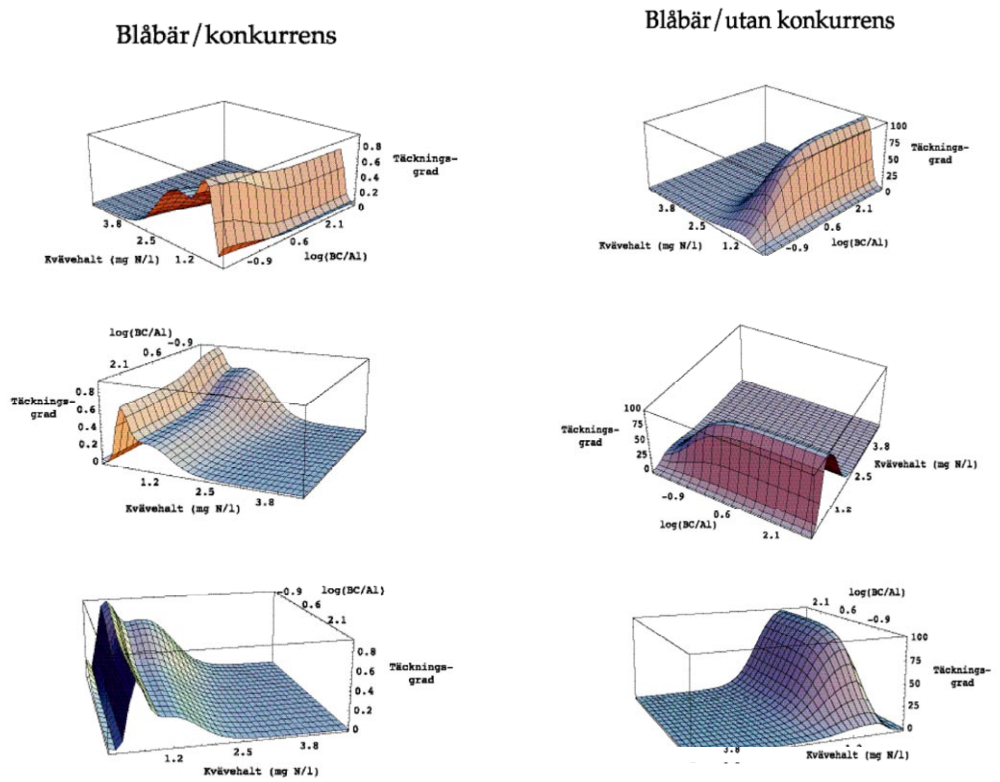


Figure 5-37 The response of the blueberry plant class with and without competition from other 6 plant classes in the same plot, assuming differentiated below-ground competition, but equal terms aboveground competition.

The presence of competition is important and also shows that field observations need to be subject to stratified filtering of the effects to come down to the pure acidification response or the pure nitrogen response. The field response is the final product of

1. Acidity
2. Nitrogen
3. Temperature
4. Water
5. Root competition
6. Aboveground competition for light

As these vary independently between sites, field response curves must be descrambled to these response functions in order to be applicable to other points in the region. This descrambling of field data is mathematically not always solvable, and thus field observation of response is much more valuable for testing the models built bottom up from individual response functions.

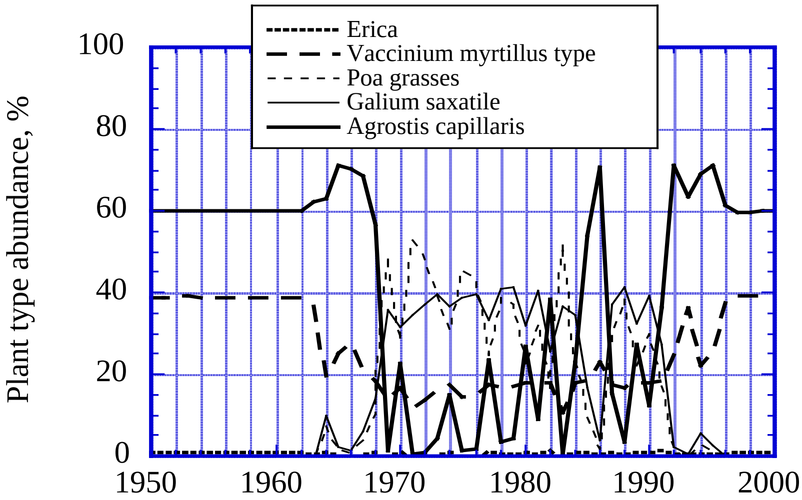


Figure 5-38 The development of species abundance at Fårahall research site as a function of increased acidification from 1960 and significant nitrogen inputs 1970 with a gradual reduction according to the multiprotocol after 1990.

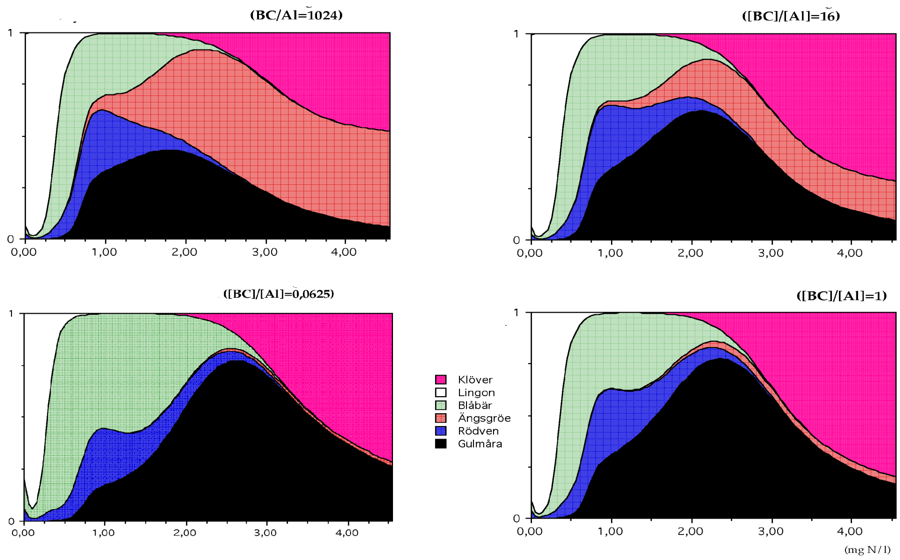


Figure 5-39 The figures show the response for six vegetation classes which we gave the preliminary names “lingonberry heather” (white), “blueberry“ (green), “common bent” (blue), “heath bedstraw” (black), “clover” (magenta) and “ryegrass” (red).

In Figure 5-38 the development of species abundance with time at Fårahall research site is shown as a function of increased acidification from 1960 and significant nitrogen inputs 1970 with a gradual reduction according to the multiprotocol after 1990. Plant response was in this case assumed to be instant, but the figure shows what kind of response and output the final model may give.

In Figure 5-39 we have used the model to study different responses as a function of soil acidity, nitrogen and mutual competition between the plants. It can be seen that the specific response of a plant class to increase nitrogen will be different if the soil acidity is changed.

This reflects that in the multi-dimensional response space, soil acidity, temperature and soil moisture may each or in combination significantly change the field response of a plant class to change in nitrogen input. It explains why it is notoriously difficult to get clear results derived from simple field experiments or regional surveys along correlated gradients. In the second phase, ASTA field data and models will therefore be used together to derive better critical loads with a closer connection to ecological effects than earlier estimates.

Our model which is being developed within ASTA will be the tool used to sort out these feedbacks and to predict responses under different, changing conditions that may occur during initial acidification and subsequent recovery from acidification in a changing climate.

5.5 Interactions between acidifying air pollutants and land use

In the ASTA programme tools are developed to optimise national abatement strategies for nitrogen and acidifying components with the focus on areas where transboundary air pollution and forestry and other land-use practices contribute to environmental effects. The work is concentrated to two topics: Nitrogen budgets in forest soils; and acidity and base cation budgets in forest soils. Important results from the preliminary mass balance calculations and dynamic modelling include reports on:

- Regional (South Sweden) calculations of the present leaching of nitrogen from managed forests. The leaching from clearcuts is calculated as a function of nitrogen deposition, and the contribution can be substantial in areas with high deposition.
- Regional (South Sweden) calculations of historic changes during 50 years of total nitrogen content in forest soils and a prediction of future development in the coming 50 years with a scenario including reduced deposition and increased intensity in forestry (whole tree harvesting). The prediction indicates that the present accumulation of nitrogen in forest soil can be decreased in the future, and some areas will show a net loss of nitrogen.
- Model calculations (MAGIC) of the long-term impact of different harvest intensity, and compensatory fertilisation, on recovery from acidification. The case study indicates the importance of forest management for the recovery, and the methods used will be a basis for regional calculations in the South part of Sweden.
- The land use in Sweden has been mapped, together with the Mistra programme RESE. The mapping is a basis for regional integrated assessment of air pollutants and land use. The database will be complemented by data on deposition, soil, surface water, hydrology etc. necessary for biogeochemical model calculations.

The studies concerning nitrogen are initially focused on future accumulation and leaching of nitrogen from managed forest soils. The calculated accumulation of N in forest soils during the last 50 years in South Sweden is shown in Figure 5-40. The results so far implies that the future accumulation of nitrogen in soils in Sweden will decrease (depending on harvest intensity) and the main contribution to leaching, caused by forest management, will come from clearcuts as today.

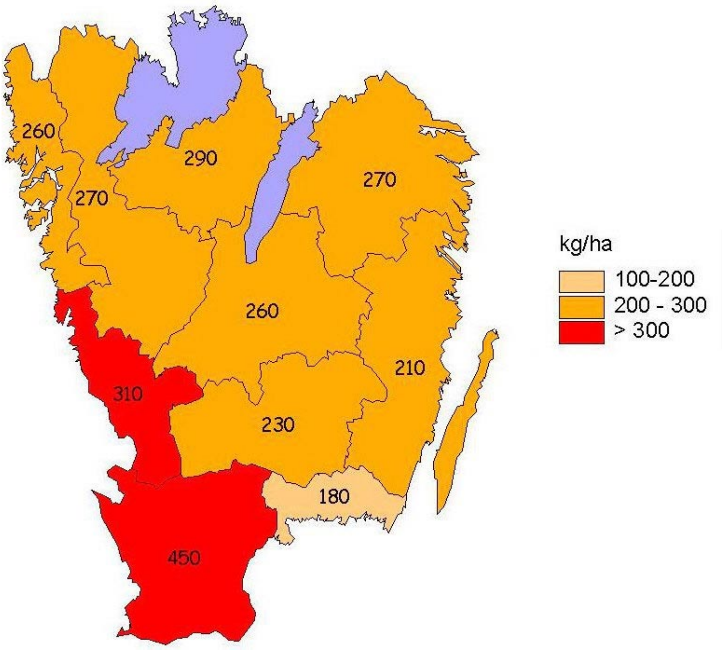


Figure 5-40 Calculated accumulation of nitrogen in forest soil in South Sweden during the period 1950 to 2000.

In Figure 5-41, the calculated present leaching of nitrogen from clearcuts is shown as a function of N deposition. The results also indicate that nitrogen saturation in soils will not be the critical effect of nitrogen deposition in productive forests in Sweden in the future, if whole tree harvest is applied (Figure 5-42).

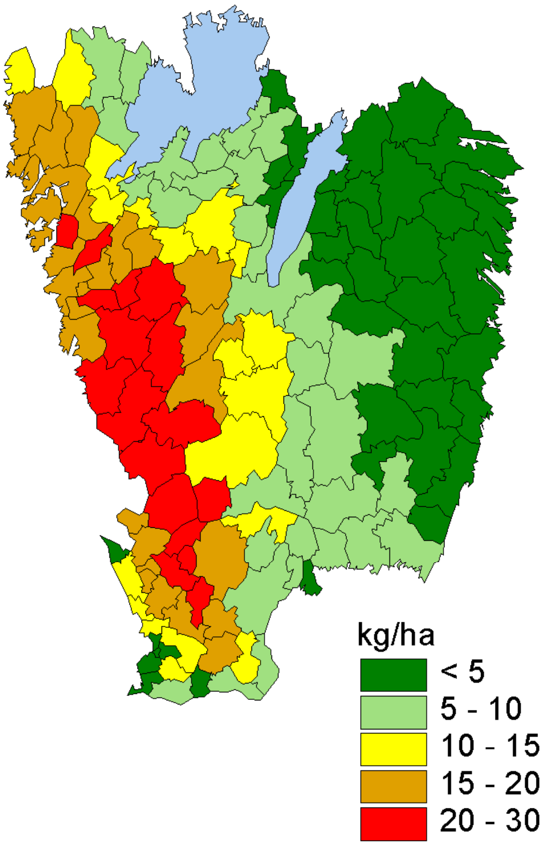


Figure 5-41 Model calculation of the present leaching of inorganic nitrogen from clearcut areas. Leaching per year during an average clearcut phase of five years.

