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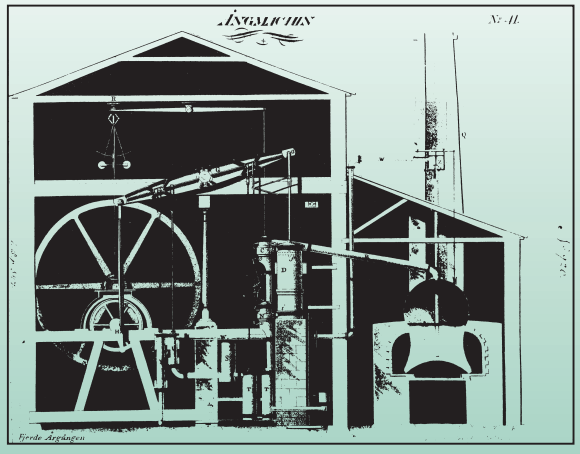

SANNA SYRI

Air pollutants and energy pathways: Extending models for abatement strategies

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19

Sanna Syri

Air pollutants and energy pathways: Extending models for abatement strategies

Yhteenveto: Ilmansaasteet ja energiaskenaariot: malleja rajoitusstrategioiden suunnitteluun

FINNISH ENVIRONMENT INSTITUTE, FINLAND
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Contents

List of original publications and the author's contribution

This study consists of the following original publications, which are referred to by their Roman numerals. The author's contribution in each article is described below.

I Syri S., Johansson M. & Kangas L. 1998. Application of nitrogen transfer matrices for integrated assessment. *Atmospheric Environment* 32 (3): 409–413.
The author of this thesis was responsible for manuscript coordination, implementation of new nitrogen transfer matrices, development and testing of the DAIQUIRI model code and for comparison calculations. Development of the comparison method and assessment of results were done jointly with the other authors.

II Ruoho-Airola T., Syri S. & Nordlund G. 1998. Acid deposition trends at the Finnish Integrated Monitoring catchments in relation to emission reductions. *Boreal Environment Research* 3: 205–219.
The author was responsible for deposition estimates using the DAIQUIRI model and model-measurement comparisons. Assessment of results was done jointly with the other authors.

III Ahonen J., Rankinen K., Holmberg M., Syri S. & Forsius M. 1998. Application of the SMART2 model to a forested catchment in Finland: Comparison to the SMART model and effects of emission reduction scenarios. *Boreal Environment Research* 3: 221–233.
The author of this thesis developed the DAIQUIRI model code to obtain site-specific historical and future deposition trends and was responsible for the modeled deposition trends.

IV Syri S., Johansson M., Grönroos J. & Ekqvist M. 1999. Assessing the effects of national and international energy scenarios and emission reduction strategies on acidification in Finland. *Environmental Modeling & Assessment* 4: 103–113.
The article describes assessment done for the Finnish National Acidification Committee, in which the author acted as scientific secretary. The author is responsible for the manuscript, model calculations (excluding ammonia emissions) and the assessment of results.

V Syri S., Amann M., Schöpp W. & Heyes C. 2001. Estimating long term population exposure to ozone in urban areas of Europe. *Environmental Pollution* 113(1): 59–69.
The author of this thesis is responsible for the manuscript, development of calculation routines and calculations. Method development and assessment of results were done jointly with the other authors.

VI Syri S., Amann M., Capros P., Mantzos L., Cofala J. & Klimont Z. (in press). Low-CO_2 energy pathways and regional air pollution in Europe. *Energy Policy*.
The author is responsible for implementation of energy data for RAINS model calculations. SO_2 and NO_x emission calculations, assessment of results and manuvNript preparation were done jointly with other authors.

VII Syri S., Suutari R. & Posch M. 2000. From emissions in Europe to critical load exceedances in Finland – Uncertainty analysis of acidification integrated assessment. *Environmental Science & Policy* 3(5): 263–276.
The author is responsible for the manuscript and task coordination. The method development and assessment of results were done jointly with the other authors.

VIII Syri S. & Karvosenoja N. (in press). Low-CO_2 energy pathways versus emission control policies in acidification reduction. *Water, Air and Soil Pollution* 128.
The author is responsible for the model calculations (excluding Finnish SO_2 and NO_x emissions), for the manuscript preparation and for the assessment of results.

Symbols and abbreviations used in the study

AOT60	Accumulated exposure Over Threshold of 60 ppbv
B1	Emission reduction scenario used in the development of the National Emission Ceilings Directive of the EU
CCE	Coordination Center for Effects under the Convention on Long-Range Transboundary Air Pollution
CLE	Current Legislation –emission scenario
CLRTAP	Convention on Long-Range Transboundary Air Pollution
DAIQUIRI	Deposition, AIr QUality and Integrated Regional Information –model developed at the Finnish Environment Institute
€	Euro
eq	equivalent (moles of charge) e.g. 1gS = $\frac{1\,g \cdot 2\,eq\ mol^{-1}}{32\ g\ mol^{-1}}$ = 0.0625 eq and 1 gN= $\frac{1\,g \cdot 1\,eq\ mol^{-1}}{14\ g\ mol^{-1}} \approx 0.0714$ eq
EMEP	Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe
EMEP/MSC-W	Meteorological Synthesizing Centre – West of the EMEP
EU-15	European Union with the 15 member countries
FMI	Finnish Meteorological Institute
FMI-RM	The Regional Model of the FMI
IAM	Integrated Assessment Model
IEA	International Energy Agency
IIASA	International Institute for Applied Systems Analysis
IPCC	Intergovernmental Panel for Climate Change
MFR	Maximum Feasible Reductions -emission scenario
NEC Directive	National Emission Ceilings Directive
NO_x	nitrogen oxides (NO_x = NO + NO_2); compounds emitted in combustion
NO_y	reactive nitrogen (NO_x + products of atmospheric oxidation of NO_x); compounds deposited to ground
OECD	Organisation for Economic Co-operation and Development
ppbv	volumetric parts per billion
PRIMES	Energy system model for the EU-15 of the National Technical University of Athens
RAINS	the Regional AIr pollution INformation System – integrated assessment model of IIASA
REF	Emission scenario incorporating all implemented and agreed upon emission control legislation
RIVM	the Dutch National Institute of Public Health and the Environment
SYKE	Finnish Environment Institute
UN/ECE	United Nations Economic Commission for Europe
UN/FCCC	United Nations Framework Convention on Climate Change
VOCs	Volatile Organic Compounds
WHO	World Health Organization

Air pollutants and energy pathways: Extending models for abatement strategies

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Syri, S. 2001. Air pollutants and energy pathways: Extending models for abatement strategies, Monographs of the Boreal Environment Research No. 19, 2001.

This study presents the development and applications of regional and local scale models for use in integrated assessment of air pollution effects in conjunction with large-scale models. A regional deposition model called DAIQUIRI (Deposition, AIr QUality and Integrated Regional Information) for integrated assessment purposes in Finland was constructed, and regional matrices for nitrogen oxides and ammonia were developed from the results of the regional air quality model of the FMI. DAIQUIRI produced similar estimates of deposition from Finnish sources as the original model, and long-term trends and the average level of deposition estimated with DAIQUIRI were found comparable with the monitored deposition levels and trends. For the mid-nineties situation, the regional nitrogen modeling resulted in 9 % to 19 % (depending on the region compared) larger estimates of areas with acidity critical load exceedances than when using European scale nitrogen deposition modeling. In this work, also a method for estimating the impacts of local NO_x emissions on urban and sub-urban ozone levels was developed and tested. The study concentrated on representing the destruction of ozone by fresh NO emissions in urban areas for future use in integrated assessment modeling of ozone control strategies. Correlation coefficients between measured daytime ozone values in the study area were found to improve from 0.64 (correlation between urban and surrounding rural measurements) to 0.85, on the average. The average correlation between daytime large-scale model estimates and urban site measurements was found to improve from 0.37 to 0.58. In the study, also integrated assessment model applications were carried out at European, national and local levels. The synergies between control strategies for CO_2 and acidification and ozone formation in the case of the UN/FCCC Kyoto protocol and the air quality targets of the EU were assessed with the help of coupled models. With two alternative energy scenarios reflecting the Kyoto targets for CO_2, reductions of sulfur and NO_x emissions between 12 % and 22 % and 8 % to 12 %, respectively, were estimated by 2010 in the EU-15 with the present emission control legislation. Due to the lower activity levels generating less emissions and the cleaner energy forms used, 35–43 % cost savings in further technical emission controls required for achieving the EU air quality targets would be achieved with the scenarios studied. Case studies for Finland indicated that there has been a decrease of 60 % in the area at risk of acidification from 1990 to 1995, and that the declining trend is expected to continue due to the recent international emission reduction agreements within the UN/ECE and the EU. Implementation of the Kyoto protocol in Finland and in the whole of EU-15 (with the present emission legislation) could bring up to 8 % more reduction of ecosystems at risk of acidification in Finland by 2010 than the recent UN/ECE protocol. An uncertainty analysis of acidification integrated assessment modeling in Finland indicated that critical loads dominate the uncertainty. Estimates are becoming more robust, as the general level of deposition is decreasing. In Finland, further efforts to reduce the overall uncertainty should be mainly directed to more accurate description of critical thresholds. In areas affected by major nearby emission sources, also uncertainties in emissions and deposition are significant. The models and their applications presented in this study contributed to identifying the problem characteristics and have supported environmental policy development at international, national and regional levels.

Keywords: regional modeling, nitrogen, deposition, acidification, tropospheric ozone, fossil fuels, sulfur, nitrogen oxides, carbon dioxide

1 Introduction

1.1 Anthropogenic emissions to the atmosphere and resulting environmental problems

The global energy consumption has increased exponentially during the past two centuries. The growing energy need has been satisfied predominantly with fossil fuels. In 1990, the global energy consumption was growing at a rate of about 2.5 % per annum. The Intergovernmental Panel on Climate Change (IPCC) forecasts a continued growth of energy demand in its recent scenarios (Nakićenovič 2000). Fig. 1 illustrates the growth of the global primary energy consumption in the main source categories from 1860 to present. (IEA 1991,1997, IPCC 1996, Nakićenović 2000). Also shown are the ranges of the IPCC scenarios projected up to the year 2100, with the main scenario with the highest energy consumption (IPCC scenario A1F1) in Fig. 1(a), and the lowest (IPCC scenario B1) in Fig. 1(b) (Nakićenović 2000).

The industrialized countries, which comprise less than 20 % of the world population, consume presently about two thirds of the total commercial energy. In the future, the growth of energy consumption and especially the use of fossil fuels is predicted to concentrate in developing countries. The projections of plausible energy futures of the IPCC cover a wide range of alternatives, as illustrated in Fig. 1. In all main scenarios of the IPCC, fossil fuel consumption is predicted to grow at least until 2040.

The expanded burning of fossil fuels has caused large emissions of combustion products into the atmosphere. Burning of carbon-containing fuels (fossil fuels and biomass) causes the carbon (C) to be oxidized and emitted into the atmosphere. Coal and oil contain usually between 1–4 % sulfur, most of which is oxidized and emitted into the atmosphere during combustion. In all combustion processes nitrogen oxides are formed, the amount depending on the nitrogen contents of the fuel and on burning conditions. Also industrial processes (*e.g.* metals production, pulp and paper production, etc.) are considerable sources of SO_2 and NO_x emissions. Fig. 2 displays the development of global CO_2, NO_x and SO_2 emissions during 1860–2000 (IPCC 1996, Dignon and Hammed 1989, Nakićenović 2000). In Fig. 2, the slowing down in the growth of sulfur emissions is visible after 1980. This is caused by the introduction of sulfur abatement policies especially in Western Europe and North America. In developing countries also sulfur emissions are still growing. Also the growth of global CO_2 emissions has slowed down after the 1980s. This is due to improved energy efficiency in the western countries and economic recession in the former Soviet Union and Eastern Europe.

Also agriculture has been greatly intensified during the 20th century to increase yields to feed a growing population. The introduction of artificial fertilizers and increased animal husbandry have caused emissions of large quantities of ammonia (NH_3) into the atmosphere.

The atmospheric changes caused by human activities have induced a manifold of local and regional environmental problems. High concentrations of sulfur oxides are poisonous to humans and

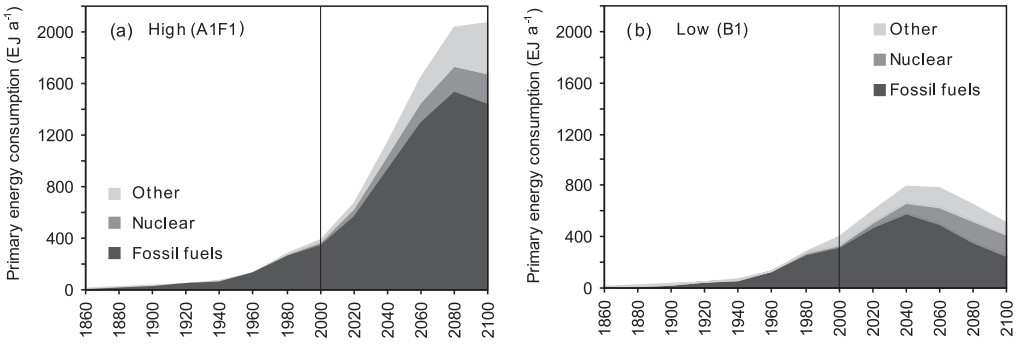

Fig. 1. Global primary energy consumption 1860–2100 with the main source categories. Category "Other" comprises biomass and other renewable energy sources. The vertical line marks the year 2000. The highest energy consumption in the IPCC main storyline scenarios is shown in (a), and the lowest one in (b) (Nakićenović 2000).

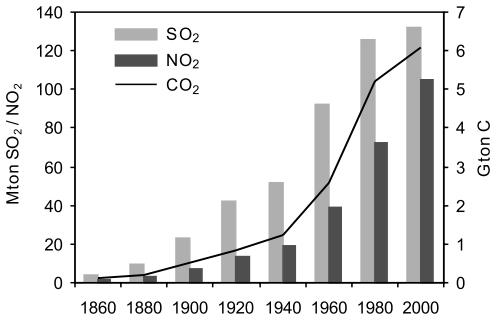

Fig. 2. Estimated annual global energy-related CO_2, NO_x and SO_2 emissions during 1860–2000.

vegetation. Depositions due to sulfur oxide, nitrogen oxide and ammonia emissions cause acidification of ecosystems. Nitrogen oxides contribute also to the formation of ozone in the lower troposphere (*i.e*. at ground level). In addition, nitrogen dioxide is hazardous to human health. Both nitrogen oxides and ammonia cause eutrophication of ecosystems.