The calculated and predicted deposition of N in South Sweden (Götaland), or input, during two 50-year periods shown in Figure 5-42 is rather similar. The same yearly deposition during 2000 to 2050 and no further reductions after 2010 can be regarded as a worst case. The historic output of N during 50 years (harvest and leaching) from forest was much lower than the input (Figure 5-42). The average accumulation in soils was half of the deposition

The calculated future output of N by harvest is much higher than the last 50-year period (Figure 5-42), resulting in an average net loss of nitrogen from forest soils in South Sweden. Leaching of N will not increase during the coming 50 years according to the calculations, and this is a consequence of the absence of further soil accumulation. The increased harvest of N during the coming 50 years is caused by both increased growth, estimated to 30% mainly due to improved management methods, and intensive harvest of tree-sections with high concentrations of N (e.g. tops, branches and needles).

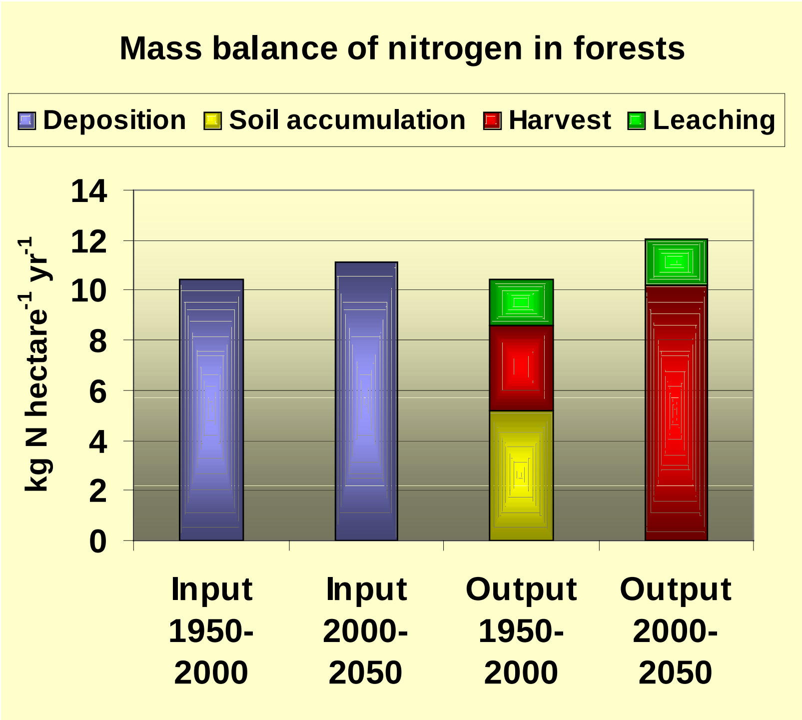


Figure 5-42. Mass balance of nitrogen in forests in South Sweden during two 50-year periods. The small net effects of nitrogen fixation and denitrification are neglected in the calculations. The historic harvest is based on data from the National Forest Inventory in Sweden. Future harvest is a scenario with intense forestry and extraction of forest fuels.

The studies concerning acidification and recovery are focused on the correlation between acidification in soil and surface water and the different acidification processes connected to deposition of strong acids compared to growth and harvest of the forest. The work so far comprises analyses of model tools for describing recovery from acidification and the influence of forest growth, harvest and fertilisation. A case study in the South part of Sweden indicates the importance of the intensity of harvest, and compensatory fertilisation, for the recovery process in soil and surface water.

In Figure 5-43, an example of dynamic modelling of recovery from acidification after reduction of the sulphur deposition is shown for a spruce forest with three different scenarios of forest management. The modelling indicates that the future ANC levels in run off is dependent of the removal of base cations by harvest. Full compensations via fertilisation of base cations will allow a more complete recovery, in comparison to harvest without compensation.

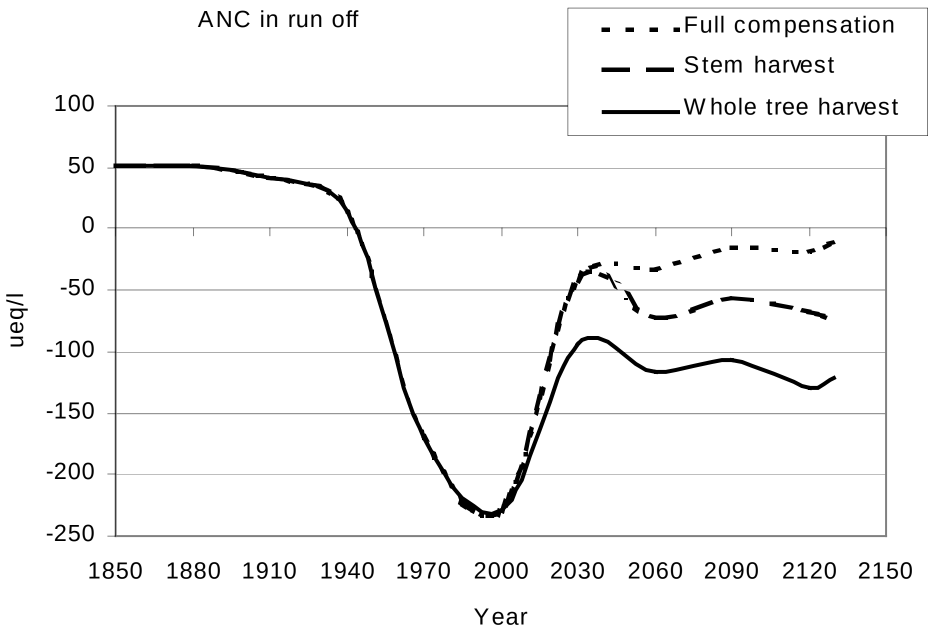


Figure 5-43 Historic and future ANC in run off from acid forest soil with different intensity in forest management. A case study with model calculations (MAGIC) in a spruce forest in South Sweden.

The application of dynamic modelling (MAGIC and SAFE) on managed forests on acid soils is a basis for development of indicators of recovery from acidification and sustainable forest production. The indicators will be used for regional assessment of the future need of decreased deposition and special management methods including liming and fertilisation of soils.

5.6 Development of new concept for modelling effects of ozone on crops and forests.

The key results from the work within ASTA with ozone effects on plants are the steps taken from concentration based exposure response-relationships to ozone uptake based relationships.

5.6.1 Crops

Experimental data has been used to evaluate the correlations between relative yields of crops and the cumulative uptake of ozone to the leaves ozone uptake (CUO). In Figure 5-44 the relative yields of potato and wheat are presented as functions of both ozone uptake-based relationships and AOT40. Better correlations were obtained both for wheat and potato when using uptake based relationships.

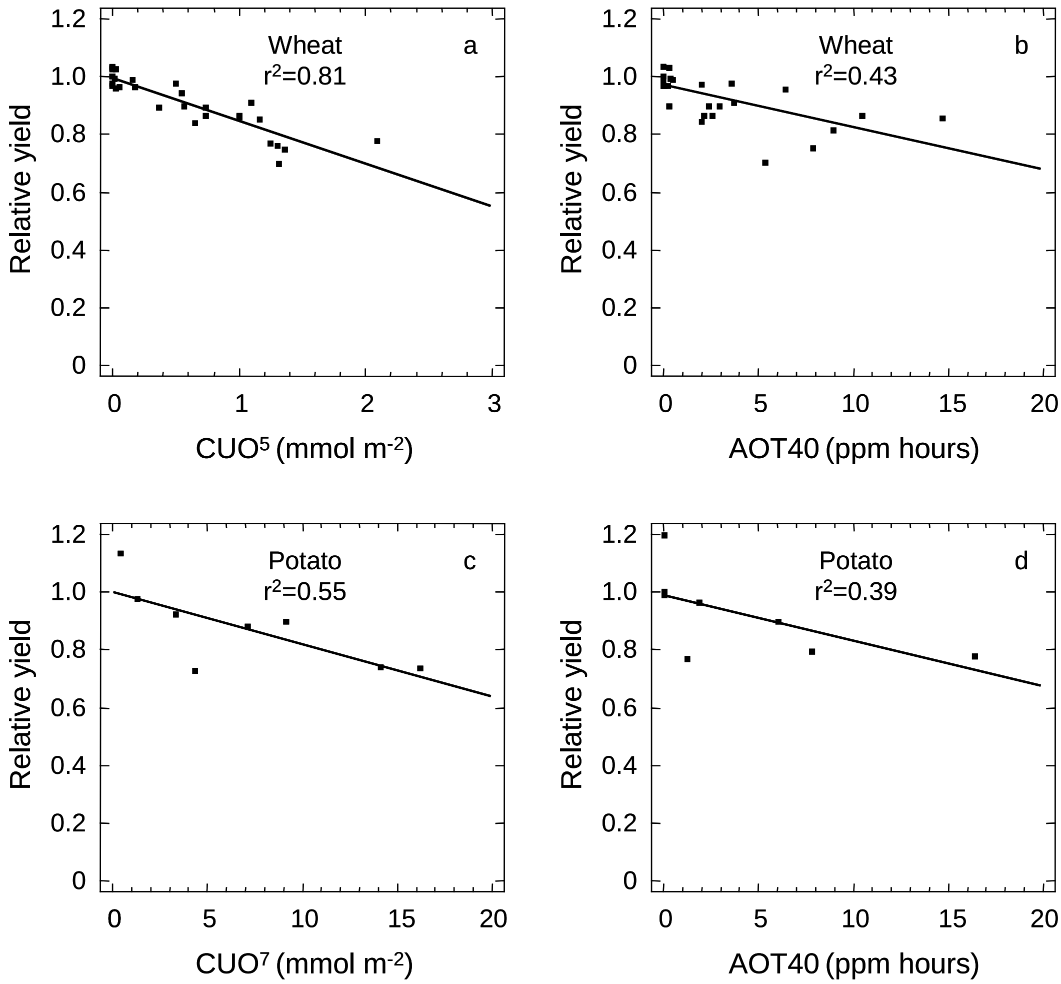


Figure 5-44 Relative yield in wheat (a, b) and potato (c, d) in relation to an uptake- (CUO) and a concentration based (AOT40) exposure index, respectively. Wheat data from five years experiments at Östad säteri (Danielsson et al, submitted). Potato data from two years of experimentation in Sweden, Finland, Belgium and Germany (Pleijel et al., 2002).

It has been suggested within the ICP-Vegetation under CLRTAP to divide crops into three categories based on their sensitivity to ozone. ICP-Vegetation has undertaken an extensive review on this. The categories are: 1. very sensitive, 2. moderately sensitive and 3. insensitive. Wheat is the most well studied example of category 1 and potato of category 2. Barley is an example (see e.g. Pleijel et al 1992) of category 3. Crops belonging to this last category will probably be excluded from mapping etc, since the ozone effects are so small.

This development has been based on extensive efforts in conductance modelling. For more information, see section 4.2.1.

5.6.2 Forest trees

5.6.2.1 DOSE-RESPONSE RELATIONSHIPS BASED ON AOT40

Dose-response relationships for ozone impact on Norway spruce (Picea abies) and European silver birch (Betula pendula) have been derived within the ASTA project, based on experimental results from Östad. As a first step AOT40 – response relationships have been constructed (Figure 5-45, A and C).

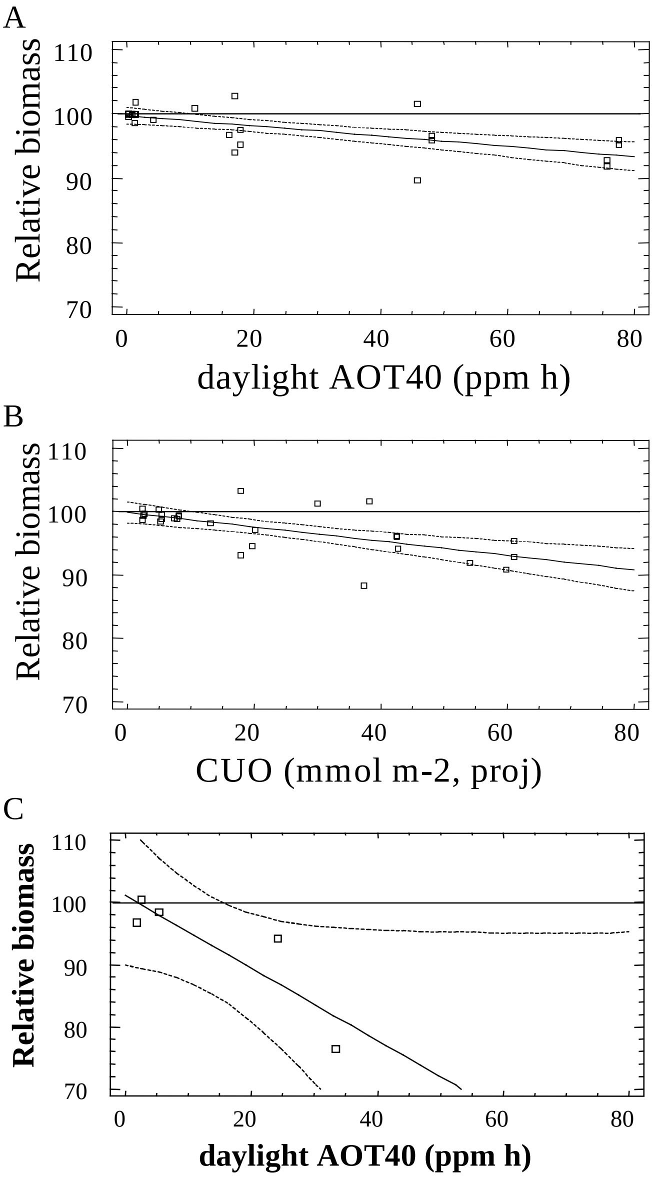


Figure 5-45 Impact of ozone on the total biomass of young Norway spruce trees in relation to daylight AOT40 (A, Skärby et al., manuscript) and in relation to cumulative ozone uptake (B, Karlsson et al. 2002b) and on the total biomass of young Silver birch trees in relation to daylight AOT40 (C, Karlsson et al., 2002d). The young Norway spruce trees were exposed in open-top chambers during four growing seasons to different ozone concentrations in combination with drought stress and in combination with phosphorous deficiency. The young Silver birch trees were exposed to different ozone concentrations in open-top chambers during two growing seasons at optimum water and nutrient availability.

The dose-response relationship, using AOT40 as the dose index and % biomass reduction as the response, was by far steeper for birch, in comparison to spruce. There are two possible explanations for these results. One is that the stomatal conductance and thus ozone uptake by unit leaf area is higher for birch compared to spruce (see further discussion below). Another explanation is that birch is a faster growing species than spruce, i.e. it has a higher relative growth rate (RGR). If it is assumed that ozone affects the RGR, then a similar % reduction in RGR will results in a higher value of % biomass reduction for the fast growing birch in comparison to the slower growing spruce. In fact the reduction in RGR per 10 ppm h AOT40 was estimated to –2% for birch and -0.75% for spruce.

5.6.2.2 DEVELOPMENT OF STOMATAL CONDUCTANCE AND OZONE UPTAKE SIMULATION MODELS

A stomatal conductance simulation model has been developed for young, well-watered and drought stressed Norway spruce trees in open-top chambers (Karlsson et al. 2000). The model is based on a multiplicative concept described in Emberson et al. (2000). Based on the simulated stomatal conductance and measured ozone concentrations, the cumulative ozone uptake (CUO) to the needles was estimated for periods July - September during three years (Figure 5-46). This was then compared to the daylight AOT40 during the same periods.

There was a substantial difference between the different periods in the relative magnitudes of the CUO and the AOT40 (Figure 5-46). The difference in the AOT40 index between 1993 and 1994 was much larger, compared to the difference in the CUO.

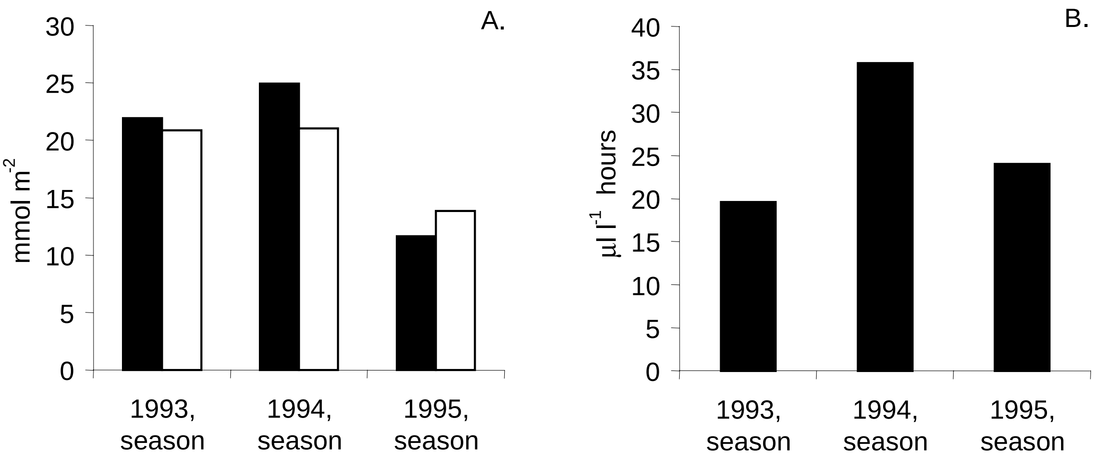


Figure 5-46 The estimated cumulative ozone uptake during July - September 1993, 1994 and 1995, compared to the daylight AOT40 during the same periods. A; Cumulative ozone uptake, B.; daylight AOT40. In A filled bars indicate well-watered saplings and open bars drought stress treated saplings. From Karlsson et al. 2000.

During 1993, the ozone concentrations were relatively low but the weather conditions favourable for ozone uptake, while the opposite was true for 1994. The drought stress caused partial stomatal closing and therefore substantially reduced the CUO.

A stomatal conductance and ozone uptake simulation for birch is presently under development and will be used to estimate the CUO during the two-year open-top chamber experiment at Östad.

5.6.2.3 OZONE UPTAKE - RESPONSE RELATIONSHIPS FOR YOUNG BIRCH AND NORWAY SPRUCE

An ozone uptake – response relationship has been developed for Norway spruce (Figure 5-45B) and is under way for birch.

Our results for young Norway spruce showed a 3% reduction of the total plant biomass per 10 mmol m^{-2} CUO on a total needle area basis. This was similar to results obtained in studies of mature Norway spruce trees in Austria (Wieser 1997), where an approximately 7% reduction per 10 mmol m^{-2}, was found for the ozone impact on the photosynthetic capacity.

The cumulated ozone uptake to needles of Norway spruce over the growing season under field conditions is not yet known. However, a preliminary study by Emberson et al. (2000) estimated the ozone uptake to beech leaves in Sweden during one growing season to 6.5 - 7.0 mmol m^{-2}, on a total leaf area basis.

5.6.2.4 SCALING OZONE - RESPONSE RELATIONSHIPS FROM JUVENILE TO MATURE TREES

An important aspect when comparing the ozone impact on juvenile and mature trees is the rate of ozone uptake (Samuelson and Kelly, 2001). In order to assess these aspects, two projects have been started with the aim to estimate ozone uptake to mature Norway spruce and birch trees.

Preliminary results are available from the project, where a sky-lift was used to enable access to the crowns of mature European silver birch trees around Asa Experimental Park in Småland, south Sweden. Climate parameters and ozone concentrations are measured at the nearby meteorological station at Asa and soil water availability is measured close to each experimental tree. The leaf stomatal conductance is measured at regular intervals using a gas exchange system.

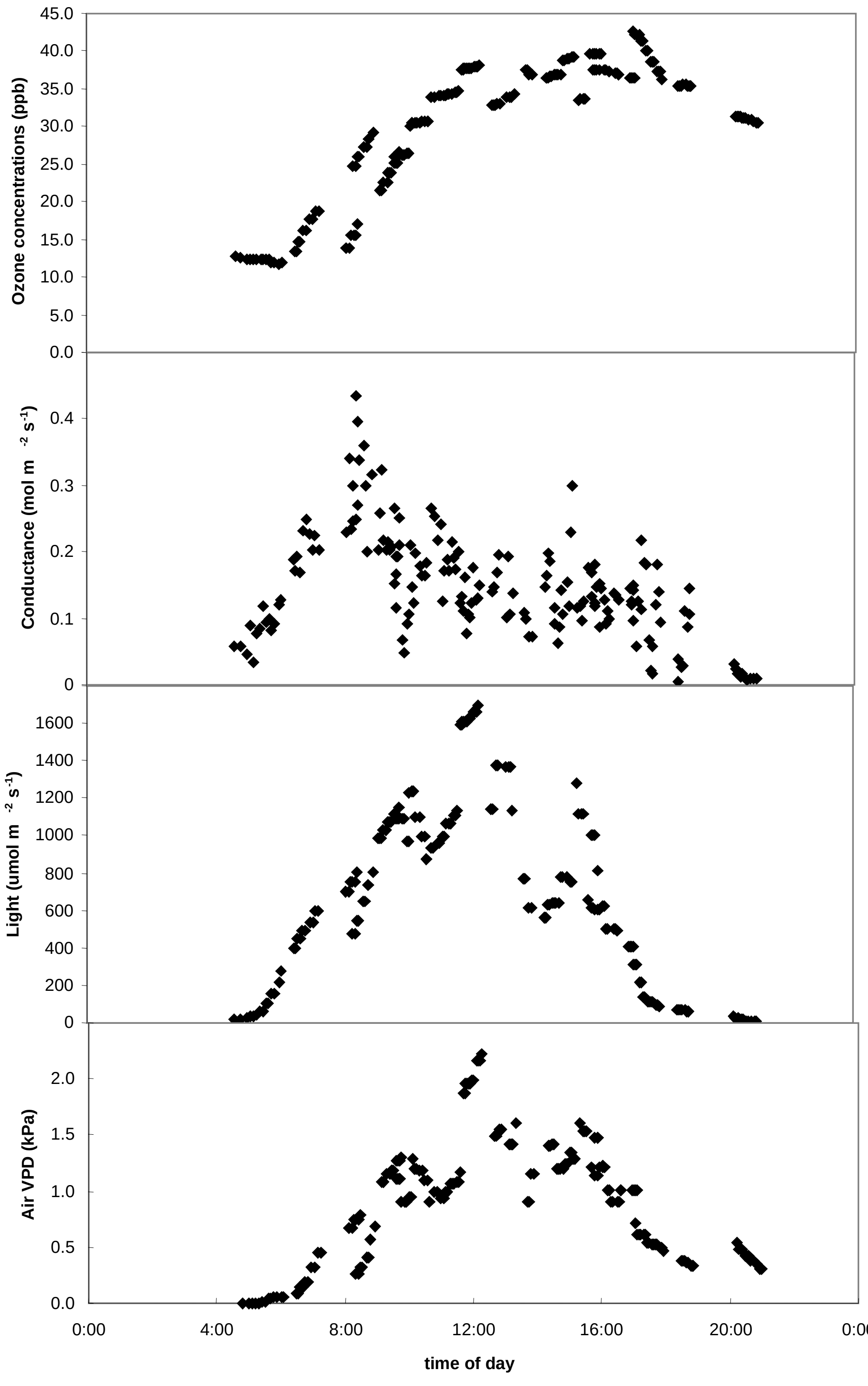


Figure 5-47 Preliminary results from stomatal conductance measurements on leaves from the upper canopy of mature birch trees during six days in August 2001.

Results from measurements during 6 days in August 2002 are shown in Figure 5-47. Ozone concentrations follow the general pattern for inland forest landscapes in south Sweden, with low night-time concentrations. Ozone concentrations increase in the morning in parallel with the light and reach a maximum in the afternoon. Leaf conductance in the upper canopy is generally low during night time. It then increases rapidly in the early morning. However, it reaches a maximum before noon and then decreases, mainly due to the increasing air water Vapour Pressure Deficit (VPD). Thus, the diurnal course of the ozone uptake to the leaves of these mature birch trees is very different from the diurnal pattern of the ozone concentrations. The maximum leaf conductance of the mature birch trees was approximately 0.4 mol m^{-2} s^{-1}, projected leaf area, which is about 2/3 of the maximum leaf conductance found for young, well fertilised birch trees. Thus, there was not a substantial difference in maximum conductance between young and mature birch trees.

Results from measurements of ozone uptake to mature Norway spruce trees will be available during 2002.

5.6.2.5 VALIDATING OZONE IMPACT ON ADULT NORWAY SPRUCE TREES

Stem circumferences have been measured at approximately weekly intervals during the growing season since 1993, with dendrometer bands on five Norway spruce trees on each of 10 different plots within 3 km from the Asa Experimental Forest in Småland, south Sweden. The relative yearly increments in stem basal area were correlated to measurements of soil humidity at the plots and weather parameters measured at a nearby weather station at Asa experimental forest. The independent parameters tested are shown in **Table 5.2**.

A multiple linear regression analysis demonstrated highly significant negative impacts of average daylight ozone concentrations (ozdayavg), as well as daylight AOT40 (AOT40day), on the yearly basal area increment of mature Norway spruce trees (Table 5.3 and Table 5.4, Karlsson et al, 2002c). Stem size, soil water potential (soilWPav), and global radiation (Glob24av) also showed highly significant impacts on the yearly stem growth. The stem growth varied considerably between individual trees within the plots. None of the parameters measured in this study could account for this variation between individual trees. This explains the relatively low correlation coefficients for the total statistical models. The magnitude of the effects was complicated by autocorrelation (colinearity) between the ozone indices and several environmental parameters and thus remains to be established. However, this statistical analysis of the correlations between yearly stem growth and different soil moisture and weather parameters provides strong evidence for an ozone impact on the growth of adult Norway spruce trees.