The air pollution problems caused by fossil fuel burning have been long known. In London, coal burning was prohibited for air pollution reasons for the first time in 1273. The problem of acid rain, with its harmful consequences on forest and lake ecosystems, was discovered by Swedish scientists in the late 1960s (Oden 1968), and research confirmed that it was due to polluted air masses transported over long distances. Acidification has been among the most serious threats for natural ecosystems for decades in large areas of Europe and North America. In 1990, for instance, 24.7 % and 16 % of the total ecosystem area in the present EU-15 countries and in the whole of Europe, respectively, were estimated to be at risk of acidification (European Commission 1999, Amann *et al*. 1999b). Correspondingly, 55 % of the ecosystem area (excluding sea regions) in the present EU-15 countries and 25 % in Europe was estimated to receive eutrophying nitrogen deposition in excess of their tolerance limits in 1990 (European Commission 1999, Amann *et al*. 1999b).

Leaching of nitrogen from agricultural and forestry activities together with discharges from industry and households cause the main part of eutrophying nitrogen loading of waters. However, the share of atmospheric deposition is also significant and growing due to the increased control of the direct sources. Eutrophication of the Baltic Sea caused by extensive inputs of nitrogen and phosphorus is one of the most serious environmental problems in Northern Europe. Along with increased control of large point source polluters and diffuse pollution from agriculture, the contribution of nitrogen originating from atmospheric deposition is receiving growing attention. Atmospheric nitrogen deposition is mainly in inorganic form, and thus readily available for algae production.

Tropospheric ozone concentrations regarded as harmful for human health are frequently encountered in Central and Southern Europe during summertime. Throughout the nineties ozone levels regarded as potentially hazardous for human health have been recorded in most European countries (*e.g.* Sluyter and van Zantvoort 1997). Model studies and observations have indicated also ozone levels harmful for vegetation in all European countries in the 1990s (European Commission 1999, Amann *et al*. 1999, Laurila 1996).

Besides the regional and transboundary environmental problems caused by sulfur, nitrogen and volatile organic compounds (VOC), it is widely recognized that the anthropogenic inputs of primarily CO_2 to the atmosphere are causing a notable warming effect of the global climate. This is at present regarded as the most profound impact of human activities on the global environment.

To summarize, during the past century the changes in the atmosphere caused by human activities, primarily fossil fuel burning, have reached an extent at which environmental pressures necessitate a change in the current practices of exploiting the fossil fuel reserves.

1.2 Agreements to control long-range air pollution and greenhouse gas emissions in Europe

The issue of transboundary air pollution first reached international attention at the UN Conference on the Human Environment held in 1972 in Stockholm. Consequently, The Organization for Economic Cooperation and Development (OECD) launched a monitoring program for observing long-range transported acidifying air pollution in Europe. In 1979, the Convention on Long-range Transboundary Air Pollution (CLRTAP) under the UN Economic Commission of Europe (UN/ECE) was signed (UN/ECE 1979). The CLRTAP has

been the driving force for reducing transboundary air pollution in Europe, as protocols were signed to abate emissions of sulfur (1985, 1994), nitrogen oxides (1988) and volatile organic compounds (VOCs) (1991). In December 1999, a protocol to abate acidification, eutrophication and ground-level ozone was signed in Gothenburg, Sweden (UN/ECE 1999a). The protocols have resulted in a notable limitation of especially sulfur emissions in Europe during the 1980s and 1990s. With the Gothenburg protocol this positive trend is expected to continue in many parts of Europe and involve nitrogen and VOCs as well.

In the 1990s the European Union assumed a more active role in fighting transboundary air pollution. The European Commission published its strategies against acidification and tropospheric ozone in 1997, which resulted in a proposal for a National Emission Ceilings Directive in 1999 (European Commission 1999).

The international response to mitigate climate change was organized under the United Nations Framework Convention on Climate Change (UN/FCCC) at the UN Conference on Environment and Development in Rio de Janeiro in 1992. In 1997, the first protocol with concrete emission reduction requirements was signed in Kyoto (UN/FCCC 1998), but its entry into force is still uncertain. The European Union developed a burden sharing agreement, in which the member countries were allocated a reduction target taking into account their circumstances and development trends.

1.3 Modeling approaches

In the past, legislation for controlling harmful emissions used to address single sectors, single substances or single environmental problems, *e.g.* the limitation of sulfur emissions (*e.g.* Sulphur Committee II 1993, UN/ECE 1994). The control of long-range transported air pollution in Europe started with flat-rate emission reduction agreements of single substances during the 1980s. It was, however, soon realized that this was not the most efficient way of tackling the problems, as both the severity of environmental problems and the average emission reduction costs varied greatly across Europe. Therefore, integrated assessment models (IAMs) were developed to analyze possibilities for more cost-efficient reduction strategies.

The features of IAMs and related concepts are described, *e.g.* in Hordijk and Kroeze (1997) and Johansson (1999). In brief, integrated assessment modeling brings together information from a broad range of disciplines and helps to communicate the features and possible remedies of the problem between scientists and policy-makers. Integrated assessment models have to be transparent and oriented towards showing the general features rather than describing in detail the biological, physical, chemical or technical processes of concern. Integrated assessment models of air pollution link information about emissions, their control costs, atmospheric transport and transformation of the pollutants and the effects on the environment or human health.

Research on the reaction mechanisms of ecosystems to acidifying deposition and on their tolerance against acid inputs resulted in the introduction of environmental criteria suitable for use in control strategy development. In 1988 the concept of critical loads, defining the maximum pollutant load that a specified (sensitive) part of an ecosystem can tolerate without harmful long-term effects, was introduced (Nilsson and Grennfelt 1988). Similarly, as a response to increased concern about European population and vegetation being exposed to harmful ozone concentrations, threshold values for the protection of human health and vegetation have been established. They are expressed as cumulative concentrations above a threshold concentration over a defined period. For vegetation, the agreed threshold is 40 ppbv, with critical exposure amounts and accumulation periods given in Fuhrer *et al.* (1997). For human health, the threshold concentration of 60 ppbv has been used in the European control strategy development, reflecting the ozone air quality guidelines of the World Health Organization (WHO 1995).

The European assessment work is partly based on the Regional Air Pollution Information and Simulation (RAINS) model, developed at IIASA, Austria, which has been the prominent model supporting the European emission reduction efforts (*e.g.* Alcamo *et al*. 1990, Schöpp *et al.* 1999). RAINS was used to support the negotiations on the Second Sulphur protocol in 1994, where for the first time environmental tolerance, defined as critical loads, was taken as the basis of the protocol (UN/ECE 1994). RAINS contains optimization modules, which allow to calculate cost-minimal emission reduction allocations across Europe to achieve a predetermined environmental improve-

ment. For the Second Sulphur protocol, the optimization mode of RAINS was used to find the least-cost emission reductions for European countries to achieve a 60 % reduction in excess sulfur deposition everywhere in Europe, which was taken as the basis for the political negotiations.

In the nineties, the so-called multi-pollutant/multi-effect approach in integrated assessment modeling was developed. It addressed the emissions of four pollutants: sulfur dioxide, nitrogen oxides, ammonia and volatile organic compounds, which contribute to acidification, eutrophication and ground-level ozone. The multi-pollutant/multi-effect methodology responded to the need to consider possible side benefits of measures, which is essential in the design of cost-effective control strategies. The concept made it possible to analyze the multiple environmental impacts and interactions of emission controls of the pollutants considered.

The multi-pollutant/multi-effect concept was applied for the UN/ECE Gothenburg Protocol (*e.g.* Amann *et al*. 1999b) and for determining the national emission reduction obligations of the National Emission Ceilings (NEC) Directive proposed by the European Commission (Amann *et al*. 1998–1999). In these processes, the RAINS model was used to find cost-optimal emission control allocations for reducing simultaneously acidification, eutrophication and ground-level ozone according to predetermined environmental targets.

1.4 New modeling challenges

The recognized scale of problems caused by emissions from fossil fuel utilization has grown from local and regional to global due to the long-range and hemispheric transport of air pollutants and their impact on climate (*e.g.* Alcamo *et al*. 1998, Jonson *et al*. 2001, Posch *et al.* 1996). As the emissions of CO_2 and acidifying, eutrophying and ozone-forming compounds originate to a large extent from the same source, the exploitation of fossil fuels, also significant synergies between reducing regional air pollution and combating global warming could be expected. Structural changes in energy production have been largely neglected in air pollution control strategies, or at best treated as sensitivity analysis, and air pollution strategies have been solely based on technical control possibilities ('end-of-pipe' technologies). As most planned CO_2 emission reduction measures affect also acidifying, eutrophying and ozone-forming emissions, the UN/FCCC process is bringing new aspects into the air pollution control strategy development. Conversely, regional air quality issues should be considered in the design of greenhouse gas abatement strategies to ensure maximum benefits of planned measures. However, one has also to be aware that regional air quality improvements reduce the amount of sulfate aerosols, which are recognized to have a cooling effect on the climate (IPCC 1996).

The present integrated assessment models operate at a relatively coarse spatial resolution due to the large geographical area in concern and the related extensive data needs. The continent-scale models are neither designed nor suitable for analyzing regional or local problems. Regional models are needed to provide accurate information about the effects of alternative local emission control policies to support the design of regional environmental policies, and thus to complement the international analysis. It has also been demonstrated that the coarse scale of deposition modeling and ignoring local variability in deposition can result in serious underestimates of acidification and eutrophication critical load exceedances (Hirst *et al.* 2000). In addition, critical load is a static concept, which is not able to represent time-dependent processes or ecosystem responses to changing acidifying deposition. To this end, dynamic modeling of the acidification processes is needed.

The integrated assessment modeling of the ozone control strategies within the UN/ECE and the EU has been done utilizing the results of a coarse scale model intended for estimating rural background ozone levels in Europe (Heyes e*t al.* 1996, Schöpp *et al.* 1999). However, small-scale phenomena in urban areas can significantly change ozone levels from those of the surroundings. The 'rural background' ozone as calculated by the present integrated assessment models tends to overestimate ambient levels in urban areas, and underestimate ozone concentrations prevailing in urban plumes or in cities with a closed geographical location. As most of the European population lives in cities and suburban areas, the modeling of population exposure to ozone is subject to considerable uncertainties. Accurate modeling of these small-scale and short-term effects posing risk to human health is a demanding task and requires detailed atmospheric models. The demand for high-

quality input data and computer time imposed by such models makes it impractical to use them for a continental scale scenario analysis, and simpler tools are needed for future support of further European ozone control policies.

Present international emission abatement strategies are based on calculations, where the effects of model uncertainties on national reduction requirements have not been quantitatively assessed. Yet the European policies aimed to reduce environmental problems caused by long-range transported air pollution require substantial abatement efforts. The National Emission Ceilings (NEC) directive of the EU, for instance, has been estimated to cost about 7.5 billion (10^9) € annually (Amann *et al*. 1998–99) on top of implementing the present legislation assuming a conventional energy pathway with increased fossil fuel use. This is because the cheap emission control potential is to a large extent exhausted in many western European countries (*e.g*. Schärer 1995, Karvosenoja *et al.* 2001). For many countries achieving the emission ceilings will require costly abatement installations with high unit reduction costs. The emission ceilings and related reduction costs can change considerably with relatively small changes in deposition targets or other underlying assumptions. The significant economic consequences stress the importance of knowing the reliability and limitations of the integrated assessment models used in support of the decision-making process. This calls for information about the confidence limits of the models and a probabilistic approach, where emission reductions and their costs can be weighed against the (changes in the) probability of ecosystem or population protection from adverse effects.

1.5 Objectives and structure of this study

This study aims at developing modeling tools for improving the spatial representation of integrated assessment models of long-range air pollution. The main objectives are the development of regional scale nitrogen deposition modeling in Finland and the development of a method for estimating the impacts of urban NO_x emissions on local ozone levels, which could be used to improve integrated assessment modeling of ozone at European scale. The second objective is the applications of integrated assessment models supporting environmental decision-making, including studies of side benefits of CO_2 control strategies on air pollution reduction. The third aim of this study is to assess and quantify the uncertainties in the integrated assessment modeling for Finland and the effects of the uncertainties on ecosystem protection estimates. The study targets regional, national and European scales.

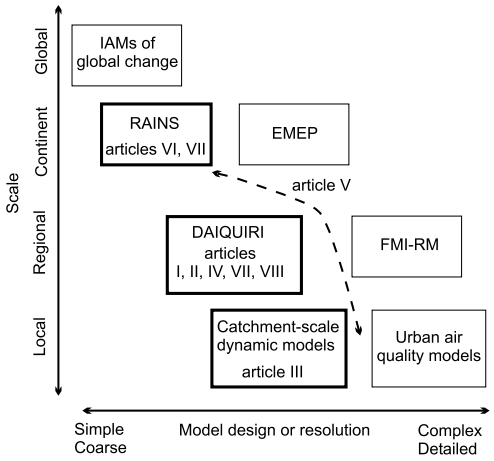

Fig. 3. Features of models of air pollution, global change and related environmental impacts designed for different purposes. The parts addressed in this work are drawn in bold and the relevant articles mentioned.

In air pollution modeling, the terms ‘local’ and ‘regional’ are often used to describe quite different scales, depending on the background of the speaker. In this work, local scale refers to distances from about one kilometer to some tens of kilometers, and regional scale, *i.e*. meso-scale, encompasses distances from tens of kilometers to some hundreds of kilometers.

Fig. 3 illustrates the features of some air pollution and integrated assessment models for different purposes with respect to their modeling domain (vertical axis) and resolution or the complexity of the system representation (horizontal axis). The parts addressed in this work are marked with bold in the Figure, with respective articles denoted.

Integrated assessment models of global change (*e.g.* Alcamo *et al.* 1998) usually operate with a coarse spatial resolution and simplified systems (population, energy-industry-emissions, terrestrial environment, etc.) descriptions due to their broad modeling scope and domain. Integrated assess-

ment models of air pollution designed for international policy development (*e.g*. RAINS) address one subset of the global change issue at one continent or region and thus are able to describe the relevant processes in more detail (*e.g*. at country-level). The EMEP models (EMEP 1998a, b), in turn, are examples of long-range air pollution models operating at European scale and containing detailed descriptions of atmospheric processes, and their results are used in integrated assessment models. The regional air quality model FMI-RM (see section 2) calculates air quality and deposition in Southern and Central Finland with a fivefold horizontal resolution compared to the EMEP models. The results of the FMI-RM are used in the regional deposition model DAIQUIRI for integrated assessment purposes developed in this work (see section 2). Catchment-scale dynamic acidification models (*e.g.* Posch *et al*. 1993) describe the ecosystem processes in more detail than the critical loads used in integrated assessment models, but they are usually used for smaller areas, and they lack the other components of integrated assessment models. Urban air quality models (*e.g*. Moussiopoulos *et al.* 2000, Brücher *et al*. 2000, Karppinen *et al.* 2000) are a further step down from the regional air quality models, operating at levels from individual street canyons to grid resolutions in the order of a kilometer.