Table 5.2 List of independent parameters tested in the multiple linear regression analysis of yearly relative stem growth. CV, coefficient of variation(s.d. / average *100).

parameter	unit	average	s.d	CV, %	max	min	interpretation
Dependent variables							
stgroabs	(mm^2) $year^{-1}$	779.7	402.6	51.6	2271.4	90.2	absolute basal stem area increment during the growing season
stgrorel	0/00 $year^{-1}$	49.1	36.8	74.9	204.4	8.0	relative basal stem area increment during the growing season
Independent variables							
Plot	1 - 10	-	-	-	-	-	Plot
Tree	1 - 5	-	-	-	-	-	Tree number within the plot
year	1993 - 1999	-	-	-	-	-	year of the measurements
stemsize	mm^2	19249	9329	48.5	50675	4057	stem basal area at the beginning of the yearly measuring period
ozdayavg	ppb	32.8	2.96	9.0	37.2	27.4	average daylight ozone concentration during the growing season
AOT40day	ppb h	4978	2042	41.0	8660	1521	daylight accumulated ozone exposure over a threshold 40 ppb, for the yearly measuring period
soilWP10	MPa	-0.164	0.180	109.7	-0.028	-0.687	24h average soil water potential at 10 cm depth during the growing season
soilWP40	MPa	-0.142	0.138	97.2	-0.027	-0.576	24h average soil water potential at 40 cm depth during the growing season
soilWPav	MPa	-0.153	0.150	-98.0	-0.027	-0.565	soil water potential, 24h average for 10 and 40 cm depth, during the growing season
temp24av	C	13.16	0.92	7.0	14.49	11.90	24h average air temperature during the growing season
VPD24av	mbar	1.80	0.49	27.2	2.50	1.19	24h average air water vapour pressure deficit during the growing season(a function of air temperature and relative humidity)
glob24av	W m^{-2}	173.1	14.5	8.4	191.9	144.9	24h average global radiation during the growing season
prec24av	mm h^{-1}	0.0999	0.0085	8.5	0.1157	0.0901	24h average precipitation during the growing season

Table 5.3. The results from fitting a multiple linear regression model to describe the relationship between the relative yearly stem basal area increment and several independent parameters. The ozone index was average daylight ozone concentration (ozdayavg). The p-value for the ANOVA of the model was <0.0000. The model explained 31.2% of the variability in relative stem growth (stgrorel).

parameter	parameter estimate	Std error	t-value	p-value
intercept	25.17	48.12	0.52	0.60
stemsize	-0.001616	0.000181	-8.92	0.0000
soilWPav	89.62	17.08	5.25	0.0000
ozdayavg	-6.610	1.256	-5.26	0.0000
Glob24av	2.074	0.313	6.617	0.0000
Prec24av	-614.9	261.7	-2.35	0.019
temp24av	0.17	3.94	0.043	0.97
VPD24av	-8,224	9.457	-0.870	0.385

Table 5.4 The results from fitting a multiple linear regression model to describe the relationship between the relative yearly stem basal area increment (stgrorel) and several independent parameters. The ozone index was daylight AOT40 (AOT40day). The p-value for the ANOVA of the model was <0.0000. The model explained 30.7% of the variability in relative stem growth (stgrorel).

parameter	parameter estimate	Std error	t-value	p-value
intercept	-210.6	69.1	-3.05	0.0025
stemsize	-0.001610	0.000181	-8.856	0.0000
soilWPav	92.83	17.83	5.21	0.0000
AOT40day	-0.01067	0.00212	-5.029	0.0000
Glob24av	2.412	0.375	6.43	0.0000
Prec24av	-465.2	258.3	-1.801	0.073
temp24av	-0.0271	4.157	-0.0065	0.99
VPD24av	-14.43	9.61	-1.50	0.134

5.6.2.6 Uncertainties and consideration of climate factors for ozone effects on plants

The most profound consequence of the work with ozone effects on crops/plants in ASTA is the strong dependence of ozone effects on ozone uptake, which, in turn, is driven both by ozone concentrations and climatic variables that determine stomatal conductance. A certain set of concentrations will then have much larger effects in cool and humid climate than in dry and hot climates – as suggested already by Emberson et al (1999). Relatively soon we expect to be able to quantify effects in the field and to incorporate the new response relationships with models like RAINS.

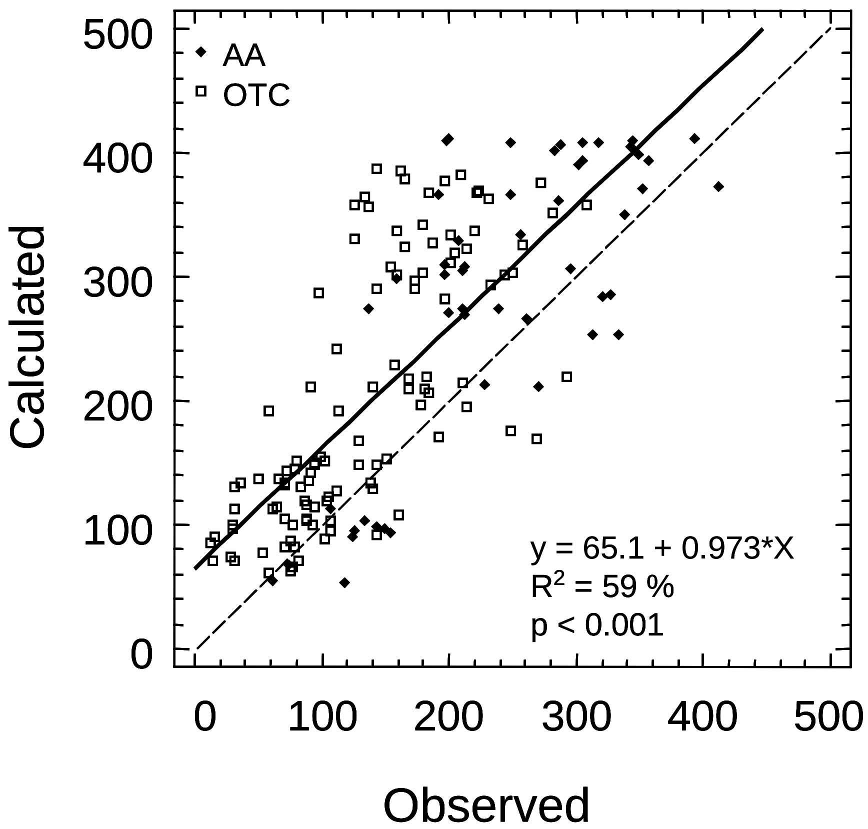


Figure 5-48 Stomatal conductance in wheat. Values calculated using a multiplicative model sensitive to temperature, solar radiation, vapour pressure deficit, ozone exposure, phenology (time) and carbon dioxide are plotted against observed conductance. Broken line: theoretical 1:1-relationship. Solid line: regression.

In Figure 5-48 the relationship between modelled and observed conductance in wheat is shown. The multiplicative model concept (environmental variables act multiplicatively to set the stomatal conductance) put forward by Emberson (1999), based on earlier work by Jarvis (1976) and Körner (1994), was calibrated for our data on conductance for wheat. A relatively strong relationship was obtained, although it does not overlap entirely with the 1:1-line. This should be compared by the inherent assumption of an AOT40 relationship that the line in the relationship is horizontal at an arbitrary level, i.e. it completely ignores the actual conductance during daylight hours. A similar comparison with a similar performance was obtained for potato (Pleijel et al., 2002).

5.7 Particles and human health

5.7.1 Concentrations and sources of PM2.5 and PM10

A monitoring network for PM2.5 and PM10 was set up from Malmö (ca 55°N) in the very south of Sweden to Lycksele (ca 65°N) in the north. The network includes two background sites in rural areas, roof and kerb stations in the three major cities of Sweden; one mid-size city (Umeå) in the north and two minor cities, one in the south (Växjö) and one in the north (Lycksele) (see also Areskoug et al., 2002).

The measured annual mean concentrations are surprisingly similar at all sites except at the most polluted stations. When examining the reasons it was found that long distant transport of pollutants from the continent has a large influence in the south which decreases towards the north. In addition, local sources become more important in the far north due to more frequent meteorological inversions during the winter.

The data was classified and evaluated according to type of sampling site. In urban background sites, such as parks, rooftop or suburban areas, local sources make quite limited contributions to PM10, about 35% during the hours when most influence is expected. For PM2.5, hardly any increase is detected at these sites in comparison to the rural sites. Street side locations show an overwhelming increase in PM10 of a factor of 4 at peak rush hours, while the PM2.5 only increases at the most with 50% during the same time period (see Figure 5-49)

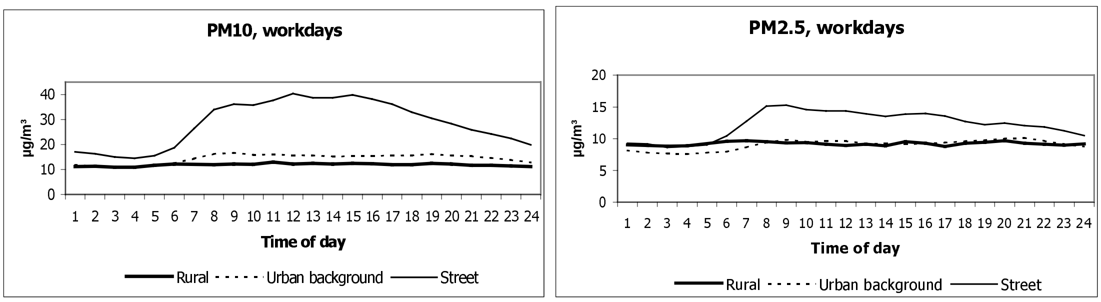


Figure 5-49 Daily variation of PM2.5 and PM10 concentrations, measured during one year (2000) at 12 sampling sites in Sweden.

Long range transported aerosol thus has quite a dominating influence on PM2.5 levels in Sweden. For PM10 coarse particles from resuspended road dust is an important additional source. Surprisingly, significant contributions to PM2.5 can also be attributed to resuspended road dust. This is very nicely shown in Figure 5-50, were the volume size distribution going from the most polluted inner city to outside the city is shown. The variation is caused by the varying influence of local sources. The exhaust particles show up around 100 – 150 nm diameter and the resuspended particles around 1 to 10 microns. The long range transported fraction is seen in the size range 300 to 500 nm.

However a significant volume fraction is seen in the background data in the size range between 1 and 2.5 μm. In Swietlicki et al. (2002), it was shown that sea spray and soil dust together accounted for 41-45% (~5.5 μg/m^3) of the PM10 and 36-41% (~4 μg/m^3) of PM2.5, based on measurements at a background site in Southern Sweden. The sea spray is a natural contribution but it is questionable whether the resuspended dust is natural. From the results obtained in Stockholm, it is likely that the cities on the continent have large emissions of resuspended dust.

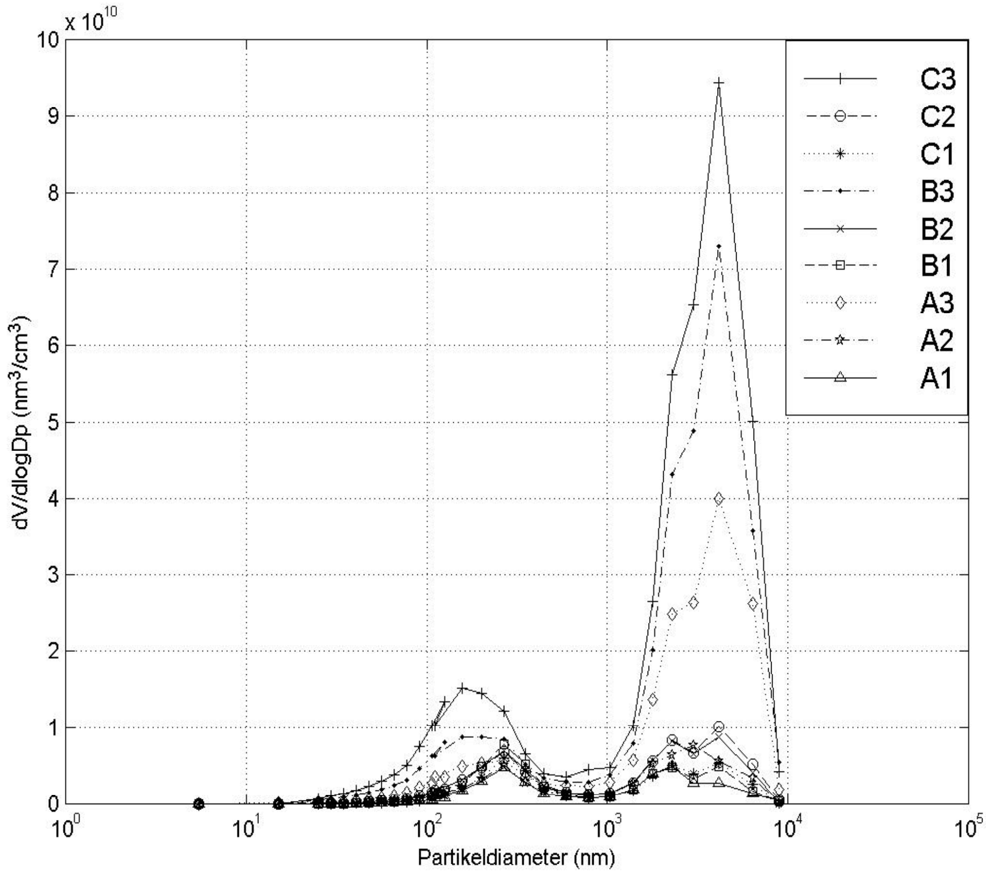


Figure 5-50 Particle volume in different urban environments measured in Stockholm. C = most polluted inner city sampling sites, A = least polluted sites outside city.

In southern Europe Saharan dust event is a common feature. Exceedance of limit value for PM10 due to dust events is excluded in the legislation. In Figure 5-51, two events in Spain during February and October 2001 are shown. The largest impact of a dust event is in PM10 levels but it is also traceable in PM1 and results in a significant increase in the PM2.5 concentrations.

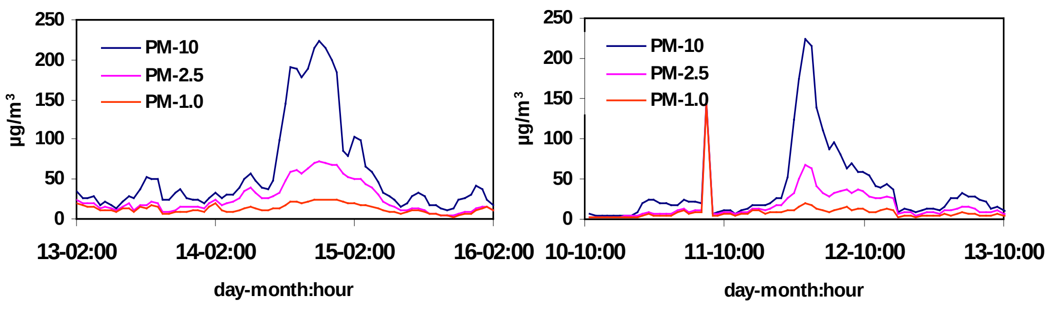


Figure 5-51 Time series of PM1, PM2.5 and PM10 during dust events in Huelva, Spain (Querol pers. comm.).

The research has so far shown that long range transport is the most important source of aerosols to even in the urban environment, but also that coarse particles, e.g. particles originating from resuspended road dust or desert dust range down in size to about 1 µm, thus giving a considerable contribution to PM2.5.

5.7.2 Other characteristics of the major aerosol types

Epidemeological studies usually show a significant correlation between observed health effects and particle mass concentrations. However, there is no scientific evidence that it is the particle mass in a certain size fraction that actually causes the observed health effects. To obtain a better understanding of which components in the aerosols that are actually harmful, a better chemical and physical characterisation is needed.

The particle number size distribution when moving from a highly trafficked street in central Stockholm to a rural site outside Stockholm shows that the number in most urban environments is totally dominated by local sources emitting particles in the size range below 200 nm (Figure 5-52)).

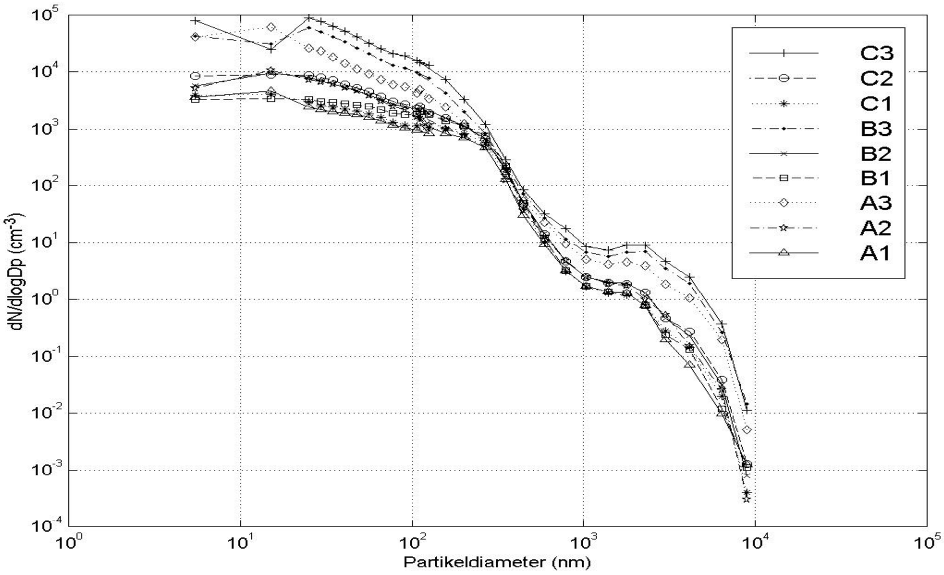


Figure 5-52 Particle number size distributions measured in Stockholm, 2000, in different environments ranging from highly trafficked street to rural suburban areas.

This implies that depending on the dominating source in an urban environment, the PM2.5 or PM10 does not necessarily represent the number of particles. This is confirmed by a comparison of a European set of measurement performed by J-P Putaud, JRC as shown in Figure 5-53 (Baltensperger et al., 2002).

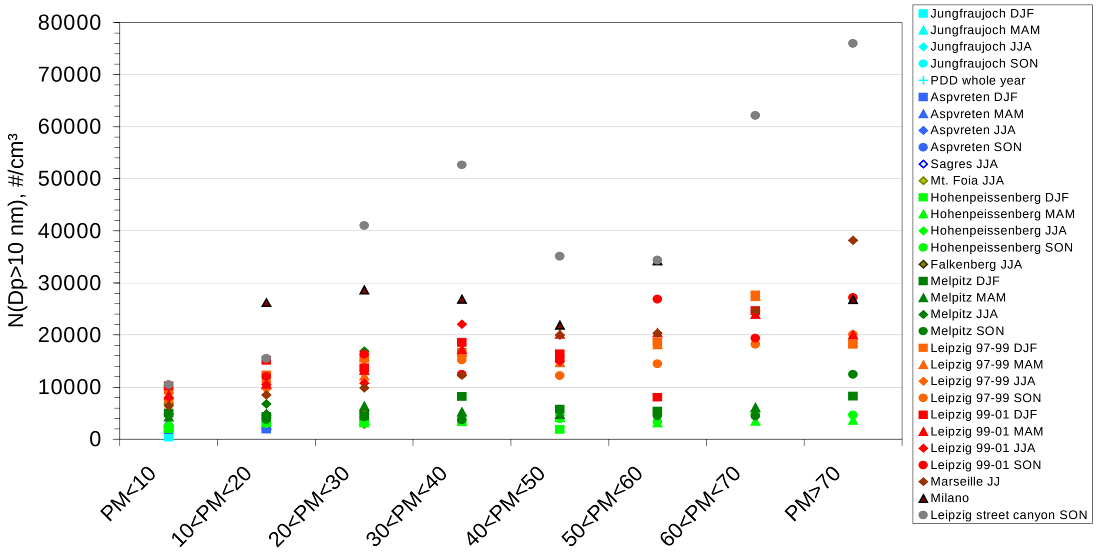


Figure 5-53 Measured total number of particles larger than 10 nm in a variety of European environments sorted after PM10 mass load (Baltensperger et al., 2002).

The chemistry is also largely dependent on the source composition. The chemical composition of a certain PM concentration can vary strongly due to different source influence as demonstrated by comparing measurements in Barcelona, Bologna and Gent (Baltensperger et al., 2002).

The ambient aerosol originates from a complex mixture of sources and thus becomes a chemical and physical heterogeneous mixture. The different sources also emit several totally different types of particles. A diesel vehicle emits particles in exhaust, brake debris, tire wear, road wear and resuspended road dust. The exhaust contains several aerosol types originating from engine wear, carbon from incompletely combusted fuel and particles from nucleating condensable organic gases. All these particle types have different chemical composition (Matter et al., 1999).

The nature of the different sources and varying influence of different sources gives a quite varying physical and chemical characteristic of the ambient aerosol. Consequently, control measures directed towards a certain particle mass fraction , will not necessarily limit other potentially toxic or harmful components such as number and surface concentrations or solubility and chemical components e.g. PAH.

5.7.3 Network of advanced background stations, particle super stations.

Advanced particle measurements at single background stations such as Hyytiälä, Finland and Mace Head, Ireland have provided new and valuable information on particle formation processes in the atmosphere. However a network of stations is needed to achieve quantitative descriptions of the rates of several important processes in the atmosphere, besides getting a general view of the absolute concentrations and their variability. A network has been established on the initiative of Institute of Applied Environmental Research (ITM) in close co-operation mainly with University of Helsinki, Finnish Meteorological Institute (FMI) and Lund University. The network operates a common database where data from both physical and chemistry measurements from five stations Figure 5-54 are collected. The network is now expanding with the start of similar particle size measurements in Norway and Lithuania, in co-operation with the Norwegian Institute for Air Research (NILU) and University of Vilnius.

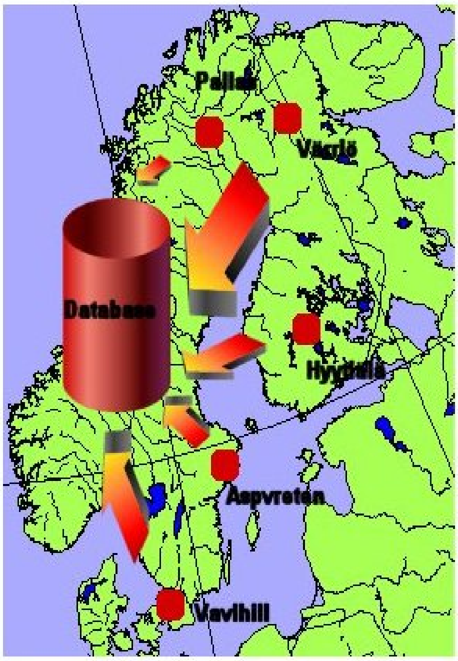


Figure 5-54 Super stations in the Nordic network.

This co-operation and common database facilitates investigations of the climatology of the Nordic aerosol and to evaluate how the long range transported aerosol varies with season and origin. The measurement data can be used to follow a certain air trajectory detecting changes in properties and composition caused by atmospheric processes occurring during transport and influences from sources located between the stations. A first report by Tunved et al., 2002, shows that there does exist a pronounced seasonal variation of number concentrations as well as shape of the size distributions. The number concentrations reach a maximum during summer time. This applies to all stations. Furthermore, the shape of the size distribution is found to be typically bimodal during wintertime with a larger fraction of accumulation mode particles in comparison to the other seasons. Highest Aitken mode concentrations are found during spring and summer. An established nucleation inventory shows that the maximum of nucleation events is confined to the spring months. Nucleation events do occur during other seasons as well, although less frequently.

There are also large differences between stations. Especially in terms of absolute number concentrations. Lowest concentrations were encountered at the northernmost stations Pallas and Värriö. Also the shape of the size distribution differs markedly when comparing stations at different locations. Pallas and Värriö exhibit data with well-separated Aitken and accumulation modes, while we observe a more smoothed size distribution at the southern stations. The seasonal characteristics of the aerosol at the stations depend also on the meteorology, which exposes the stations to air masses of different origin.

Using clustering techniques, hundreds of samples with trajectories passing the same areas under similar meteorological conditions can be collected. This gives an opportunity to calculate mean values and variations and to study source functions and transformations. The first result obtained from this approach was that the largest contribution to particle volume came from trajectory clusters of continental origin. Another interesting feature found was that the integral number concentration did not vary considerably when comparing clusters of different origin. Clusters arriving from the continent were clearly associated with size distributions shifted towards the accumulation mode. This behaviour was most pronounced at the southerly-located stations.