This thesis consists of the following parts:

– development, validation and applications of a regional scale nitrogen deposition model DAIQUIRI for integrated assessment purposes in Finland (articles I, II, III, IV,VIII)
– development of an integrated assessment modeling methodology of long-term population exposure to ozone with improved accuracy (article V)
– analysis of the side benefits of low-CO_2 energy pathways in controlling acidification and ozone-formation at European (article VI) and national scales (article VIII)
– model applications supporting national and international policy-making and identifying the problem characteristics (articles III, IV,VI,VII, VIII)
– uncertainty analysis of acidification integrated assessment modeling in Finland and development of a probabilistic modeling method (article VII)

Chapter 2 of this study presents the development and validation of the regional scale nitrogen deposition module for integrated assessment purposes in Finland. The validation results are discussed and areas for further research are pointed out. In Chapter 3, a method for estimating the impacts of local NO_x emissions on urban and suburban ozone levels is presented and tested. This part of the work aimed at finding methods for improving the representation of population exposure to ozone in the European integrated assessment modeling of ozone control strategies. The applicability of the method and further research priorities are discussed. Chapter 4 describes a model study investigating the possible impacts of the EU climate change policy on recent EU air quality strategies. The cost-savings potential in air pollution control achieved with CO_2 abatement measures is analyzed and the findings are discussed. Chapter 5 presents national and local scale case studies partly arisen from the needs of policy-making. National-level assessment of future acidification is presented, and the impacts of alternative domestic and international technical reduction measures are compared with the plausible side-benefits of implementing the Kyoto protocol in the EU. Section 5.4 describes, how the emission and deposition scenarios can be utilized in dynamic acidification modeling to assess the impacts of alternative European emission reduction pathways on ecosystem recovery from acidification at catchment level. Chapter 6 presents an uncertainty analysis of acidification integrated assessment modeling in Finland. The derivation of the uncertainty estimates is shown and the results are linked to the international reduction strategy development by illustrating the differences in the ecosystem protection estimates between the probabilistic method developed in this work and the deterministic approach used in international policy-making.

During the course of this study, input data were updated and extended as new information became available. Therefore, some figures presented in this summary are updates of originals in the articles I–VIII.

2 Modeling nitrogen deposition at regional and local scales

In the control of acidifying deposition, the compounds of nitrogen are gaining increasing attention, as sulfur emissions have been reduced considerably in Europe during the 1980s and 1990s

(EMEP/MSC-W 1999). As nitrogen deposition also causes eutrophication of soils and surface waters, thereby altering the species composition of the ecosystems, the need for nitrogen emission reductions is emphasized.

Continent-scale long-range transport models of acidifying pollutants used in integrated assessments in Europe are neither designed nor suitable for analyzing local effects of individual emission sources or impacts of national emission control policies. Especially ammonia deposits in considerable amounts also within short distances. Regional models are needed to estimate properly the deposition caused by domestic emissions and the effects of local emission control options, thereby supporting the design of regional environmental policies and complementing continent-scale analyses.

2.1 The regional deposition model DAIQUIRI

Air quality models require detailed input data and significant computing resources. Their applicability in integrated assessment models is limited, and thus simplified descriptions of depositions have been developed from their results for use in integrated assessment. Instead of running the very data- and computation-intensive original meteorological and chemical models to estimate the depositions of alternative emission scenarios, only long-term average results (from one year to several years) are utilized in integrated assessment models. So-called transfer or transport matrices are used to describe long-term deposition patterns caused by an emission source or an emitter area. At European scale, transfer matrices calculated from the results of the EMEP ROOT150 model (EMEP/MSC-W 1998a) have been widely used in the integrated assessment modeling of European emission reduction strategies. The modeling domain covers Europe with a grid resolution of 150 km × 150 km, and the matrices describe the deposition in every grid cell of the domain due to emissions in each European country.

Within this work, a regional deposition model called DAIQUIRI (Deposition, AIr QUality and Integrated Regional Information) for integrated assessment purposes in Finland was developed (articles I, II). In DAIQUIRI, regional sulfur and nitrogen transport matrices for estimating deposition caused by Finnish emissions are linked with the long-range transport matrices of the EMEP model. The regional transport matrices for sulfur have been developed earlier at the Finnish Meteorological Institute (FMI) and used in acidification research in the late 1980s and throughout the 1990s (*e.g.* Johansson *et al.* 1990). In this work, regional matrices for nitrogen oxides and ammonia were developed using the regional air quality model (FMI-RM) of the FMI (Hongisto 1992). The matrices describing annual average transport of nitrogen oxide and ammonia emissions in Finnish conditions calculated with the FMI-RM were combined with the existing regional sulfur matrices and with the EMEP long-range transport matrices in the DAIQUIRI model. In addition, a module for calculating point-wise deposition time series for the needs of dynamic acidification modeling was included in DAIQUIRI (article III, Forsius *et al.* 1997). The meso-scale nitrogen transfer matrices have a resolution of 30 km × 30 km. Deposition fields calculated both with the meso-scale matrices and with the EMEP long-range transfer matrices are interpolated bilinearly to the DAIQUIRI grid of $^1/_4° \times {}^1/_8°$, which is about 14 km × 14 km in Southern Finland.

With DAIQUIRI total nitrogen deposition is obtained by adding the deposition fields caused by all individual sources in Finland and including the long-range transported deposition from all other countries using the EMEP transfer matrices. This construction allows a fast computation of deposition patterns for various national and international emission reduction scenarios, analogously to earlier integrated models of acidification (*e.g.* Johansson *et al*. 1990).

2.2 Development and validation of the regional nitrogen deposition module

Regional nitrogen transfer matrices developed in this work were calculated at the FMI from the results of the regional air quality model FMI-RM (Hongisto 1992). FMI-RM is a Eulerian grid model, in which the advection, diffusion, chemical conversion and deposition of nitrogen compounds are calculated by solving numerically a set of partial differential equations. The model has seven vertical layers of variable thickness, extending from ground to a maximum height of 2500 m. The horizontal grid size is 30 km × 30 km, and the original calculation area covers Southern and Central

Finland with a total calculation area of 600 km × 600 km.

Matrices were generated at the FMI for the years 1990, 1993 and 1995 for Southern and Northern Finland (article I, Kangas and Syri 2001). The transfer matrices cover a domain equal to the FMI-RM calculation area, *i.e.* 600 km × 600 km. Every cell of each regional transfer matrix describes the annual amount deposited to the cell from a unit source located in the center of the matrix. For nitrogen oxides, matrices for three emission height classes were calculated. As practically all ammonia emissions originate from activities at ground level (*i.e.* agriculture), one height class was considered sufficient for ammonia. In addition, separate matrices were generated by polynomial fitting and extrapolation to describe the approximate deposition outside the matrix ranges, up to 900 km from the source.

A validation of DAIQUIRI was carried out for the year 1990 (article I). The aim of the validation was to identify how well DAIQUIRI can (i) reproduce the deposition fields of the more accurate model FMI-RM, and (ii) follow the annual depositions measured at background sites throughout Finland. Based on the results, the suitability and accuracy of DAIQUIRI nitrogen modules with respect to integrated assessment applications could be judged. Deposition fields resulting from Finnish emissions as calculated with FMI-RM and DAIQUIRI were compared and found similar especially in forested inland areas. DAIQUIRI deposition estimates were also compared with annual bulk deposition values from the background monitoring stations of the FMI (Leinonen and Juntto 1991). The stations belong to different measurement networks, and precipitation sampling time ranges from a day to a week. Fig. 4 shows the locations of the stations used in the comparison. The stations Haapasaari and Sotkamo were excluded from the comparison for 1990, as the temporal coverage of the measurements was less than 75 %.

The correspondence between the measurements and DAIQUIRI estimates was better for deposition of oxidized nitrogen (NO_y) than for reduced nitrogen (NH_z). One reason for this was that some of the stations were located in the vicinity of considerable agricultural activities causing local-scale deposition not captured with the regional-scale model. Overall, the correlation coefficients calculated for ten available inland stations (r^2=0.92 for NO_y and r^2=0.76 for NH_z) indicated a good correlation between DAIQUIRI estimates and the measurements at all deposition levels in 1990. The linear coefficient was greater than one, which is in accordance with the fact that DAIQUIRI calculates total deposition, whereas the bulk measurements contain only a fraction of dry deposition.

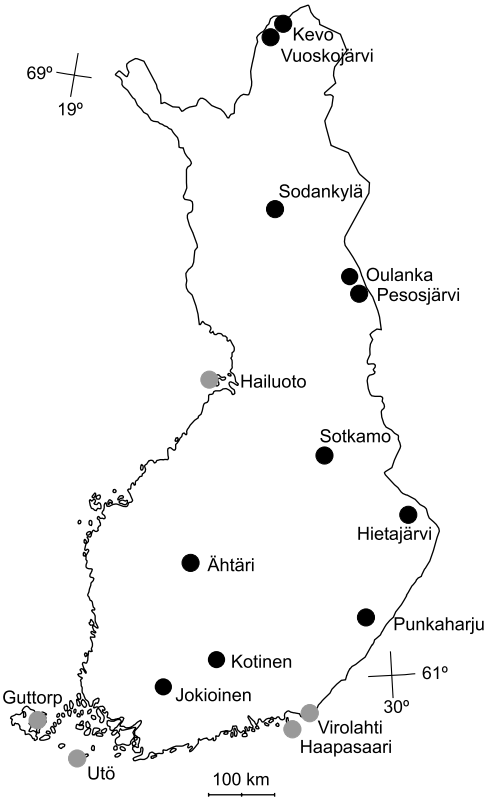

Fig. 4. Locations of the FMI background monitoring stations used in the DAIQUIRI validation. Inland stations are shown as black dots and coastal stations as gray dots.

In the second phase of validation, comparisons for the years 1993 and 1995 were carried out (Kangas and Syri 2001). Fig. 5 shows depositions in 1995 from emissions within the southern modeling domain for (a) oxidized and (b) reduced nitrogen calculated with FMI-RM (left) and DAIQUIRI (right). The borders of the FMI-RM and DAIQUIRI modeling domains are drawn in Fig. 5(a). The results of both models were interpolated bilinearly to the DAIQUIRI grid of $^1/_4° \times {}^1/_8°$.

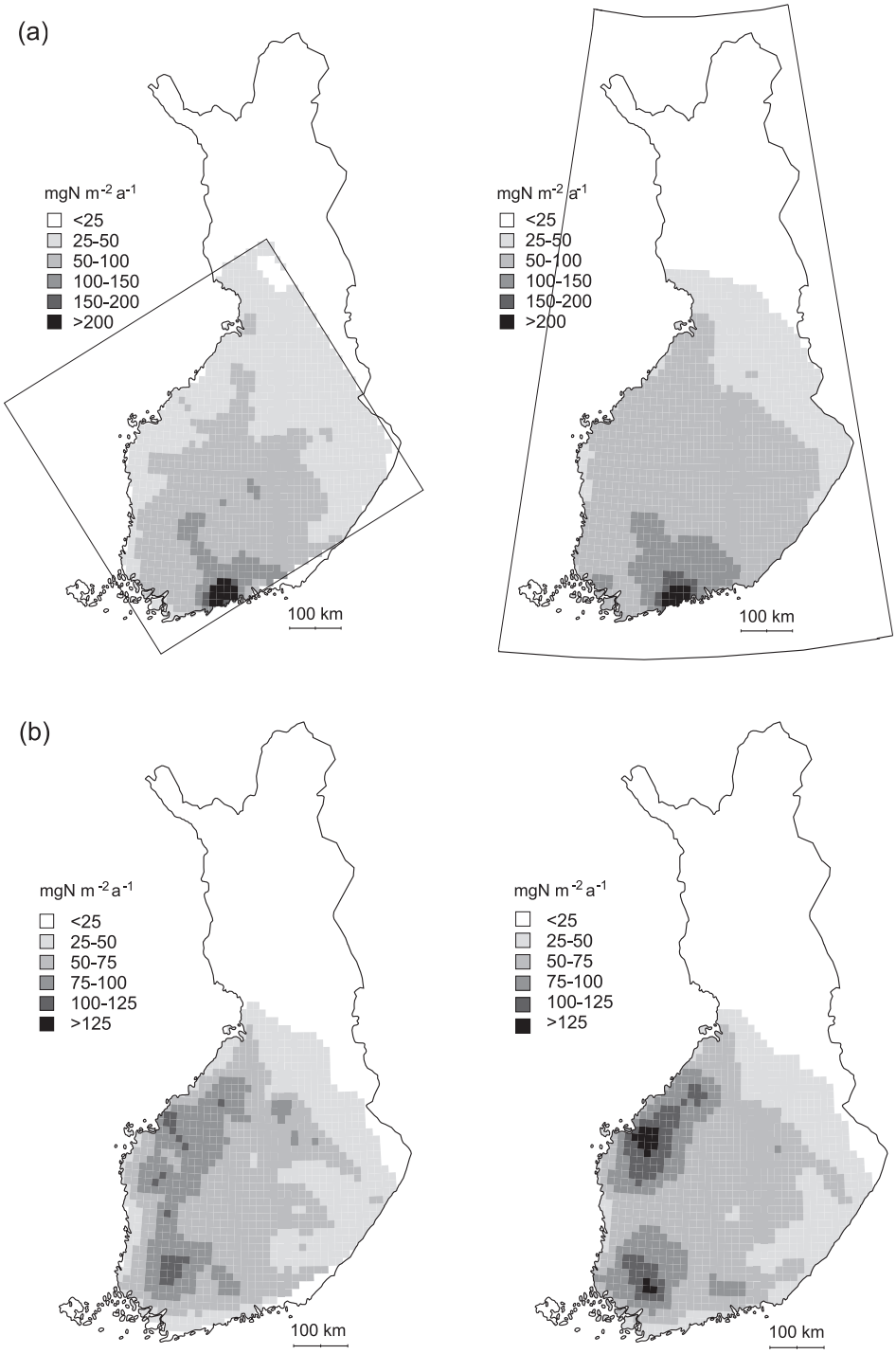

Fig. 5. Deposition fields from emissions within the FMI-RM calculation grid in 1995 for (a) oxidized and (b) reduced nitrogen calculated with FMI-RM (left) and DAIQUIRI (right). The modeling domains are drawn in (a).