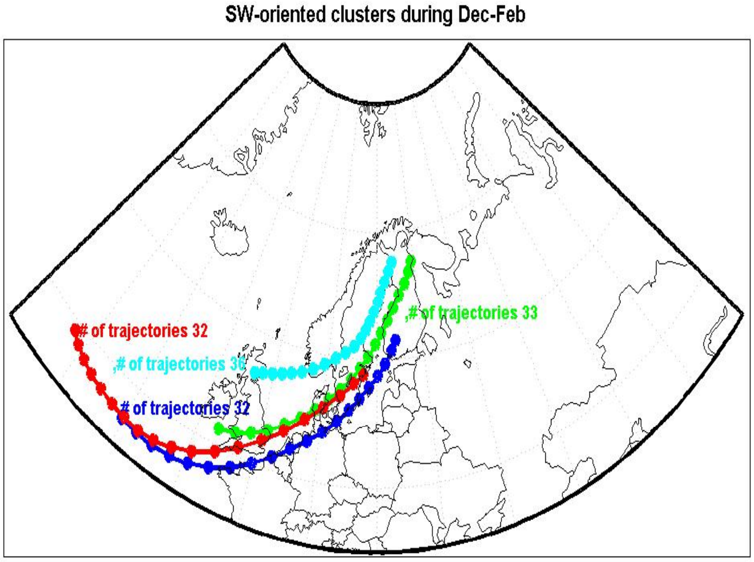


Figure 5-55 NE oriented clusters arriving the different stations. Clusters are represented as means, each endpoint corresponding approximately 5 h.

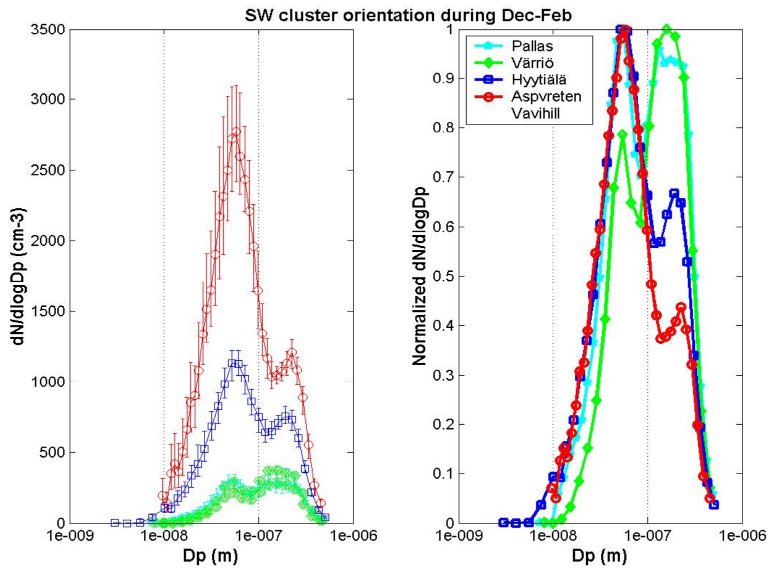


Figure 5-56 Comparing resulting median size distribution between different stations with clusters of similar orientation. (to the left) Resulting size distribution and normalised size distribution for period DEC-FEB. Error bars indicate quantile ranges.

When evaluating the measurement results from the different stations on occasions when the same meteorology seemed to affect all stations, significant differences where detected. In air masses originating from the north, sources located between the measurement sites contributed to the number of Aitken mode particles. Nucleation in the atmosphere is also a probable source. When following air masses originating in the south, Figure 5-55, a strong decrease was observed when the air masses moved north, as shown in Figure 5-56, most likely dependent on wet deposition.

In total, the first investigations show that the size distribution of particles over the Nordic region is dependent of source areas, transport distance and atmospheric processes occurring during transport. The variation in particle number at a site is surprisingly small over the year, while the particle volume is strongly dependent on source area. This indicates that emissions of precursors to condensable gases mainly add to volume while emissions of primary particle do not add to the background number of particle as seen in air masses from the north. However the number concentrations of several thousands per cc, measured at the southerly stations, is beyond what is considered the natural background concentration. The work will be continued including application of dynamic aerosol models on well characterised cases to quantify sources and processes.

The results are fundamental for the development of useful regional models capable of describing size and chemistry in the airborne aerosol as well to describe the deposition accurately.

5.7.4 Develop and evaluate parameterisations based on the data collected within the network.

The present model approach of EMEP is to add the major components by mass to describe the total particle mass in the two size fractions PM2.5 and PM10. The formation of secondary organic mass is estimated according to the Pankow/Odum approach. However, depending on temperature effects (Simpson et al., 1997) these estimates are very uncertain. The organics in the present estimates range to roughly 20% of the total PM10 or rather PM1 as no coarse particle emissions are included in the emission database for primary particles.

About half of the observed concentrations of PM10 for the Nordic sites are explained by the present mass EMEP model (Lazaridis et al., 2001). However organics, sea salt and resuspended dust are not included in the estimate. The observations by Swietlicki et al, (2002), summarised above, estimate that the two latter sources contribute roughly 40% of the total particle mass which corresponds well to the missing fraction in the model estimates for southern Sweden. These results stress the importance of including resuspended dust and sea spray in models estimating both PM2.5 and PM10 concentrations.

Another important Swedish contribution is a parameterisation for sea salt emissions, extracted from laboratory experiments and flux measurements at sea, which will be useful in estimating the sea salt contributions (Mårtensson et al., 2002). This can be used either in a mass or dynamic aerosol orientated model.

In a project funded by the Nordic Ministers Council (and co-ordinated with the ASTA activities), further development of the aerosol description in the EMEP model is being made. The project is also focussed on support to emission estimates used as model input. Within this project, DNMI has implemented the Pankow/Odum approach to calculate the mass of organic carbon (Andersson-Sköld and Simpson, 2001). Univ. of Helsinki have developed a multi mono-modal dynamic aerosol routine that is able to simulate the effects of nucleation, coagulation and condensation on the initiated size distribution (Pirjola et al., 2000). The new multi mono dynamic aerosol routine will soon be tested in different case studies based on data from the Nordic network. This project is co-ordinated by EMEP MSC-West (DNMI) and with participation from Univ. of Helsinki, ITM, NILU and FMI.

EMEP has recently taken action to improve the understanding of the transboundary transport of particles by making new recommendations for their monitoring program. Determination of the organic and dust fractions of the aerosol has started by different tests. Other physical parameters such as number and size distribution are also important. The establishment of EMEP particle super sites is a high priority for the increaed understanding of source - receptor relationships of particles.

5.7.5 Implementation and testing of a dynamic aerosol module in the EMEP-model.

There are some regional/global atmospheric chemistry transport models which include dynamic aerosol processes, i.e. which are capable of describing the formation and transformation of particles in the atmosphere. Different ways of describing the aerosol have been chosen, like sectional, lognormal modes, moments and multi mono. All methods involve considerable simplifications either in details of physics or chemistry, mainly due to computational limitations. These simplifications make the evaluation of model performance using measurement data extremely important.

To discuss and to possibly issue recommendations on how the models should be developed and made suitabile for policy use, an EMEP workshop on implementation of dynamic aerosol models for large scale applications was organised in Helsinki, 30/1 –1/2 2002. The workshop was initiated by ITM.

In the workshop all models available in Europe were presented and discussed. Important uncertainties were identified. The workshop concluded that one of the most important missing factors was source descriptions for sea spray and soil dust. There is also insufficient information on key thermodynamic properties of many chemical components. Several processes were reviewed at the workshop and difficulties were acknowledged. However it was also recognised that several models were able to complete most of the identified deliverables as given in Table 5-5.

Table 5-5 Model deliverables related to aerosols.

Area of interest	Parameters	Comments
Acidification / Eutrophication	Particulate mass of SO_4, NO_3 and NH_4	
Health	PM10 and PM1 N(Aitken), N(accumulation)	Mass closure needed to accomplish source identification
Climate	optical properties such as scattering and absorption	

The workshop recommended that PM1 rather than PM2.5 should be used as a measure of fine particles. PM1 will reflect the fine particulates much better than PM2.5 and almost completely eliminate the contribution of dust sources. Furthermore, the need for monitoring data on all parameters was emphasised, as it is a necessity to achieve good model performance.

5.7.6 *PM1 or PM2.5 as particle indicator for health effect studies?*

There is presently a discussion within the EU CAFE programme concerning choice of suitable indicator for fine particles. The US EPA has decided that PM2.5 shall be used as indicator for fine particles, as it has been used in epidemiological studies yielding stronger correlation to health effects than PM10. However, there is strong evidence, as shown above, that both anthropogenic and natural dust contribute significantly to the PM2.5 levels. In Figure 5-57 an example of results from a measurement campaign running for 1 month in Stockholm is presented. Measurements were performed at a heavily trafficked street in the inner city and at a calm street in a suburban area. The number distributions at both places show absolute maxima to the right in the figure at sizes of 15 – 30 nm. The mass distribution of the inner city site has an absolute maximum at about 5 µm, but a minor maximum in range 0.1 – 0.3 µm. In the suburban site there are two roughly equal peaks at 0.3 and 2-5 µm range. It is obvious that PM2.5 concentrations are strongly affected by coarse particles in both environments. PM1 however is very much less affected. Using PM10 and PM1 will strongly simplify the assessment of sources. PM1 is almost only dependent on primary particle emissions from combustion and secondary formation in the atmosphere. PM10 will contain both PM1 and mechanically produced particles as e.g. resuspended dust. The coarse particle mass can be calculated using PM10 – PM1. Thus the two source types can be separated and control measures are more easily applied to the correct source type.

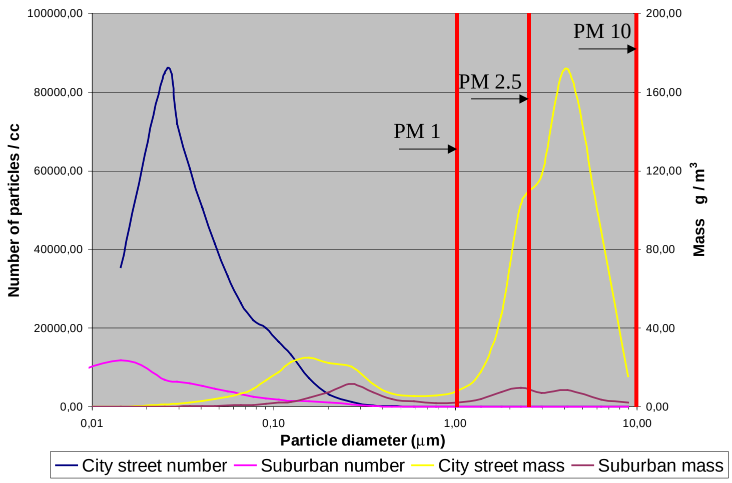


Figure 5-57 Number and mass size distribution measure at an inner city and a suburban site in Stockholm. PM1, PM2.5 and PM 10 are indicated.

There is also concern about the applicability of PM2.5 as an indicator of health effects. American epidemiological studies show quite a linear relationship to mortality (Dockery et al., 1994). An understanding of the effect of different particle sizes can be obtained by examining the IRCP Human Respiratory Tract Model for Radiological Protection as shown in Figure 5-58 (Låstbom and Campner, 2000). Superimposing the shown size distributions Figure 5-57 on the deposition efficiency in the human lung shows that the minimum in lung deposition coincides with the minimum in particle size distributions Coarse particles are efficiently deposited in the extra thoracic region, while the fine particle mass most efficiently deposit in the bronchial region. Looking at the number of particles, the largest fraction is deposited in the bronchial to alveolar region. However the deposition of the very finest particles can be considerable in the upper airways due to diffusion.

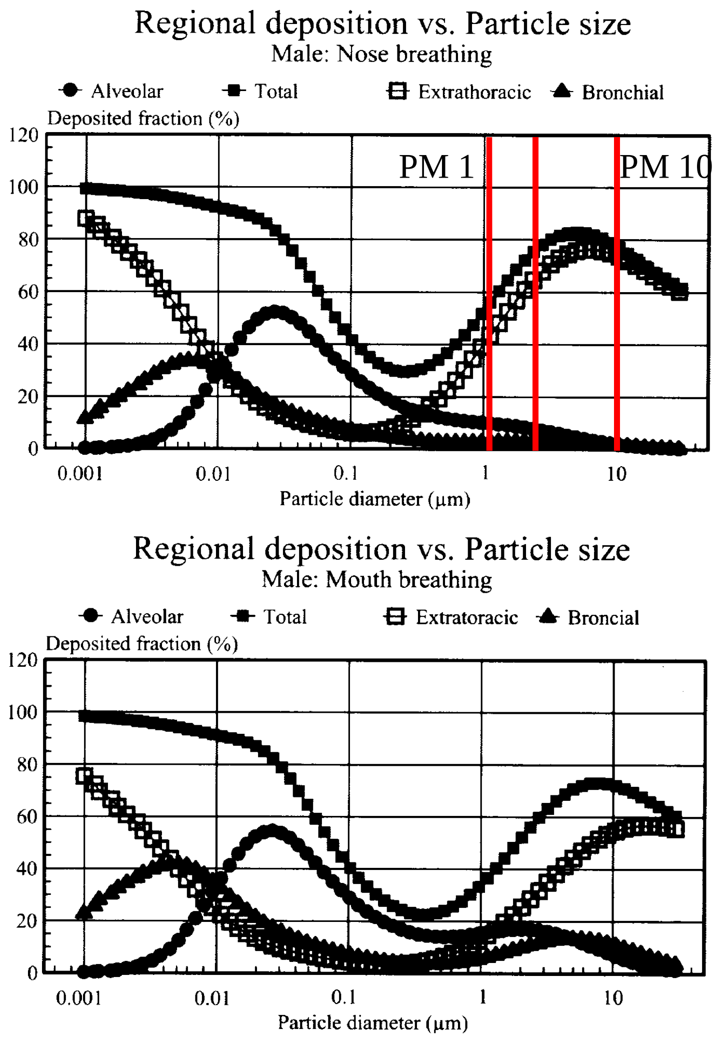


Figure 5-58 IRCP model for deposition of particle to the human lung when nose breathing. PM1, PM2.5 and PM 10 are indicated.

Consequently PM2.5 particles deposit in all regions of the respiratory system, while PM1 relate more to deposition in the bronchial and the alveolar regions.

Health effects due to elevated levels of PM1 will most likely depend on particles affecting the body through interactions via the bronchial and alveolar regions, while effects of coarse particles, i.e. PM10 –PM1 is more likely to be related to the upper airways. PM2.5 will thus not be that decisive in separating either health effects or particle source type.

6 Beyond 2010

6.1 Introduction

The policy work today aims for control measures which to a large extent will be implemented after 2010; target years that are mentioned are 2015 and 2020. For such distant time horizons, there will of course be difficulties to predict the driving forces and the status of relevant scientific issues which will influence our views on air pollution problems.

In this chapter we aim to discuss issues related to the development of future air pollution policies and what the main possibilities and obstacles might be. The chapter has not the aim to propose best solutions but rather to form a basis for further scientific research and discussions in structures forming the future air pollution strategies. In the chapter we will cover both issues of particular relevance for the policy development and issues that are most relevant from a scientific perspective

6.2 Driving forces

6.2.1 CLRTAP or EU

6.2.1.1 Legislative power

One question often raised in connection with future policies on transboundary air pollution is if it will be the European Union or The Convention on Long-Range Transboundary Air Pollution that will take the lead in Europe. Will EU with its more powerful legislation instruments drive the process or will the Convention with its wider geographical coverage and well developed organisational system of science-policy interactions be the forum that forms the strategies? Or will the two organisations find different roles within a joint process? These questions are of course of importance not only for policy formation but also for the development of scientific tools and concepts as well as the ways science may interact with policy.

From the scientific point of view, there is also the more fundamental question of to what extent science will be important for policy. The Convention has favoured scientifically derived policies in relation to policies driven by best available technologies (BAT), even if some countries have had a different opinion. The European Union policies and legislation are to a large extent formed from a BAT basis, in particular for motor vehicles and for large combustion plants. However, the EU also has some legislation based on environmental or health impacts e.g. the Air Quality Directives. Within the recently decided NEC directive and the policy work under CAFÉ a further change towards more effect-oriented and cost-effective approaches is emerging. Since the main instruments for the Commission are legislation towards the sectors, there are always strong arguments for a more technical approach in the legislation. This does not necessarily substantially decrease the needs for science. Scientific assessments will be important in the process of forming legitimacy for any new environmental legislation, especially when considering the increasing costs of most new control measures. The integrated assessment modelling approaches within CAFÉ and CLRTAP will be well suited for these purposes.

The Gothenburg Protocol was signed before the NEC Directive was finally negotiated and decided. Because of delayed negotiations on the NEC Directive, the European Union did not sign the Gothenburg protocol in December 1999. The agreed reductions were considered to be too weak from an EU perspective. When there was a final agreement on the NEC Directive, it was however obvious that the differences between the achievements in the Gothenburg Protocol and the NEC directive were very small. So, even if there were strong expectations on a much more stringent outcome of the NEC directive, the EU countries in most cases signed up to the same emission ceilings for 2010 in both the Protocol and the NEC directive.

International conventions rarely have any instruments for sanctions for countries which do not fulfil their obligations. This is also true for the CLRTAP and this is assumed to be a drawback of the Convention. One example of the lack of legally binding enforcement is the VOC protocol, where several countries have announced that they have not been able to reduce their emissions by 30% according to the protocol. At this stage it is not possible to say if the lack of fulfilment will have a large influence on the power of the Convention or if it is more a question of an unexpected delay in the implementation process. EU directives will have a stronger legislative power and countries not fulfilling their obligations may be subject to sanctions.

6.2.1.2 Participation in the process

During more than 20 years CLRTAP history, the science-policy collaboration has engaged a large number of scientists and scientific research has been established in many countries in support of the policy processes. Since the Convention almost by definition has to rely on initiatives from the participating countries, scientific contributions were identified as an important form of policy support and there has been a reasonably equal sharing of objectives, activities and resources between countries. Scientific networks were initially formed on an informal basis but have become formally adopted as part of the CLRTAP organisation. EU as a party under the Convention has, however, been very little visible as an organisation directed to promote collaboration between science and policy. Through its research Directorate, the Commission has financed scientific research of importance for air pollution strategies but it has to a very small extent been visible and used in connection with the policy development within the Commission.

6.2.1.3 Compounds and environmental problems considered

The CLRTAP process is directed towards compounds of importance from the transboundary point of view. The convention itself was originally formed to combat transboundary air pollution. The Convention will thus focus less on local (urban) air pollution even if these problems today are considered more important than effects occurring due to regional air pollution. For the EU, urban air pollution and health effects are given higher priority than the regional aspects, and the EU will in this respect probably be a more important driving force than the convention.

6.2.1.4 Geographical area

CLRTAP includes practically all countries in Europe and US and Canada. The European coverage includes in practice also Turkey and most of the former Soviet Republics. EU has until now only included the 15 member states but will in the future also include the accession countries. The Commission has also stated that the work within CAFE should also include all accession countries.

The Convention has already from its start established a close collaboration with the scientific community. Today, this support is substantiated in a large number of sub-groups and centres handling various issues for the support of scientific knowledge, models and data. These sub-groups and centres are mainly financed by the countries themselves (centres are financed by the country hosting the centre). In the CAFÉ programme there is no such system of scientific support and CAFÉ has, at least for its preparation of a revision document in 2004, declared that it will to a large extent rely on the Convention within these areas. There is presently no other solution in sight and the Convention is assumed to remain as the main organisation for the support of scientific knowledge and data.

6.2.1.5 CONCLUSIONS

With the present insight into the policy processes for transboundary air pollution in Europe, we are convinced that CLRTAP and EU both will be of crucial but somewhat different importance for the development of control strategies in Europe, at least for the coming five years. EU and the CAFÉ programme will not be able to compete in areas where CLRTAP already has established a functioning system. Instead the Commission has to rely on what is prepared under the Convention and on the delivery schedules established under the Convention. These areas include issues on source-receptor relationships on transboundary fluxes, all effects except effects on human health, emission inventories, evaluations of the outcome of control measures undertaken. The Commission will mainly concentrate its work to other areas, in particular health effects, particulate matter, emission scenarios and target settings. The work by the Commission will also cover local scale pollution.

We also believe that the different ways of policy directions may benefit from a parallel action of CLRTAP and EU. The determined way of forming policy through directives as done by the Commission may complement the bottom-up and process-oriented procedures established within the Convention.

6.2.2 Interactions with other policies

Emissions of air pollutants will of course also be influenced by other policies than those exclusively directed at air pollution. Policies on Climate Change are of particular interest and importance, since much of the emissions of air pollutants are directly related to combustion of fossil fuels. Climate change policies will therefore directly influence the emissions of sulphur dioxide and nitrogen oxides and a decrease in the use of fossil fuels will thus in most cases have a corresponding positive effect on acidification, eutrophication and effects from particulates and tropospheric ozone. The Kyoto protocol will mainly influence the situation until 2012 and will only reduce emissions less than 10%. Control measures after the Kyoto process are however expected to be larger and will thus have a larger impact also on emissions of air pollutants.

Agriculture is another area of importance for future policy. The Common Agriculture Policy (CAP) has formed the agriculture practice within EU-Europe and it has also had an influence on emissions of ammonia. The reductions of ammonia emissions within the Gothenburg Protocol and the NEC Directive are very limited and there is a need for much larger reductions if critical loads on eutrophication should be reached.

It is today very difficult to predict the future development within this area, in particular in relation to the expansion of EU to about 25 countries.

Energy and transport are the key sectors when developing policies on air pollution and long range transport. Different future scenarios for these sectors will be assessed in the integrated assessment work within the CAFE work and the results will form the basis of future abatement strategies.

6.2.3 Sustainable development

Sustainable development is a leading policy within EU and is aimed to influence the development of practically all sectors. Environment and resource utilisation is one of the fundaments and the implementation of the concept will certainly influence air pollution policies.

The sustainable development concept may even influence the settings of critical loads (Grennfelt 2000). The present critical loads concept for acidification allows acidification of soils and ecosystems and a substantial loss of alkaline substances. If, instead, a concept of sustainable ecosystems is applied a long term productivity of the soil will be required, i.e. that the soil will be kept at certain level of base saturation and nutrients. Such a change will give significantly lower critical loads and will thus require larger emission reductions.

6.2.4 Aiming for a semi-global strategy

Regional air pollution has so far been considered as continental problems although several investigations show that there is a substantial transport of pollutants between continents. So far, the influence from intercontinental transport has not been considered to be so large that it has requested special attention for policy. From a scientific point of view, it is mainly for ozone and to some extent sulphur that the intercontinental transport is so large that it may influence European air pollution control strategies.

In connection with the negotiations of the second sulphur protocol it was recognised that already the background deposition of sulphur was enough to exceed the critical loads for sulphur. In order to achieve realistic control strategies, adjustments were made in the maps of critical loads.

Background ozone concentrations have increased over the last century. There are strong indications that the yearly mean concentrations are more than twice as high in comparison to the concentrations 100 years ago. Present background concentrations are at levels where sensitive plants and ecosystems will be damaged or close to the critical levels used in the abatement strategies. Model calculations using the IPCC scenarios indicate a further increase in the ozone concentrations over the Northern hemisphere by approx. 20%. Such concentrations may certainly influence ozone damages to plants and thus air pollution strategies.

6.2.5 Optimised approaches or sector strategies?

One of the large successes of the Oslo and Gothenburg Protocols as well as the NEC Directive was that they were based on an optimisation of the least costs to fulfil agreed environmental targets. The optimisation procedure was a large step forward in the development of environmental strategies and there are expectations that the same type of strategy may be used even for the coming revisions.

The use of optimised strategies may however face problems that will become more obvious if used to reduce pollutant levels that approach the critical loads and levels;

problems, which need to be considered before the negotiations. The problems are both of a scientific nature and related to policy. The scientific problems are connected to the difficulties associated with control measures and energy, traffic, agricultural policies, reaching the critical loads and that optimisation becomes less robust at levels near the critical loads.

6.2.6 Problems related to the use of cost-efficiency approaches

A consequence of a step-wise cost-efficient strategy is that the cheapest control measures are taken in connection with the first negotiations and that the more expensive will be taken at later occasions. This means that for the renegotiations the costs for additional reductions may be substantially larger in comparison to the control measures undertaken in the Gothenburg protocol. At the same time the environmental benefits will be less obvious since more and more ecosystems are protected after each round of agreements. This is an inevitably consequence of the strategies. There may therefore be a communication problem to argue further reductions, where the environmental benefits will be less and the costs substantially higher. The use of the static critical loads concept will, as pointed out earlier, amplify this impression. Environmental benefits thus need to be expressed using more dynamic approaches and for systems with a better resolution than that of 150 km used for the Gothenburg Protocol.

Another problem in connection with future negotiations is that emission control requirements may become more unevenly distributed than in previous protocols. Control measures may be required in countries where the benefits will be very small – at least in relation to critical loads and levels. These problems may cause obstacles in the future negotiations and need to be considered in connection with both the choice of scientific research areas and in connection with the development of policies.

6.3 Scientific understanding and future needs for research and data

6.3.1 Introduction

The issues of scientific support to policies can also be regarded in a long-term perspective i.e. beyond the time scales of the next rounds of negotiations in CLRTAP and CAFE. CAFÉ has as already mentioned the objective to come back with a revision of the strategies and proposals for revisions of directives every five years and there will thus be a need for support after 2005. There are also needs for support for policy that can be made available already within the next three years and serve as support to policy although not being part of the present ASTA programme and objectives so far. This section will bring up some issues of crucial importance in this respect and discuss them in terms of further development and further needs for policy development.

6.3.2 The possibilities of reaching clean air in Europe

The final objective of air pollution strategies in Europe is to reach levels where air pollution will not cause any harm to human health and the environment. Based on what is expected as achievable within the EU- and CLRTAP processes, it is not expected that this objective will be reached through the decisions taken in the coming revisions of protocols and directives. Further actions will therefore be necessary in order to reach pollutant levels below critical loads and other environmental quality objectives.

The possibilities to reach sufficiently low emission levels will at the end depend on several factors, technology development and policy interest probably being the most important. Of the different environmental problems handled within ASTA, none will probably be solved even after the upcoming negotiations, even if the problems associated with acid deposition and regional ozone episodes will be diminished to very low levels in comparison to the situation at the end of the 20th century.