Fig. 5 shows that the level and the general patterns of indigenous deposition calculated with the FMI-RM were quite well reproduced with DAIQUIRI especially in forested inland areas, which are the main point of interest in applications for acidification research. Compared to FMI-RM, DAIQUIRI overestimated the depositions during all years investigated in areas where other surface types, mainly fields or waters, dominated the land use. This is explained by the fact that in the generation of the transfer matrices, actual surface types in the FMI-RM calculation area were used, and the dominating land use type in the vicinity of the emission source in the center of the matrix was forest. For forests the dry deposition velocities of gaseous nitrogen compounds are higher than for fields, grasslands and waters, thus resulting in higher dry deposition amounts (Seinfeld and Pandis 1998).

In addition to validation carried out in (article I), deposition estimates calculated with DAIQUIRI for the years 1993 and 1995 were compared against annual deposition measurements from FMI stations (Leinonen 1994, 1997). In this exercise, also separate matrices for wet and dry fractions of deposition were available from the FMI-RM calculations and from EMEP (Kangas and Syri 2001). Fig. 6 shows a comparison of measurements and DAIQUIRI estimates of (a) oxidized and (b) reduced nitrogen for 1995, indicating the modeled shares of wet and dry deposition and the error ranges of the measurement analyses (Leinonen 1999).

It can be seen that especially at inland stations DAIQUIRI predicts higher total depositions than the measurements, which collect wet deposition from precipitation and include only a fraction of dry deposition. At inland stations the modeled wet depositions were close to the measured values. The Punkaharju station is located on an isthmus between two lakes, and the modeled overestimation of deposition may be due to local meteorological variations, which can not be captured with the transfer matrix concept. The stations Virolahti and Haapasaari show interestingly the effects of coastal conditions and for ammonia the impact of nearby agricultural emissions as well. Haapasaari is located on an island, and Virolahti is some tens of kilometers northeast of Haapasaari, close to the coast and near agricultural activities. The local ammonia emissions cause a deposition peak not captured with the regional scale model. Also the measured NO_y deposition is considerably higher at Virolahti. The differences in NO_y deposition probably reflect the coastal meteorological effects not captured by the regional models.

At all coastal stations of the Gulf of Finland the correspondence between measurements and model results was found considerably poorer than at inland stations, especially for ammonia during all years considered in the validation. At the coastal stations the discrepancies between model results and measurements reflect the impact of several factors. Firstly, the calculated values South of Finland consist mainly of long-range transported deposition, which is estimated with the EMEP model's transfer matrices. In addition, both the EMEP model and FMI-RM can only partially represent the complex transport and deposition processes over coastal areas. High measured depositions in relation to model results along the South coast of Finland can also be a reflection of underestimated emissions in the Baltic countries and in the St. Petersburg region used in the models. At coastal stations also measurement uncertainties can be considerably larger than in inland regions as the evaporation caused by wind may lead to overestimated deposition amounts.

The ability of DAIQUIRI to estimate both wet and dry fractions of deposition enables a better evaluation of differences between modeled depositions and measurements. In this work, modeled wet depositions away from coastal areas were in general close to the monitored depositions, indicating that the share of dry deposition present in measurements would not be large. The inclusion of wet and dry fractions of deposition in DAIQUIRI is directed also to facilitate dynamic modeling, where modeled long-term deposition time series are usually calibrated to site-specific monitored values, which are typically bulk measurements (article III).

The ability of DAIQUIRI to reproduce observed deposition trends was evaluated for 1987–1995 in (article II), where DAIQUIRI was utilized to indicate the origin of observed deposition trends at background monitoring stations belonging to the network of the UN/ECE Integrated Monitoring Programme (Kleemola and Forsius 1997). In (article II), the regional nitrogen transfer matrices estimating total deposition averaged over the years 1990 and 1993 were applied, as the matrices for 1995 and wet and dry fractions of deposition were not yet available. Finnish nitrogen emission data

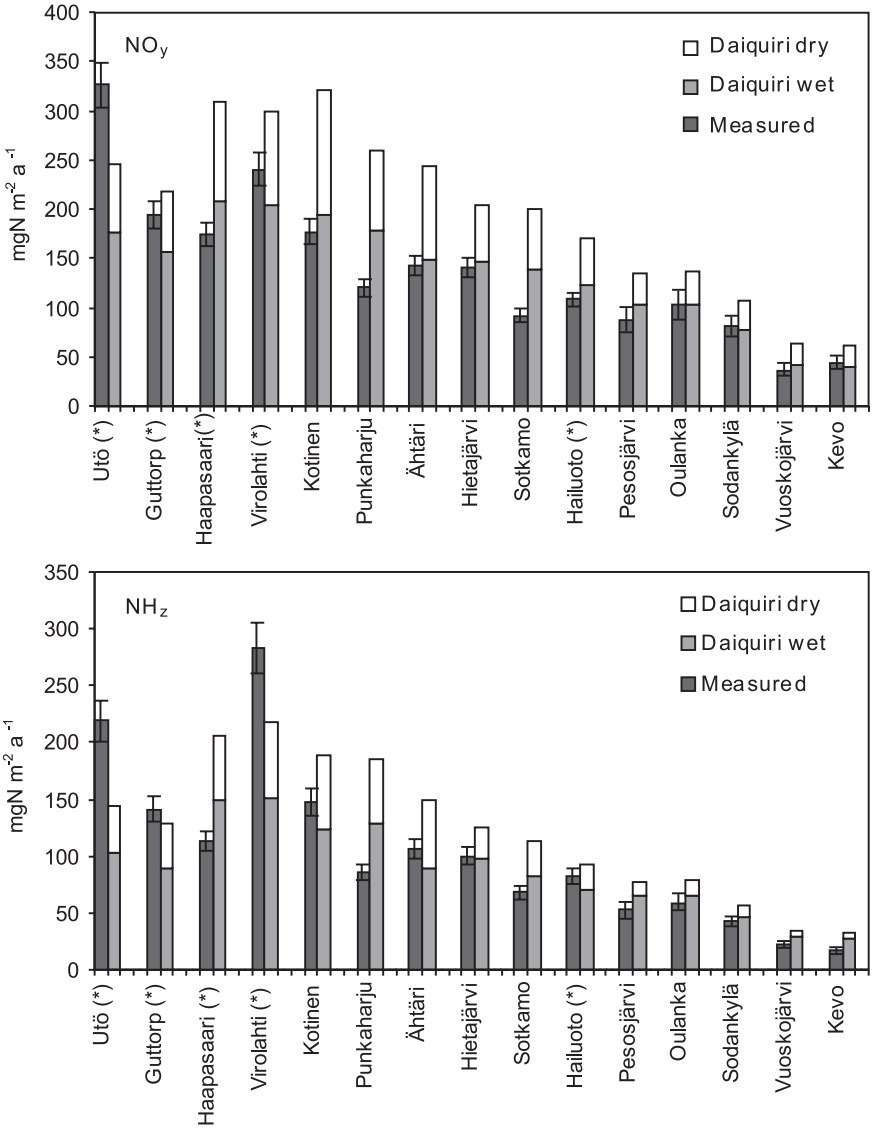

Fig. 6. Measured and modeled deposition of (a) oxidized and (b) reduced nitrogen in 1995. Both the wet and dry shares of the modeled deposition are shown. Coastal stations are marked with an asterisk (*). The measurement analysis error ranges are indicated with vertical bars.

were available from CORINAIR and other national inventories (Melanen and Ekqvist 1997). At all the monitoring stations the main part of nitrogen deposition was long-range transported, which was estimated with the EMEP transfer matrices.

Modeled long-term trends were found similar to those indicated by the measurements. Individual years' depositions were not always reproduced, which is probably mainly due to local meteorological factors not captured with the regional and long-range transport matrices averaged over several years. The interannual variations in measured deposition were the largest for ammonia, and they were only partially followed by the model.

In (article II), DAIQUIRI provided information about the origins of observed trends in deposition, which could not have been derived using only measurements and information about emission changes in European countries. It was found that the slight downward trend in NO_y deposition in Finland was mainly due to emission reductions abroad, as NO_y deposition in Finland is dominated by imported deposition and during 1987–1994 the Finnish NO_x emissions stayed almost constant. Measurements implied a 10 to 20 % decrease in NH_z deposition during the study period. DAIQUIRI runs indicated that this was partly due to decreases in Finnish emissions and partly caused by reductions outside Finland. At Kotinen in Southern Finland the model runs showed that reduction in domestic deposition has been significant in the decreasing trend. In the case of ammonia, however, uncertainties both in emission inventories and in deposition measurements are considerable.

In the remote areas of Northern Finland the share of domestic origin in nitrogen deposition was found negligible. This implies that regional modeling of nitrogen deposition is needed primarily in Southern and Central Finland affected by significant local emissions. There regional modeling can give more precise information about the effects of domestic emissions and about the impacts of national emission reductions in relation to international measures than available from continent-scale models alone.

2.3 Effects on environmental indicators

The impacts of regional nitrogen deposition modeling on the estimates of atmospheric loading and on the exceedances of acidity critical loads were calculated to quantify the significance of the indigenous deposition modeling for environmental assessment. The results of the regional nitrogen deposition module with detailed point-source emission data as input (Melanen and Ekqvist 1997) were compared with the estimates of the EMEP long-range transfer matrices, which utilize EMEP emission data at 150 km resolution (EMEP/MSC-W 1998).

Fig. 7 illustrates the difference in (a) NO_y and (b) NH_z deposition estimates in 1995 between the regional modeling of DAIQUIRI and the deposition fields obtained with the EMEP transfer matrices. The comparison was done for the EMEP deposition fields interpolated to the $^1/_4° \times {}^1/_8°$ grid of DAIQUIRI. The dark shadings show areas where DAIQUIRI gives higher deposition estimates, and the two light shadings illustrate regions where the EMEP transfer matrices give higher estimates. A maximum difference of about 240 mgN m^{-2} a^{-1} in NO_y deposition fields from Finnish sources was found in the Helsinki region, where the maximum emission density occurs in Finland. For NH_z, DAIQUIRI estimated at maximum about 70 mgN m^{-2} a^{-1} more indigenous deposition than the EMEP model in the agricultural and fur production areas of Ostrobothnia.

The model estimates of total masses deposited to Southern and Central Finland (confined by the intersection of FMI-RM and DAIQUIRI calculation grids, shown in Fig. 5) from sources located within the area were compared. Table 1 illustrates the differences between the estimates of FMI-RM, DAIQUIRI and the EMEP transfer matrices. The EMEP results include deposition from all Finnish sources, but practically all the Finnish nitrogen emissions are in the calculation area used in the comparison, and emissions from Northern Finland are mainly transported towards North East.

Table 1 shows that the more accurate representation of nearby deposition from NO_x emission sources in Southern and Central Finland gives larger estimates of the resulting loading. Both FMI-RM and DAIQUIRI arrive at considerably larger total NO_y deposition than the EMEP model. DAIQUIRI overestimates both NO_y and NH_z deposition in relation to FMI-RM especially in areas dominated by fields or waters. The calculation area in Table 1 includes the sea regions near the Finnish West Coast, which partly causes the overestimated total deposited amounts in comparison to FMI-RM. For forested areas, the deposited masses of DAIQUIRI and FMI-RM results agree better. Compared to FMI-RM, DAIQUIRI overestimates the total indigenous NO_y deposition by 18 % in 1993 and by 33 % in 1995, and ammonia

Table 1. Deposition from Finnish sources (ktN a^{-1}).

	NO_y		NH_z	
	1993	1995	1993	1995
EMEP	11.0	10.3	16.6	15.3
FMI-RM	14.6	14.6	16.2	13.6
DAIQUIRI	17.3	19.4	20.8	15.1

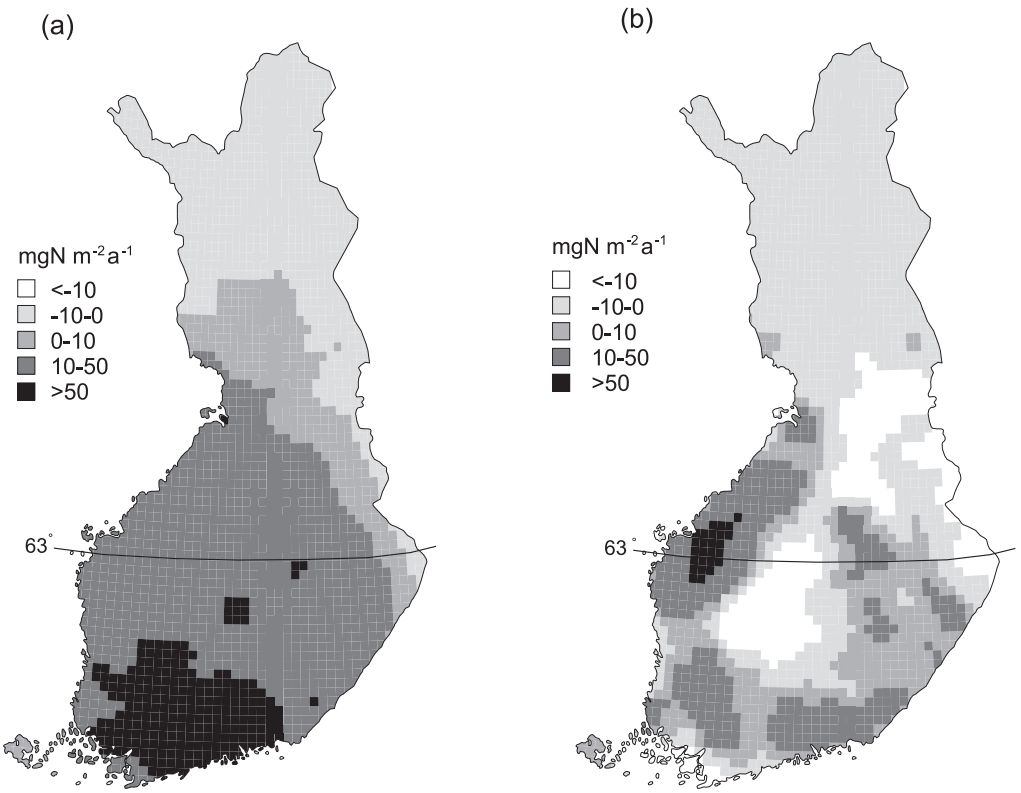

Fig. 7. The difference in deposition between the meso-scale modeling of DAIQUIRI, using Finnish point-source emission data, and deposition calculated with the EMEP transfer matrices and EMEP emission data for Finland for (a) NO_y and (b) NH_z in 1995.

deposition by 28 % and 11 % in 1993 and 1995, respectively. However, for inland areas the relative overestimation is considerably smaller.

The Lagrangian EMEP model assumes an instantaneous mixing of nitrogen emissions to the atmosphere up to the mixing height and estimates only the average deposition to grid cells of 150 km × 150 km. Thus it is not able to portray the in-grid variation of deposition, and the local in-grid deposition depends largely on the local correction factor (Seland *et al*. 1995). Both FMI-RM and DAIQUIRI calculate considerably more NO_y deposition than the EMEP model. For ammonia, the Lagrangian EMEP transfer matrices used indicate bigger domestic deposition to the calculation area used here than the Eulerian model FMI-RM. These results are in accordance with Bartnicki (1999), who compared the Lagrangian and new Eulerian long-range transfer matrices of EMEP. He found that the Eulerian modeling concept estimates more indigenous NO_y than the Lagrangian model, and for domestic NH_z deposition the results were the opposite. However, in our application the EMEP model overestimated NH_z deposition to coastal areas and far from local sources compared to the FMI-RM and DAIQUIRI, but significantly underestimated NH_z deposition close to the sources.

Table 2 shows the ecosystem areas in Finland with exceedance of acidity critical loads in 1995 and in 2010, assuming the implementation of the UN/ECE Gothenburg protocol. Nitrogen deposition from Finnish sources was calculated using the nitrogen transfer matrices averaged from the years 1990, 1993 and 1995. Long-range deposition was estimated with the long-term average EMEP transfer matrices (EMEP/MSC-W 1998a). For comparison, critical load exceedances estimated using

Table 2. The share of total ecosystem area with acidification critical load exceedance in Finland in 1995 and in 2010 (Gothenburg protocol). Nitrogen deposition from Finnish sources is calculated either with the average meso-scale matrices or with the long-term average EMEP transfer matrices.

	DAIQUIRI % of area	EMEP % of area	Difference as $\frac{\text{DAIQUIRI}}{\text{EMEP}} \cdot 100\ \%$
1995			
Finland	5.39	4.96	109 %
<63°	6.94	5.83	119 %
2010			
Finland	3.56	3.51	101 %
<63°	4.24	3.92	108 %

only the EMEP transfer matrices are shown as well. The comparison was done both for all ecosystem areas in Finland and for ecosystems located in Southern Finland only (South of 63° latitude), where the majority of Finnish acidifying emissions is located. Sulfur deposition from domestic sources and from nearby sources in Estonia and Russia was calculated using the existing meso-scale module for sulfur (Johansson *et al.* 1990) and the long-range transported deposition was estimated with the EMEP/MSC-W matrices.

Table 2 demonstrates that the higher calculated domestic inland nitrogen depositions calculated with the meso-scale module result in more pessimistic estimates of the areas exceeded than when using the EMEP model only. The differences are significant, taken into account that most of the deposition of acidifying compounds is imported and that sulfur deposition, which is in the same order as nitrogen deposition in Finland, was not varied in the comparison. Table 2 corroborates that meso-scale modeling has significance in areas near emissions. The differences in the estimates of areas at risk are more pronounced if only the ecosystem areas South of 63° latitude are considered. With the expected future decrease of acidifying deposition, the differences in critical load exceedances caused by meso-scale modeling will be less pronounced.

2.4 Concluding remarks

The regional deposition model DAIQUIRI was found to perform well in estimating long-term nitrogen deposition in forest-dominated areas. For areas dominated by other surface types and in coastal areas, DAIQUIRI overestimated nitrogen deposition, which illustrates the potential problems induced by the movable transfer matrix concept. However, the problems related to model representation of agricultural and coastal areas are of minor importance in the model application for acidification assessments, as the ecosystems of primary interest are forests and lake catchments usually covered mainly by forests.

Based on comparisons with measurements, DAIQUIRI was able to represent the long-term depositions with satisfactory accuracy. Individual years' depositions were not reproduced at all sites, but long-term trends and the average level of deposition were comparable with the monitored deposition levels and trends. These together with the origin of deposition are the relevant parameters in the integrated assessment modeling of acidification and eutrophication.

The regional nitrogen deposition modeling with DAIQUIRI has provided more precise information about acidifying and eutrophying deposition from Finnish emission sources than what is possible with coarse-scale long-range transport models. The differences against the continent-scale modeling used in international control strategy development were significant in Southern and Central Finland. The regional deposition modeling indicated considerably higher NO_y deposition from local sources in Southern Finland than the Lagrangian EMEP model with 150 km resolution, which was also reflected in the estimates of acidity critical load exceedances. For ammonia, the average levels of indigenous deposition were comparable, but regional modeling with DAIQUIRI was able to represent the spatial patterns and the impacts of local sources in deposition in more detail. Thus it is important to estimate domestic deposition with regional or local scale models in order not to underestimate or mislocate the impacts of domestic emissions. The computationally simple technical structure of DAIQUIRI facilitates various applications and scenario studies in support of environmental policy development (*e.g.* article IV, Forsius *et al.* 1997, Lepistö and Syri 2001).

Estimation of nitrogen deposition at coastal areas is fraught with considerable uncertainties, as was demonstrated by model-measurement comparisons and model intercomparisons in this work. More research should be devoted to estimating ni-

trogen deposition to waters and coastal areas more reliably in order to facilitate the assessment of effective control strategies against eutrophication of surface waters.

3 Modeling urban ozone patterns for European control strategies

The previous section described how regional nitrogen deposition models can be developed and used together with spatially detailed emission inventories for improved estimation accuracy in support of environmental policy development at national and sub-national level. Correspondingly, tools for linking the results of large-scale ozone models used in international control strategy development and local phenomena can be developed for efficient local and national control policy development, utilizing local information about NO_x levels for better representation of local effects in ozone formation and destruction.

The integrated assessment modeling of ozone control strategies for the UN/ECE and the EU has been done by deriving a simple statistical model from the Lagrangian-type photochemical oxidant model of EMEP (Heyes e*t al.* 1996, Schöpp *et al.* 1999). The EMEP photochemical model calculates photochemical reactions and transport of air pollutants in a single atmospheric layer with a horizontal resolution of 150 km × 150 km, applying a chemical scheme with about 70 species and 140 reactions (Andersson-Sköld and Simpson 1999). Ground-level ozone concentrations throughout Europe are computed with a 6-hour time-step, at 0, 6, 12 and 18 GMT. Due to its coarse spatial resolution, the model is mainly designed for estimating ozone values in rural background areas over long periods of time (month – year), and it is not suited for describing phenomena occurring in urban areas.

However, small-scale phenomena in urban areas can significantly change ozone levels from those of the surroundings. The 'rural background' ozone, as calculated by the present integrated assessment models, tends to overestimate ambient levels in urban areas, and underestimate ozone concentrations prevailing in urban plumes or in cities with a closed geographical location. As most of the European population lives in cities and suburban areas, the modeling of population exposure to ozone is subject to considerable uncertainties. In this work, a method for estimating the impacts of local NO_x emissions on urban and sub-urban ozone levels was developed and tested (article V).

3.1 Representing the effect of local NO_x levels on ozone concentration for large-scale models

This study concentrated on representing the 'titration effect', *i.e.* the destruction of ozone by fresh NO emissions in urban areas, for future use in integrated assessment modeling of ozone control strategies. The key questions were to what extent titration can explain differences between rural background and observed ozone levels in urban areas, and how small-scale information on urban NO_x levels could be used to improve the accuracy of population exposure estimates derived from large-scale modeling or from local good-quality monitoring networks. The effect of urban NO_x concentrations on ozone levels was derived from the chemical equations describing the ozone balance (article V).

Ambient levels of ozone are a result of complex chemistry with several thousands of known chemical reactions (Seinfeld and Pandis 1998, Colbeck and McKenzie 1994). Essentially, increased ozone levels occur in the presence of NO_x, VOC and sunlight. An ozone molecule is produced through the recombination of molecular oxygen (O_2) and atomic oxygen (O). In the presence of solar energy, hν, the NO_2 molecule may dissociate into NO and O as follows:

$$NO_2 + h\nu \rightarrow NO + O \qquad (R1)$$

Then the recombination of O and O_2 will occur:

$$O + O_2 + M \rightarrow O_3 + M \qquad (R2)$$

where M is any inert molecule. On the other hand, a NO molecule may undergo a rapid reaction with an ozone molecule:

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (R3)$$

The balance of these reactions is dependent on the sunlight and the NO_x available. In the presence of VOC, there may occur series of chemical processes, which will convert NO to NO_2 and therefore reduce the possibility for the reaction R3, resulting in a net ozone production.

If there is initially a lot of NO in relation to NO_2, reaction R3 will be more dominant. A new

equilibrium state with decreased ozone concentration will emerge. This is often referred to as the titration of ozone. Titration is independent of sunlight and therefore reduces ozone concentrations also during nighttime together with the deposition to ground. Nitrogen oxides are mainly emitted as NO, most of which oxidizes in the atmosphere within a few minutes to form NO_2. Thus in the immediate surroundings of emission sources the share of NO in total NO_x (NO_x=NO+NO_2) is higher than further away from the sources.

In the absence of other processes to convert NO to NO_2 and assuming steady state conditions, one can derive from R1, R2 and R3:

$$[O_3] = K \cdot \frac{[NO_2]}{[NO]} \quad (1)$$

where the equilibrium constant K is the ratio of the photolysis rate coefficient (reaction R1) and the rate coefficient for reaction R3. The stoichiometric reactions of $[O_3]$ and $[NO]$ and the conservation of nitrogen imply (Seinfeld and Pandis 1998, p. 236):

$$[O_3]_0 - [O_3] = [NO]_0 - [NO] = [NO_2] - [NO_2]_0 \quad (2)$$

where the subscript 0 denotes the initial state. From (2) it follows that the sum of oxidizing compounds, $[O_x]=[O_3]+[NO_2]$ remains constant. The same holds for nitrogen oxides, *i.e.* $[NO_x]_0=[NO_x]$. Thus for an urban location, in which we assume that NO_x is injected and the new equilibrium is reached, equation (1) can be written as:

$$[O_{3,U}] = K \cdot \frac{[NO_2]_0 + [O_3]_0 - [O_{3,U}]}{[NO]_0 - [O_3]_0 + [O_{3,U}]} \quad (3)$$

where $[O_{3,U}]$ is the urban ozone concentration after the equilibration. The concentrations of nitrogen compounds can be calculated from the conservation of nitrogen by defining the initial urban NO_x concentration $[NO_{x,U}]_0$ as the sum of NO_x concentration in the surrounding rural areas $[NO_{x,R}]$ and the local emission $\Delta[NO_x]$. In this work, $[O_3]_0$ was assumed to be equal to the nearby rural ozone concentration $[O_{3,R}]$, available either from measurements or from the EMEP model. Thus eq. (3) becomes:

$$[O_{3,U}] = K \cdot \frac{[NO_{2,U}]_0 + [O_{3,R}] - [O_{3,U}]}{[NO_U]_0 - [O_{3,R}] + [O_{3,U}]} \quad (4)$$

This is a quadratic equation for $[O_{3,U}]$ with a unique positive solution. In this work, equation (4) was applied to both measured and modeled time series of rural O_3 and measured urban and rural NO_x to explore to what extent it can explain (i) differences in measured ozone concentrations between urban and rural areas, and (ii) between measurements and the large-scale predictions of the EMEP model.

In this study the *K*-factor was estimated from the available rural measurement data. For this it was assumed that the concentration of VOC at the rural (background) stations is rather small, so that one can neglect the chemical reaction of VOC and assume that the reactions R1-R3 will be in steady state for these stations. Under this assumption, equation (1) holds and K can be approximated as

$$K \approx \frac{\overline{[O_3]}.\overline{[NO]}}{\overline{[NO_2]}} \quad (5)$$

where $\overline{[NO_2]}$ and $\overline{[NO]}$ represent the average rural NO_2 and NO concentrations surrounding the urban monitoring stations, estimated from the monitored NO_x and NO_2 concentrations at the rural stations, and $\overline{[O_3]}$ designates either the average monitored rural ozone concentrations around the urban area or the EMEP model's ozone predictions.

3.2 Test application

The method was tested with NO_x and ozone measurements from both urban and rural areas in Switzerland and with the ozone predictions of the EMEP photochemical model. The study utilized EMEP model calculations for the 1994 summer period (1 April – 30 September).

In Fig. 8, calculated and measured frequency distributions at the urban station of Zürich are compared. In Fig. 8(a) measured rural background ozone was used as $[O_{3,R}]$, and in Fig. 8(b) EMEP model ozone concentrations were used as the $[O_{3,R}]$.

It can be seen from Fig. 8 that the inclusion of the titration effect improved the fit over the entire range of ozone concentrations. While the frequency of concentrations between 20 and 40 ppbv was somewhat overestimated, the approach missed some individual concentration peaks.

Table 3 shows the improvement in the correlation coefficients between the urban site measurements and rural ozone concentrations and the modeled urban concentrations. It shows that including the titration effect increased the correlation considerably at all stations and for both background ozone values used. The overall urban

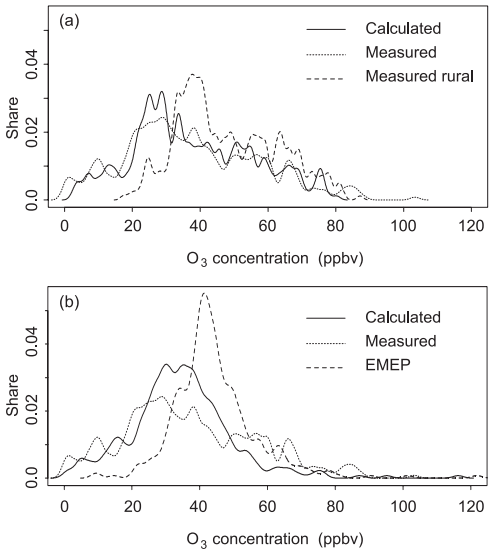

Fig. 8. Measured and calculated daytime ozone concentrations in Zürich. Dotted lines represent ozone concentrations measured at the site. (a) Solid line shows the concentrations calculated from equation (4) using the average rural ozone measurements. Dashed line designates the average rural ozone time series used as the starting point. (b) Solid line shows the concentrations calculated from equation (4) using the EMEP model ozone predictions (dashed line). NO_x concentrations used were those measured at the site.

ozone patterns were more accurate when utilizing the rural observations as the starting-point, but also the use of the large-scale model results gave satisfactory results.