6.3.2.1 Acidification. Recovery as a part of critical loads.

So far, calculated critical loads for acidification have been used in Europe to set environmental targets for lakes and forest ecosystems. The inclusion of dynamic aspects as well as land use has been a large step forward in the development of more appropriate methods for assessing acidification effects as well as developing abatement strategies. We foresee in the future a more integrated approach in the use of the knowledge where air pollution deposition will be part of the overall land use management. In these respects the further application of the dynamic critical loads concept in Europe will be an important task.

A major problem in applying dynamic models and using dynamic model results as a basis for emission reduction negotiations is the difficulties in finding enough data to run the models all over Europe. The situation in Europe varies between countries; some countries have made very extensive mapping of soils and sufficient data for dynamic assessments is available. Countries with at least an acceptable availability of data include Austria, Croatia, The Czech Republic, Denmark, Finland, Germany, Hungary, Netherlands, Poland, Russia, Slovakia, Slovenia, Sweden, United Kingdom and Switzerland, in all 15 countries. When it comes to the application of the data into dynamic modelling, the situation is much more questionable. Some countries i.e. Denmark, Germany, Netherlands, Sweden and Switzerland have performed dynamic model calculations at a large number of sites. In other countries there are difficulties for one or another reason. These difficulties may arise from absence of motivation, skill, money or appropriate organisation, which in turn result in lack of data and model runs. Most common is probably the lack of financial support.

A prerequisite for dynamic effects to be taken into account in the negotiation process is that dynamic effects have been assessed for at least those grids that are likely to determine the overall emission reduction optimisation. In other words, it is more important to assess the dynamic effects, e.g. recovery times, in grids which have been subject to large exceedances than in grids which have not experienced exceedances of their critical loads. Fortunately most areas that have been subject to large exceedances have either planned/ongoing dynamic modelling activities or the potential to perform dynamic modelling activities. When dynamic effects have been assessed for several regions/countries the prospects for assessments throughout Europe – by extrapolation or model calculations are more realistic.

6.3.2.2 Ozone – background concentrations. AOT40 as a basis for critical levels

Peak ozone concentrations have decreased over central Europe since 1990, most probably due to the decreased emissions of nitrogen oxides and volatile organic compounds. In parallel there are indications of increased background concentrations as monitored at the outskirts in Europe. The increased background and the diminishing peak concentrations may influence the methodologies for estimating effects from ozone, in particular the AOT40 approach, as well as the control strategies.

AOT40 was a step forward when it was introduced. Inevitably it will, however lead to overestimation of effects in some climates and underestimation in other climates. The only solution to this is to use an ozone uptake based approach. Substantial research has now been performed showing that this is possible and that the estimations of effects are improved. Also the estimation of absolute level of effects will be improved compared to the AOT40 approach, since the problem of monitoring height (see e.g. Pleijel 1997) disappears with the flux-based models. The ozone-flux dependence on environmental variables will have to be built into the next generation of critical levels for ozone to avoid criticism that the best available scientific information has not been used.

Analysis of potato, wheat and timothy data reveal that the critical fluxes tend to start to accumulate around 20 ppb ozone if conductance is close to maximum. The difference from the AOT40 is not necessarily as large as it first may seem like, since the flux accumulation in the range of 20 to 40 ppb is not very large, but it may be large in climates where high conductance is relatively common. In dry climates the threshold may effectively be above 40 ppb. The further development of flux-based methodologies and applications need to take into account the changes in concentrations and investigate if critical levels even should be able to take into account exposures under background conditions.

6.3.3 Human health and particles

The shown relation between air pollution and health has started intensive research efforts to find the crucial chemical and/or physical components of air pollution and how they affect humans. Presently more than 100 publications on the subject or in related areas appear in literature per year. This will certainly give us a much deeper and detailed knowledge on what is harmful within the next 5 years. Within this time frame a new general concept will likely emerge about what the major focus should be. Our vision is to participate in this work on a European base and to give valuable contributions.

To be able to meet the possible future needs, the basic approach is to establish knowledge, models and measurements in a concept that make it possible to describe concentration fields of whatever component or property that might be of interest. This concept includes the description of emissions, transport, transformation and deposition and is based on a comprehensive atmospheric model. With this "tool" and the measurements necessary to support the model we will have the possibility to describe fully the connection between the occurence of particles in the air, particle precursor emissions and human health impact.

The future studies of health effects due to air pollution need substantial and detailed physical and chemical information on what an air pollution episode contains to further deepen the understanding of the relation between different health effects and air quality. This implies that an close interaction between scientist working with developing models and more broad understanding of air quality and scientists working with health studies will strongly enhance the progress concerning the understanding of the link between pollution and health.

6.3.4 Urban air

The general concept is that the urban air is more harmful to humans than "fresh" air in rural areas. The identified long range transport of air pollution shows clearly that no such "fresh" air exist. Aerosols are transformed and diluted to lower concentrations after some days transport by the winds, but may still reach considerable concentrations far from the major source areas.

In urban areas the future emissions will look quite different from the emissions of today. There is intensive work ongoing to refine the diesel engine, both for light and heavy vehicles, to a “clean” power source by adding exhaust treatments as particle filters and catalyst. The driving force is that these engines are more fuel efficient and thus give lower CO_2 emissions. If the car and truck industry succeeds to introduce particle filters on their vehicles, the particle mass, especially the soot emissions, will decrease with roughly two orders of magnitude. The emissions might still have about the same *number* of particles in the exhaust. These particles will be much smaller and mainly formed from remaining organic condensable gases in the exhaust.

Due to an expected major change in the energy production towards more use of biomass combustion, a major change is also to expect in the emissions from small to medium size power plants.

In a long term perspective the urban air pollution of particles will probably change towards considerably lower mass concentrations. Even though particle mass will decrease, the number will perhaps be in the same range. The particles will be smaller and more of secondary origin. Biomass combustion will contribute a larger fraction of the urban aerosol. These sources emit aerosols that are drastically different than those emitted from fossil fuel combustion, probably larger but fewer and more soluble.

It is difficult to state whether this change in emission properties will diminish the fraction that will be available for long range transport. The smaller particles have a longer life-time in the atmosphere, while hygroscopic particles have a shorter. The long range transported aerosol in the Nordic countries depends mostly on emissions on the continent, where different scenarios than in the Nordic countries may be relevant. Considering the large influence that the long range transported aerosol has today it is most likely that it will play an important role also after 2010.

Presently low limit values for ambient particles are expressed in particle mass, PM10. This is due to the fact that almost all investigations on the relation between particles and health are performed using particle mass as indicator of particles. This is mostly due to traditional air pollution monitoring methods, which since the days of the London smog, includes measurements of particle mass. Consequently extensive measurement networks with long time records on particle mass concentrations are available for use in epidemiological studies of the relation of particles and health. However several other investigators have suggested other size fractions, chemical components and other parameters as potential indicators or direct cause of health problems. But those investigations have by necessity been based on smaller data sets and less time coverage.

In chapter 5 we have strongly suggested PM1 as a measure for the exposure to fine particles. This size range is dominated by long range transported aerosol and by local combustion sources. Using the two PM measurement classes PM10 and PM1 gives a better possibility to assess the sources, for coarse and fine particles respectively.

Other specific components/properties that probably will be considered in relation to health is Black Carbon (BC), Organic Carbon (OC), PAH, soluble – non soluble substances and number of ultrafine particles. In several health investigations a good correlation between soot, according to the OECD-standard, and health effects has been found. Presently BC and number of ultrafine particles are discussed as new certification standards for vehicles. OC contains several toxic compounds, e.g. PAH, but the main fraction of the mass, about 80%, is still unspecified. The farthest progress has been reached in the water- soluble fraction of the OC addressing functional groups (Faccini et al., 2000). The insoluble part is still largely unknown representing roughly half the OC concentration (Zappoli et al., 1999).

There is strong possibility that new indicators and/or components affecting health will be found within the coming 5-7 years. This has to be considered when setting up the strategy for monitoring and model development.

6.3.5 Terrestrial biodiversity; forests, heathlands and bogs

Biodiversity changes due to N deposition was not considered in the optimisation procedure in connection with the Gothenburg Protocol or the NEC Directive. Nevertheless, N deposition has caused large changes and may be considered in another more important way in connection with the CAFÉ programme and the re-negotiations of the protocol. One of the reasons for leaving it out may have been the lack of coherent databases and models covering all Europe. Another may be the relation with N induced stresses and stresses caused by other environmental factors (acidification, land use changes, climate change)

Presently used models for nitrogen assessment of critical loads are either empirical or based on mass balance approaches. The empirical models have in general poor resolution and the applied values have wide ranges. Mass balance models are reasonably robust, but the connection to ecological effects still remains weak. The need for new innovative tools is therefore large.

In the plans for a second phase of the ASTA programme, the possibilities of developing a dynamic model by which the consequences of N deposition will be analysed will be explored. In a first step the model will be developed for the Southern part of Sweden. The model should include possibilities for the assessment of combined effects of N and S deposition scenarios (eutrophication and acidification effects), climate change scenarios and different forest practices. The intention is to have a model running by 2005.

We aim to integrate feedback mechanisms and cause/consequence relationships between soil acidification, nitrogen eutrophication and climate change on vegetation development, particularly for managed forest, through further development and integration of existing mathematical models. The model system will not only support decisions within the CLRTAP system but also for policy-related work in connection with the conventions on climate change and biodiversity. Nationally the model may support strategies and decisions for the management of Swedish forests.

The methodology will be based on available European knowledge and models for ground vegetation dynamics and will take into account factors such as present vegetation, soil water, temperature, soil nitrogen, soil acidity and geochemical properties of the soil.

6.3.6 Land use

Future demand on renewable materials will contribute to a more intense forest management. As already shown earlier in this report, a more intense forest management and extraction will make substantial contributions to the acidification of forest soils. This may in the long run cause nutrient deficiencies in the forest ecosystem. Already today forestry contributes to the ongoing acidification of lakes and streams and thus prolongs the recovery time of damaged ecosystems. An acceptance of a more intense forestry may therefore be dependent on the possibilities to counteract the acidification. The forest sector works with measures to counteract acidification and leakage of nitrogen caused by harvest, but there is today little interest in compensating the contribution from atmospheric deposition.

Assessments of the future need of emission reductions influencing Sweden must be based on the interactions between land use and impacts from air pollutants. Simulations of future acidification and eutrophication of forest soils must be based on realistic scenarios for land use, in parallel to deposition scenarios. Further development of dynamic models to deal with the effects caused by growth and harvest should be developed in order to assess the impact of different management scenarios (e.g. whole tree harvest, different species, rotation period and fertilisation) on the possibilities of forest soil and surface water recovery from acidification.

Future accumulation of nitrogen and the risk of nitrogen saturation and enhanced leakage in forest soils in areas in Sweden with high deposition are dependent on the intensity of forestry. Intense forestry can probably counteract accumulation of nitrogen better than unmanaged areas (Westling & Akselsson, 2002).

7 Conclusions

In this report we have described a number of new approaches and developments of importance for the future use in air pollution strategies in Europe. The intention has not been to cover all relevant aspects. The focus has been on new concepts for describing environmental effects in a more dynamic manner and present ways on how to integrate these into models and tools of importance for the development of policies.

The report is also a status report for the ASTA research programme at the end of its first phase and on the doorstep to its next. The ideas and results presented here are therefore open for discussion certainly within the ASTA community but also for anyone who has views and interests in this issue. Since the overall objective of the programme is to support policy we are particularly interested in views that can improve the scientific support and legitimacy in this process.

Scientific research in the area of transboundary air pollution in Europe works within limited timeframes. The schedules, especially for the CAFE programme is tight with defined times for deliveries. The deliveries are focused on the development of strategies and proposals based on integrated assessment modelling, by which cost-effective solutions should be obtained. Integrated assessment solutions are however not the only input to the policy process. Other factors of similar importance are improvement of scientific understanding in general, examination and evaluation of the underlying science and outcome of the integrated assessment models and deeper analyses of the outcome of already undertaken measures. Results from such research will not be limited only to the time limits of CAFÉ. The schedule for the revision of the Gothenburg protocol is less strict and since CLRTAP probably will take more interest in environmental effects, research on these issues will have possibilities to be linked into the IAM process at a later stage than in the CAFÉ programme.

By this report we have also demonstrated that the inclusion of new aspects will increase the relevance and applicability of using effect-based approaches in regional air pollution strategies. New aspects may on one hand add complexity to an already complex model structure and there is a risk that transparency and reliability may be lost. On the other hand we have also demonstrated that the previous concepts and methods suffer from serious limitations that may make them questionable in future strategies in particular at levels approaching critical loads and levels.

One of the large challenges in the further work with the inclusion of these aspects in IAMs is therefore to show the advantages with the more complicated systems.

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