The method was also tested in terms of the AOT60 index used in the development of the UN/ECE Gothenburg protocol and the EU NEC directive proposal. The AOT60 is the accumulated ozone concentration above a threshold of 60 ppbv, *i.e.*:

$$\mathrm{AOT60} = \int \max\left[0, [O_3](t) - 60\right] dt \qquad (6)$$

where the integral is taken over the whole summer period (April-September), and $[O_3](t)$ are eight-hour moving average values (six-hourly data with the EMEP model).

Fig. 9 compares the average (rural measurements or EMEP model) AOT60 values with urban site measurements and the AOT60 values derived from background concentrations using the titration method for the Basel, Härkingen, Sion and Zürich sites.

Fig. 9 shows that the titration method improved the estimate of the AOT60 at all sites. In this study, with respect to AOT60 the utilization of average rural observations was less successful than the use of modeled ozone time series as starting point. Especially for Basel, the use of rural time series gave too high results. This is probably because the rural observations used were not fully representative for the surroundings of Basel. However, when interpreting the results it should be remembered that the AOT60 is a very sensitive measure since it depends only on the upper extremes of the whole time series. A systematic error of a few percent either in the observed or in the modeled ozone concentrations can result in large deviations of the AOT60 value, as illustrated for example in EMEP/MSC-W (1998b). Therefore these comparisons in terms of AOT60 should be considered only indicative for the quality of the titration method. The results calculated with the whole data sets, as shown in Fig. 8 and Table 3, demonstrate more comprehensively the performance of the method.

Table 3. Correlation coefficients between urban measurements and surrounding average rural values (measured and modeled with EMEP) and modeled urban concentrations.

Station		Average rural O_3 measurements	Urban O_3 modeled with average rural O_3 measurements	EMEP model O_3	Urban O_3 modeled with EMEP model O_3
Urban	Basel	0.80	0.89	0.42	0.51
	Zürich	0.83	0.94	0.42	0.59
Freeway	Härkingen	0.52	0.85	0.30	0.64
	Sion	0.40	0.74	0.33	0.59
Mean value		0.64	0.85	0.37	0.58

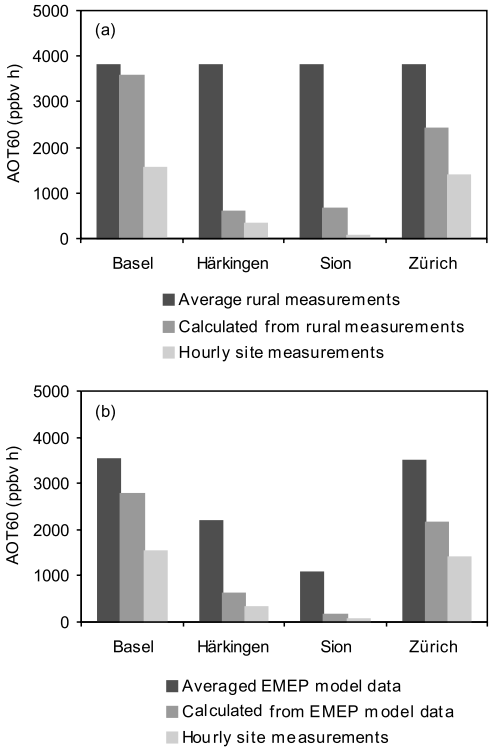

Fig. 9. AOT60 values evaluated at four urban and freeway stations. Light gray bars denote the AOT60 obtained from the hourly urban measurements. (a) Black bars show the average AOT60 obtained from the rural ozone measurements and gray bars show values obtained by the titration method using the rural ozone measurements as starting-point. (b) Black bars show the AOT60 obtained from the EMEP model and gray bars show the values obtained by the titration method using the EMEP model ozone data (averaged over the two closest grid cells for Basel, Sion and Zürich stations).

3.3 Concluding remarks

The results indicate that urban NO_x levels are a significant explanatory factor in differences between urban and surrounding rural ozone concentrations and that the urban NO titration effect could be satisfactorily represented in integrated assessment models with the method presented. The method could also be applied to improve the quality of spatial interpolation methods for observed ozone to obtain more realistic maps of ozone exposure and to improve large-scale assessments of health risks caused by the synergistic effects of ozone and NO_2 concentrations (Kley *et al.* 1999).

The estimate of the reaction coefficient in this study requires good-quality rural NO_x measurements, which are not available throughout Europe. For a wider integrated assessment modeling application the method should be further developed and tested. This could be done, for example, by calculating the reaction rates from the local radiation and temperature conditions. Further research efforts should include testing of the method in more locations and analyzing the performance of more widely applicable ways of deriving the required parameters.

In the development of ozone control strategies the co-effects of local and large-scale emission reduction measures are important to consider. Studies with coupled large- and urban-scale photochemical models have shown that local urban NO_x emission reduction measures alone can result in an increase of harmful ozone levels in the area, and, on the other hand, large-scale emission reductions alone are often not sufficient for reducing ozone efficiently in urban areas (*e.g.* Moussiopoulus *et al.* 2000). The method presented here could be used to assess the co-impacts of international emission reductions and local policies if detailed urban-scale photochemical models are not available. The following section presents an example of such European level scenario analysis, where the large-scale impacts of alternative European energy scenarios on acidification and rural background ozone levels were studied. In regional studies, these European level results could then be combined with the more detailed regional and local models, as discussed above.

4 Modeling joint European control strategies for CO_2, acidification and ground-level ozone

The UN/FCCC process and its Kyoto protocol are bringing new aspects into the air pollution control strategy development, since most planned CO_2 emission reduction measures affect also acidifying, eutrophying and ozone-forming emissions. Integrated assessment models can be applied to analyze the environmental impacts of these measures. This section presents a European level assessment of the synergies between global and regional control strategies for CO_2 and acidification and ozone formation for the case of the UN/FCCC Kyoto pro-

tocol and the air quality targets of the EU (article VI). The EU air quality targets for 2010 aim at cutting the amount of population and vegetation exposure to harmfully high ozone levels by two thirds and one third, respectively, from the level of 1990 throughout the EU. Acidifying deposition in excess of the ecosystem tolerance is targeted to be cut by 95 % from the 1990 level (European Commission 1999). The impacts of alternative 'Kyoto' energy strategies (meeting the CO_2 reduction targets of the Kyoto protocol) on the future regional air quality and on the additional efforts needed to achieve the air quality targets in the European Union were analyzed.

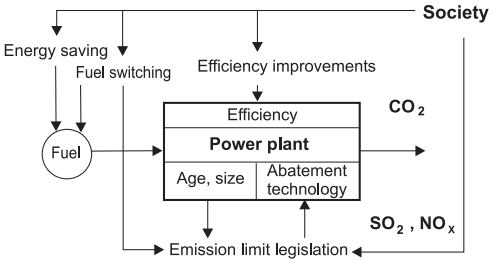

Fig. 10. The main mechanisms of CO_2 reduction measures and other air pollutant reductions (*e.g.* SO_2 and NO_x) in the energy system.

4.1 The system modeled

In general, the most cost-effective means of CO_2 reduction are energy saving measures and fuel switching to those with lower unit CO_2 emissions, *e.g*. from coal to gas or sustainable use of biomass. Energy saving measures reduce also emissions of sulfur dioxide and nitrogen oxides, as they depend directly on the fossil fuel consumption. Similarly, increases in the shares of nuclear, hydro-, solar or wind power reduce the direct sulfur and NO_x emissions as well. Since coal and heavy fuel oil have higher unit carbon emissions than gas or biomass and they have high sulfur contents as well, a shift in the fuel mix towards gas and (the sustainable use of) biomass reduces also sulfur emissions in installations where flue gas desulfurization is not required by legislation. NO_x emissions depend more on the boiler type (air flow, burner types, pulverized fuel, etc.) than on the fuel type. At present, primary NO_x emission reduction measures are standard technology in new installations, and in the EU countries required by legislation (UN/ECE 1999b, European Commission 1998), with the strictest limits on gas-fired installations. Thus a shift in energy production from old coal or oil-fired plants to newer gas-fired installations also has a NO_x-reducing effect. VOC emissions relate to energy use and industrial activities (*e.g*. solvent use). Reduction of *e.g.* transport fuel consumption reduces VOC emissions as well, but, for instance, increased wood burning in small boilers as a CO_2 abatement measure can increase VOC emissions. Fig. 10 illustrates how CO_2 reduction measures and other air pollutant reductions operate and interact in the energy production system. The principal measures to reduce CO_2 emissions induced by the society are drawn on the upper part of Fig. 10, and factors relevant for other air pollutants (*e.g.* SO_2 and NO_x) are drawn on the lower part of the Figure. Energy saving or improved efficiency of energy production or end-use result in a lower fuel use and thus have direct reducing effects on all emissions. However, efficiency improvements by higher boiler temperatures can result in increased NO_x emissions. The effect of fuel switching on SO_2 and NO_x emissions depends on the emission legislation and it varies between countries, fuels and plant size and age classes.

The results of the European-scale energy system model PRIMES, developed and used at the National Technical University of Athens (NTUA) (Capros *et al*. 1999a,b, Antoniou and Capros 1999), were used to analyze the effects of energy strategies adapted to CO_2 reduction on multiple air pollution problems (article VI). PRIMES is a modeling system simulating energy supply and demand in the EU member states, and for this study, it was used to find cost-minimal energy structure solutions meeting the EU Kyoto reduction targets for CO_2. Two alternative 'Kyoto' scenarios were used as input to the RAINS model to analyze their impacts on acidifying and ozone-forming emissions. In particular, the sulfur, nitrogen and VOC emission reductions required to achieve the EU interim air quality targets, assuming the 'Kyoto' energy scenarios, were calculated with the optimization module of RAINS and compared with those under a conventional 'Baseline' energy scenario, which assumes an absence of CO_2 abatement measures.

RAINS contains modules for calculating the

emissions of sulfur, nitrogen oxides, ammonia and volatile organic compounds. These modules include databases on energy consumption for European countries, distinguishing 21 categories of fuel use in six economic sectors. The time horizon extends from 1990 to 2010. Emission estimates are based on the CORINAIR inventory of the European Environment Agency (EEA 1996). The modules incorporate information about *e.g.* average sulfur contents of the fuels, typical unabated emission coefficients for each pollutant, control technologies at use and parameters describing the unit reduction costs in European countries (Cofala and Syri 1998a, b, Klimont 1998, Klimont *et al.* 1998). Emission reductions calculated by RAINS are assumed to be achieved only with technical control options and feedbacks of emission controls on the level and the structure of energy consumption are neglected. As a starting point, RAINS estimates the emissions for the year 2010 (given any energy scenario as input) expected to result from the presently agreed legislation on emission controls in the European Union. Such legislation includes the Large Combustion Plant Directive (EU 1988), its proposed revision (European Commission 1998) and limits on sulfur content for heavy fuel oil and diesel (EU 1998a). For mobile sources, the decisions on the Auto/Oil emission standards are taken into account (EU 1998a, b, c), together with the Council conclusions about emission standards for heavy-duty vehicles (EU 1998d) and the regulations for non-road machinery engines (EU 1998e).

Atmospheric dispersion and transformation processes of sulfur, nitrogen oxides and ammonia are modeled in RAINS using the source-receptor matrices of the Lagrangian model of EMEP. For the assessment of environmental impacts, RAINS contains a Europe-wide database of critical loads for acidification and eutrophication compiled by the UN/ECE Coordination Center for Effects (CCE) at the Dutch National Institute of Public Health and the Environment (RIVM) in the Netherlands (Posch *et al.* 1999).

4.2 Energy scenarios

In this study, two alternative 'Kyoto' scenarios obtained as least-cost energy structure solutions of PRIMES were used (Capros and Mantzos 1999). The 'No Trade' (NT) scenario assumed that the Kyoto protocol would be implemented in the EU-15 according to the EU burden sharing agreement, without any greenhouse gas emission trading taking place. The 'Full Trade' (FT) scenario supposed that CO_2 emission permits would be traded with all Annex B countries (UN/FCCC 1998) and the CO_2 reduction in every EU country is determined by the least-cost solution of PRIMES. In the 'No Trade' case, the CO_2 emissions of the EU-15 would decrease by 8 % from 1990 to 2010, following the Kyoto protocol. In the 'Full Trade' they would stabilize at the 1990 level, the rest of reductions being implemented outside the EU. In both 'Kyoto' energy scenarios examined in this study, about half of the CO_2 emission reductions would be achieved by decreases in total energy consumption, and the rest by changes in fuel mix, efficiency improvements and in the 'Full Trade' scenario also with emission trading. The average cost per ton of carbon avoided would be 180 € and 50 € in the 'No trade' and 'Full trade' scenarios, respectively (Capros *et al.* 1999a).

The environmental impacts of the 'Kyoto' scenarios were compared with a conventional 'Baseline' scenario reflecting a situation without any CO_2 reduction obligations. It is a compilation of the national official energy forecasts in the EU-15 countries from the late 1990s. For the countries for which these were not available, the 'Baseline' energy scenario developed with PRIMES in collaboration with the European Commission was used. For all the EU-15 countries, the 'Baseline' compilation used in the study assumed a high economic growth and increase in energy use, mainly met with expanded use of coal or gas. Fig. 11 illustrates

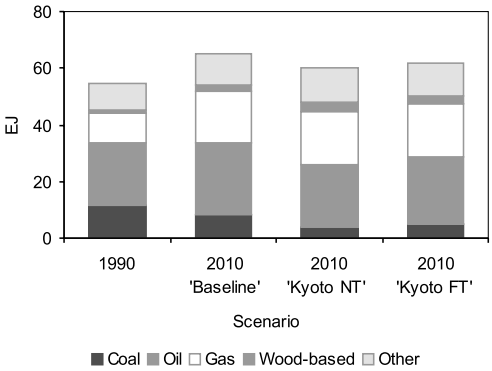

Fig. 11. Primary energy consumption in the EU-15 in 1990 and in 2010 with the 'Baseline', 'Post-Kyoto NT' and 'Post-Kyoto FT' scenarios (Capros and Mantzos 1999, article VI).

changes in the primary energy consumption for the main fuel categories in the EU-15 from 'Baseline' to the 'Kyoto NT' and 'Kyoto FT' scenarios.

4.3 Results

Table 4 compares SO_2 and NO_x emissions in 1990 with the levels expected for the year 2010 assuming regulations on emission controls agreed by 1999 for the 'Baseline' and 'Post-Kyoto NT' energy scenarios, respectively. For the 'Baseline' energy scenario, the existing and decided regulations on emission control will result for the EU-15 in considerable decreases of SO_2 and NO_x emissions from 1990 to 2010. Also the change in the energy consumption structure, particularly the lower reliance on fuels with high sulfur contents, contributes to the decrease in emissions. The effects of a changed energy structure are more pronounced in the 'Kyoto' energy scenarios. With the same emission controls in 2010, all energy-related emissions would be further reduced.

Implementation of the current legislation in the transport sector forms the main part of control costs in the EU-15 from 1990 to 2010 (Amann *et al.* 1998–99). The total technical control costs induced by the current legislation with the 'Baseline' energy projection have been estimated at about 59 billion (10^9) € a^{-1} (starting from the 1990 emission control situation) (Amann *et al.* 1998–99). Out of these numbers, about 80 % comes from the transport sector, mainly consisting of the introduction of new engine technologies in light- and heavy-duty road transport as stipulated by the Auto/Oil program (EU 1998b, c, Cofala and Syri 1998b). Due to the lower activity levels in the 'Kyoto NT' and 'Kyoto FT' scenarios there would be about 4 and 2 billion € less annual costs, respectively, to implement the present legislation by 2010 (article VI).

Table 5 shows the optimized control costs on top of implementing the current legislation for meeting the interim air quality targets with the alternative energy scenarios. Table 5 shows that with the energy scenario meeting the CO_2 reductions of the Kyoto agreement (No Trading) the achievement of the EU air quality targets would require about 3.2 billion € or 43 % less per year investments in technical controls of sulfur, nitrogen and VOC emissions. The savings are mainly due to the fact that the energy structure of the 'Kyoto NT' scenario has less consumption of fossil fuels, generating less emissions. Therefore, the additional relative reductions needed for meeting the air quality targets are smaller and the most expensive control measures are avoided. Since the 'Kyoto' energy scenario based on free greenhouse gas emission trading ('FT') would induce more sulfur, nitrogen and VOC emissions with the current legislation than the scenario without trading, the additional costs of emission controls required to meet the air quality targets would be 0.6 billion € a^{-1} higher annually than with the 'Post-Kyoto NT', still being considerably less than in the 'Baseline' scenario.

In the cost-minimized solution of meeting the EU air quality targets, the further emission reductions would mainly take place in the industrial sector in all the energy scenarios studied. The measures in the industrial sector would incur about half of the total emission reduction costs on top of the current legislation. Table 5 shows that the sulfur, nitrogen and VOC emission reductions would cost considerably less in all economic sectors under the 'Kyoto' energy scenarios.

Table 4. Comparison of emissions (in kilotons) of the EU-15 for 1990 and the 'Baseline' and 'Kyoto' energy scenarios in 2010, assuming the present legislation.

	SO_2	NO_x
1990	16430	13230
2010		
"Baseline"	4690	6850
"Kyoto NT"	3640	6010
"Kyoto FT"	4110	6280

Table 5. Annual sectoral emission control costs on top of implementing the current legislation in the EU-15 for the optimized scenarios meeting the EU environmental interim targets (in % and billion €). Percentage values show the distribution of costs among the sectors.

	'Baseline' %	'Kyoto NT' %	'Kyoto FT' %
Power generation	6	6	9
Industry	44	58	50
Domestic	13	9	12
Transport	8	4	4
Agriculture	29	24	25
Total	100	100	100
Billion €	7.5	4.2	4.8

4.4 Concluding remarks

In this section, a European level assessment of the potential synergies between global and regional control strategies for CO_2 and acidification and ozone formation was presented. The results of the study demonstrate the side-benefits of global greenhouse gas emission control strategies on regional air quality. It should be noted that the simulated least-cost energy structures are scenarios resulting from cost-optimization modeling analysis, which naturally do not take into account political and societal aspects. The exact magnitude of the side benefits as well as their distribution among countries and economic sectors will change to some extent depending on the actual implementation programs of the countries. However, assuming full greenhouse gas trading and thus portraying a distinctly different energy production and consumption structure, arrived at similar conclusions about the synergies and their magnitude, which corroborates the robustness of the general conclusions.

The results underline the importance of considering the side effects of control strategies for single pollutants or single environmental problems. The significant potential for synergy between global greenhouse gas emission control strategies and regional air quality should be taken into account in the future development of cost-efficient mitigation policies.

The European level assessments of the side benefits of global CO_2 control strategies on long-range transported air pollution in Europe can also be linked, using regional and local scale models, with national and sub-national assessments and policy development. In the following section, the European level modeling results presented above are linked with the more detailed regional modeling (see also section 2) to analyze the future outlook of the acidification situation in Finland.

5 Effects of emission controls and low-CO_2 energy pathways on acidification in Finland

This section presents national-scale applications, where alternative European emission control strategies and domestic and bilateral policy scenarios were used to assess the future acidification situation in Finland and its amelioration options. The regional deposition modules and point-source databases of DAIQUIRI were linked with the European scale models. Section 5.4 describes how the regional deposition model can be employed to produce site-specific deposition time series of acidifying compounds for studying the ecosystem responses to changing deposition.

5.1 National assessment of critical load exceedances

In parallel to the European-level air pollution control strategy development of the UN/ECE and the EU, the Acidification Committee nominated by the Ministry of Environment prepared a national-level assessment of the Finnish emission reduction possibilities and of the future outlook on acidification in Finland (Acidification Committee 1998). In a study made for the Committee (article IV), the regional deposition model DAIQUIRI and the Finnish critical load database were utilized to assess the environmental impacts of alternative energy and emission control scenarios. The work of the Acidification Committee was done during the early phases of the Gothenburg protocol deliberations and the NEC Directive proposal, as their final formulation was not yet fixed (article IV). Here the analysis with the final emission ceilings of the Gothenburg protocol and the NEC Directive proposal is presented (article VIII). In addition, the effects of energy policies fulfilling the Kyoto greenhouse gas emission targets were assessed (article VIII).

Finland committed herself to substantial sulfur emission reductions in the Second Sulphur Protocol (UN/ECE 1994). Finnish annual sulfur emissions declined from 260 ktSO_2 in 1990 to 96 ktSO_2 in 1995, thus the target of 116 kt annual SO_2 emissions was achieved, and since 1995 the emissions have kept rather constant. This has increased the relative importance of nitrogen and imported deposition in general for acidification. In 1995 the domestic share in deposition of sulfur was over one fifth only in the vicinity of major emission sources, whereas the Finnish contributions to the deposition of nitrogen oxides and ammonia were 20–50 % and 20–70 %, correspondingly, everywhere in Southern and Central Finland (article IV). In 1995, acidity critical loads were exceeded at about 5.4 % of the total ecosystem area in Finland, mainly in southern and eastern parts of the country.

In the assessments, Finnish energy scenarios of the Ministry of Trade and Industry (1997, 2001) were used. The 'Energy Market Scenario' (EMS) assumed an average annual economic growth of about 2.5 %, and it predicted an increase of 31 % in the energy consumption during 1990–2010 mainly met by expanded hard coal and gas use (Ministry of Trade and Industry, 1997). With EMS, CO_2 emissions would increase by about 33 % from 1990 to 2010. The impacts of energy policies fulfilling the Kyoto greenhouse gas emission targets were assessed using the scenarios of the Finnish national climate strategy (Ministry of Trade and Industry 2001, Hildén *et al.* 2001). The 'Gas' scenario would fulfill the EU burden sharing agreement for Finland, *i.e.* the stabilization of net greenhouse gas emissions at their 1990 level by strong shifting from coal to gas and biomass, by accelerated penetration of renewable energy technologies and by decreased total energy consumption. In the other scenario, increased nuclear capacity would be used to replace coal. Acidifying emissions in 2010 with the 'Gas' and 'Nuclear' scenarios were estimated at 91 $ktSO_2$ and 170 $ktNO_2$ and 88 $ktSO_2$ and 172 $ktNO_2$, respectively (Hildén *et al.* 2001).

A detailed point-source emission database for Finland and the nearby regions of Estonia and Russia was utilized together with the meso-scale deposition modules to estimate the impacts of domestic and nearby emissions with higher accuracy. Every combustion plant in Finland with a capacity greater than 5 MW_{th} is included in the database (Melanen and Ekqvist 1997). The emission scenarios for diffuse sources, such as traffic, domestic heating and agriculture, were resolved at municipality level, assuming the latest available spatial distribution. As there are several major sulfur emitters near Finland in Estonia and Russia, scenarios used in bilateral emission reduction negotiations were also taken into account in the assessment to reflect the potential of bilateral measures. For the assessment of ammonia emissions and the impacts of agricultural scenarios and measures, an emission model was developed at SYKE and utilized in the assessment (Grönroos *et al.* 1998).

In the study, the regional nitrogen deposition module of DAIQUIRI (see section 2) was used together with the regional sulfur module developed earlier (Johansson *et al.* 1990). Long-range transported deposition was estimated with the EMEP long-range transport matrices (Barret and Berge 1996, EMEP/MSC-W 1998). The Finnish critical load database (Johansson *et al*. 1999) was used to estimate the exceedances of critical loads.

The case that the European legislation would not have been tightened from the situation in the mid-nineties after the signing of the Second Sulphur Protocol in 1994 (UN/ECE 1994, 1995, Amann *et al.* 1996) was calculated (illustrated as scenario 'UN/ECE 1994'). The 'Baseline' emission scenario contains all emission control legislation adopted or accepted by the year 1999. During the 1990s, the EU Auto Oil program produced regulations, which will reduce effectively NO_x and VOC emissions from the transport sector in the EU countries. The proposed revision of the Large Combustion Plants Directive (European Commission 1998) was also included in the 'Baseline' estimate. For the non-EU countries, the 'Baseline' assumes that new power plants built to replace the aging capacity would fulfill the emission standards set in the Second Sulphur Protocol. The development of emissions with the scenarios studied are given in articles IV and VIII. Acidifying emissions in 2010 with the UN/ECE Gothenburg protocol are denoted as 'UN/ECE 1999' (UN/ECE 1999a). 'EU/NEC' shows situation in 2010 assuming the implementation of NEC directive proposal (European Commission 1999) in addition to the Gothenburg protocol. Finland has conducted bilateral negotiations with the Russian Federation and Estonia on reducing sulfur emissions in the bordering areas. 'BIL' includes in addition to 'UN/ECE 1999' and 'EU/NEC' these planned bilateral sulfur reductions (article IV).

In addition, a scenario introduced in section 4, 'Kyoto (NT), was used together with the Finnish 'Gas' scenario to study the effectiveness of structural changes in the energy system of the EU-15 in reducing critical load exceedances in Finland (denoted as 'Kyoto' in Fig. 12). In this scenario, the fulfillment of the UN/ECE Gothenburg protocol and the EU NEC directive proposal were not assumed, and the acidifying emissions would be determined by the 1999 emission legislation.

5.2 Effectiveness of domestic, bilateral and international emission reductions

Fig. 12 illustrates the results of the scenario analysis (article VIII). Ecosystem areas with acidity critical load exceedances are shown for 1990, 1995

and 2010 for the scenarios studied. The bar 'Baseline' illustrates the situation with the implementation of present legislation (excluding the Gothenburg protocol and the NEC Directive proposal) and assuming conventional energy pathways not fulfilling the Kyoto protocol. The bar 'Kyoto' shows a case with the same legislation as 'Baseline', but with the 'Kyoto NT' energy scenario described in section 4 and the Finnish 'Gas' scenario. In addition, the impacts of the further emission control policy options are illustrated (with the energy scenario of 'Baseline' as background assumption). For reference, Fig. 12 also shows the area at risk of acidification in Finland if the European legislation would not have been tightened from the situation in the mid-nineties, illustrated as 'UN/ECE 1994'.

The results show that there has been a strong decrease in the area at risk of acidification in the 1990s. The decrease in the early nineties can partly be attributed to the rapid decline in Finnish sulfur emissions. Other important elements are the EU legislation concerning acidifying emissions and the decline of acidifying emissions from the former centrally planned economies.

The expected decrease of long-range transported deposition will bring considerable benefits to Finnish ecosystems. Already with the new legislation in Europe after the mid-nineties ('Baseline' in Fig. 12) the ecosystem protection will improve significantly from that expected with the mid-nineties legislation. As already the 'Baseline' situation contains quite advanced emission control legislation, there is less cheap potential left for further technical measures to achieve the EU air quality targets set in the acidification and ozone strategies. The NEC Directive proposal focused largely on ozone reduction in Central and Southern Europe, and thus the reduction in acidifying deposition to Finland would be small in relation to the high costs of achieving the targets. As the majority of acidifying deposition is imported and Finnish legislation exhausts most of the technically feasible and cost-effective control options, the potential to combat acidification with further Finnish technical emission reductions alone is small.

For Estonia and Russia, the Gothenburg protocol allows growth in emissions from their present level. Further measures at the neighboring large emission sources in Russia would have significant

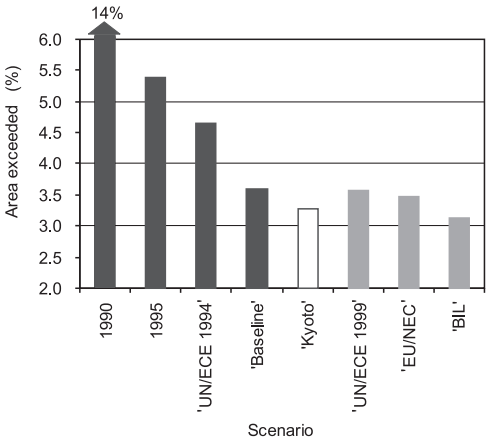

Fig. 12. The percentage of ecosystem areas at risk of acidification in Finland in 1990, 1995 and in 2010 with the scenarios studied.

beneficial effects for adjacent Finnish ecosystems. The major sulfur emitters in Estonia and in the Russian areas next to Finland cause considerable acidifying load to the bordering Finnish areas. During the 1990s, a decline of sulfur emissions has taken place both in Estonia and in the Russian areas bordering Finland. Especially in Russia, however, the decrease has been mainly due to the economic recession and the resulting decline in production instead of investments in emission control technology. The emissions may increase along with the recovery of industrial production and the energy supply, since it is often feared that environmental investments will not be high on the agenda during an economic revitalization (*e.g.* Golitsyn 1992). Fig. 12 demonstrates that further measures at the adjacent large emission sources in Russia would have significant beneficial effects for nearby Finnish ecosystems, though the reduction percentages assumed in the 'BIL' scenario were modest compared with those already achieved in Western Europe.

5.3 Impacts of CO_2 abatement policies in the EU

Fig. 12 shows also the plausible influence of implementing the Kyoto protocol in the EU-15 on ecosystem acidification in Finland. This scenario as-

sumes the same legislation as the 'Baseline' scenario, which incorporates all measures required by legislation up to 1999, excluding the Gothenburg protocol and the NEC Directive (see also section 4). Thus the bars 'Baseline' and 'Kyoto' in Fig. 12 display the difference in ecosystem protection achieved solely with the alternative energy structure.

The results demonstrate that shifts in the EU energy system towards less carbon-intensive fuels and energy saving could bring notable reduction of acidifying emissions and ecosystems at risk of acidification. This example, with Finland as receptor, does not fully reflect the environmental side-benefits of these measures, as in Eastern Finland a large part of deposition originates from Russia and Estonia. Nevertheless, structural measures in the EU taken to fulfill the Kyoto protocol (without further technical emission controls) could protect Finnish ecosystems up to 8 % more than the UN/ECE Gothenburg protocol.

However, especially when drawing conclusions from scenario analyses resulting in small differences in critical load exceedances (*e.g.* 'UN/ECE 1999' and 'EU/NEC' scenarios in Fig. 12), one should bear in mind that the critical load exceedance estimates involve considerable uncertainties (see section 6). In addition, the critical load is a relatively simple concept, which is not able to reflect the time-dependent acidification processes. In the following, the use of the deposition model DAIQUIRI (see section 2) in a case study of dynamic acidification modeling investigating the impacts of some of the above emission reduction scenarios at catchment scale is introduced.

5.4 Derivation of deposition scenarios for dynamic acidification models

The emission and deposition scenarios can also be utilized in dynamic modeling to assess the impacts of alternative European emission reduction pathways on ecosystem recovery from acidification at catchment level. In the context of an international modeling project of acidification development at research sites (Forsius *et al.* 1998), DAIQUIRI was used to produce the acidifying deposition trends during 1800–2010 for the sites studied, including the Hietajärvi catchment in Northern Karelia (article III). The historical sulfur deposition estimates of Mylona (1993) and future emission scenarios of IIASA (Amann *et al*. 1996) were utilized. For the study, site-specific deposition trends representing the 'UN/ECE 1994' scenario described in the previous section (denoted as 'REF' in article III), a preliminary scenario of the UN/ECE Gothenburg protocol development (B1) and the Maximum technically Feasible Reductions (MFR) scenario were calculated.

Dynamic modeling of the soil acidification at Hietajärvi with the SMART model showed the onset of deterioration in the stream water pH and soil base saturation in the 1960s as a response to the sharply increasing acidifying deposition. The model runs indicated that only the MFR scenario would be able to completely reverse the occurred acidification. The less stringent scenarios were able to stop and partly reverse the acidification process. This is in accordance with the critical load approach, which indicates that the area would be protected in 2010 already with the less stringent reduction scenarios (Ahonen and Rankinen 1998).

This example demonstrates the conceptual differences between the static critical loads, aimed at indicating the regional ecosystem sensitivities for large-scale control strategies, and the catchment-level dynamic modeling. The following section investigates more closely the effects of uncertainties in the critical loads and in the other parts of the integrated assessment models on the estimates of ecosystem areas at risk, which are used as criterion in the international policy-making.

6 Model uncertainties and probabilistic scenario analysis

The European policies aimed at reducing environmental problems caused by long-range transported air pollution require substantial abatement efforts. The exhaustion of cheap technical emission controls increases the uncertainties in the costs of achieving a certain environmental protection level, which are calculated by the integrated assessment models. The sensitivities of emission ceilings and associated control costs to small changes in background assumptions have been demonstrated by, *e.g.,* Bak and Tybirk (1998), who found that the emission ceilings and related reduction costs can change considerably with relatively small changes in deposition targets or other underlying assumptions. The significant costs stress the importance of knowing the reliability and limitations of integrat-

ed assessment models used in designing those policies.

Within this work, a mathematical uncertainty analysis for the integrated assessment modeling of the areas at risk of acidification in Finland due to emissions in European countries was conducted (article VII). The analysis was done by constructing a model framework, which enables a probabilistic analysis of emission reduction scenarios.

6.1 Uncertainties in acidification modeling

The assessment considered uncertainties in national inventories of acidifying emissions in European countries and their spatial locations, in the atmospheric transport of the pollutants and in the critical loads characterizing the ecosystems' sensitivity (article VII). The relative contributions of the uncertainty of individual modules of integrated assessment modeling to the overall uncertainty of ecosystem protection levels were calculated for 1990 and 2010, assuming the implementation of the UN/ECE Gothenburg Protocol. The uncertainties associated with the individual modules were mainly estimated by reviewing existing literature.

The principal sources of uncertainty in this context can be attributed to:

(i) Model structure: long-range transport (LRT) models used, how well does the steady-state critical load concept represent the acidification process, etc.
(ii) Input data: measurement errors, errors or uncertainties of modeled input parameters.
(iii) Lack of data and data sampling strategies affecting the representativeness of the input data and of the selected ecosystems, interpolation or extrapolation of data.
(iv) Lack of knowledge: *e.g.* dry deposition to different surfaces, incomplete understanding of the system to be modeled, etc.

An exact quantification of the magnitude of all individual errors caused by the sources listed above is not possible, because this would require more knowledge about the real world systems than is currently available. Instead, the sources of uncertainty were prioritized by reviewing the existing literature about how significant these could be. The shortcomings of the model structures (i) and model limitations (i, iv) concerning atmospheric transport and deposition modeling are briefly described in the following. The shortcomings concerning soil/lake acidification modeling are discussed in (article VII). Uncertainties in input data (ii) caused by possible errors in emission estimates, LRT model input parameters and input parameters for calculating critical loads are summarized in the following. Uncertainties driven by sampling strategies affecting the representativeness of the ecosystem data (iii) used here are based on Forsius (1992).

In the EU countries sulfur emissions can be considered relatively well known, because reliable statistics of fuel consumption and fuel characteristics are available. Sulfur emissions are a function of the sulfur contents of the fuel, share of sulfur retained in ash and the effectiveness of end-of-pipe reduction measures. The major part of sulfur emissions comes from large point sources, which are usually required to continuously monitor their emissions, and these emission estimates can thus be considered rather reliable. Emission estimates from disperse sources are usually based on national fuel consumption statistics and average emission factors, which makes them less reliable than estimates based on continuous monitoring. In this study, a ±5 % range of variation for European country-total sulfur emissions was assumed, which may be too optimistic for many East European countries. The influence of a larger country-emission uncertainty was investigated by assigning a ±30 % range of variation for sulfur emissions.

Nitrogen oxide emissions are more difficult to estimate than sulfur emissions. The formation of nitrogen oxides in combustion processes depends mainly on the burning conditions, such as flame temperature and amount of oxygen available, and on the nitrogen contents of the fuel. A considerable share of NO_x emissions comes from large point sources, which are required to monitor and report their emissions, and thus those emission estimates can be considered rather reliable. The largest single NO_x emitting sector, however, is transport. In the EU-15 in 1990, for instance, NO_x emissions from the transport sector were 8.43 out of 14.3 $MtNO_2$ total emissions (59 %). In the near future the share of transport in total NO_x emissions in Europe is expected to decrease considerably due to the continuing penetration of advanced catalytic converters in the vehicle fleet, as stipulated by the Auto/Oil program of the EU (*e.g.* EU 1998c, d).

Transport emission inventories are usually

based on relatively limited measurements of typical emission factors, multiplied by the total mileage driven or fuel used in a country. In this study, a ±15 % range of variation, with a sensitivity run with a ±30 % range of variation, for country-total NO_x emissions was assumed for all European countries based on assessments of individual sectors (*e.g.* Sawyer *et al.* 2000, Placet *et al.* 2000). The studies of emissions from individual sources or sectors often find larger error or uncertainty ranges than the ±15 % used in this study. However, one should remember that the sum of independent uncertain variables has smaller relative uncertainty.

Practically all ammonia emissions in European countries come from agricultural activities. Agricultural ammonia emission inventories are based on statistics about fertilizer use, animal numbers, fodder types, storage and stable types and control measures used. Based on limited measurements, average emission coefficients are assigned to each activity and control measure. The largest uncertainties are in the average emission coefficients used for calculating the evaporation rates and in the average efficiencies of the reduction techniques used. The total uncertainties of agricultural emissions have been estimated to be in the order of ±30–40 % (*e.g.* Asman and Jaarsveld 1992, Grönroos *et al*. 1998). A ±40 % range of variation for ammonia emissions was used here.

Similar inventory methods and emission coefficients for acidifying emissions are used in European countries. Therefore the uncertainties can be assumed to be correlated to some extent among countries, and in this study an inter-country correlation coefficient of 0.5 was used. The assumption of correlations leads to conservative estimates of the importance of uncertainties in emissions. If no correlations were assumed, the individual countries' emission uncertainties would tend to cancel each other out.

Long-range transport modeling of air pollutants considered in this work is based on the Lagrangian model of EMEP with a 150 km x 150 km horizontal resolution. The model represents the lower atmosphere in one layer, assuming homogenous mixing of emissions. Uncertainties in the long-range transport matrices used in integrated assessment modeling of air pollution have been analyzed by Alcamo and Bartnicki (1987, 1990), who investigated the parameter uncertainties of an earlier version of the EMEP model. They suggested a coefficient of variation (CV) of 27 %. This estimate is assumed to include also the uncertainty caused by incomplete knowledge about the location of emission sources within countries, which is of minor significance in the uncertainty of pollution transported over thousands of kilometers. The studies by Alcamo and Bartnicki considered only sulfur; however, due to the lack of other information the same CV was used for nitrogen compounds as well. A sensitivity run with a CV of 35 % for nitrogen compounds was carried out in conjunction with the more pessimistic uncertainties of a ±30 % range of variation for sulfur and nitrogen oxide emissions.

Inter-annual meteorological variability can have significant effects on the deposition to Finland, because in the immediate vicinity of the borders in the South and in the East there are large emission sources and thus weather anomalies can have large effects on the amount transported to Finland. The mean meteorology of 1985–1996 utilized in the study was found to produce approximately the average of the individual years' ecosystem protection level. Therefore the use of long-term average atmospheric transport matrices seems to eliminate the effect of the inter-annual meteorological variability and represent long-term average deposition patterns, thus justifying their use for scenario analyses.

The EMEP model's 150 km resolution is rather coarse compared with the scale of the ecosystems used to characterize ecosystem acidification in Finland. The deposition model resolution can significantly affect the estimates of ecosystem areas at risk when there are significant gradients in deposition. The coefficient of variation for in-grid deposition due to indigenous emission sources was estimated as the difference between the deposition fields calculated alternatively with the meso-scale module and with the EMEP model interpolated to the meso-scale grid of $^1/_4° \times {}^1/_8°$ (see section 2). Meso-scale deposition fields from Finnish and nearby area emissions were added to the long-range deposition. The coefficient of variation was biggest for ammonia close to agricultural areas, where the share of local emissions in total deposition is the largest.

The uncertainties in critical loads for forest soils in Finland have been investigated by Johansson (1999), who arrived at a CV estimate of ±30 % for forest soils. The uncertainty estimates of critical loads for lakes used in this work are

based on work by Forsius (1992), and a CV of ±10 % was used for lake critical loads in Finland.

6.2 Analytical formulation

The probability P_{EX} to exceed a critical load with a given deposition vector (D_N, D_S) was defined by assuming a normal distribution with standard deviation σ_{CL} around the critical load function of every individual ecosystem:

$$P_{Ex}(CL_{N,S} < (D_N, D_S)) = \begin{cases} \Phi\left(\frac{D_S - CL_{max}(S)}{\sigma_{CL} CL_{max}(S)}\right) & \text{if } D_S > D_N \frac{CL_{max}(S)}{CL_{min}(N)} \\ \Phi\left(\frac{D_N - CL_{max}(N) + D_S/\gamma}{\sigma_{CL} CL_{max}(N)}\right) & \text{if } D_S \le D_N \frac{CL_{max}(S)}{CL_{min}(N)} \end{cases} \quad (7)$$

where Φ is the cumulative Gaussian distribution, and

$$\gamma = \frac{CL_{max}(S)}{CL_{max}(N) - CL_{min}(N)} \quad (8)$$

where $CL_{max}(S)$ is ecosystem's maximum critical load for sulfur, and $CL_{min}(N)$ and $CL_{max}(N)$ are the ecosystem's minimum and maximum critical load for nitrogen. They define a trapezoid function in the N-S plane (Posch *et al.* 1999).

For the probabilistic assessment, the mean of the deposition of a compound X (where X=SO_2,NO_x,NH_3) to an EMEP 150 km × 150 km grid cell j, $D_{X,j}$, can be written as:

$$D_{X,j} = \lambda_{X,j} + \sum_{i=1}^{N} \varepsilon_{X,i} E_{X,i} \alpha_{X,i,j} A_{X,i,j} \quad (9)$$

where

$\lambda_{X,j}$ = uncertainty due to in-grid variation in grid j; expected value E[$\lambda_{X,j}$] = 0
$E_{X,i}$ = emissions from country i
$\varepsilon_{X,i}$ = relative uncertainty of the emission from country i . E[$\varepsilon_{X,i}$] = 1
$A_{X,i,j}$ = transfer coefficient from country i to grid j
$\alpha_{X,i,j}$ = relative uncertainty of the transfer coefficient. E[$\alpha_{X,i,j}$] = 1
N = number of countries.

These mean depositions are used to define a bivariate lognormal distribution f_D of correlated sulfur and nitrogen deposition (D_N, D_S) (article VII). The probability to exceed critical loads by the deposition (D_N, D_S) with distribution f_D is then calculated as:

$$P_{Ex} \mid f_D = \int_0^\infty \int_0^\infty P_{Ex}(CL_{N,S} < (D_N, D_S)) f_D(D_N, D_S) dD_S dD_N \quad (10)$$

The above equations allow to calculate the uncertainties of critical load exceedances due to uncertain emissions, atmospheric transport and critical loads for each ecosystem. To obtain uncertainty estimates for all Finnish ecosystems, these individual calculations were summarized and presented as probability distributions.

6.3 Probabilistic scenario analysis

Fig. 13 shows how the uncertainty of ecosystem protection level was found to develop from 1990 to 2010. Also shown are the relative contributions of individual modules to the total uncertainty. The robustness of ecosystem protection level estimates in Finland was found to improve significantly from the year 1990 to 2010, assuming emissions according to UN/ECE (1999). This is because the level of acidifying deposition will decrease and thus for many ecosystems depositions will be less than the critical loads with a higher probability than in 1990, even though the relative uncertainties in deposition and critical load values stay the same. Fig. 13 demonstrates that at national scale the most significant are the uncertainties in critical loads. More pessimistic assumptions on the uncertainties in emission inventories or in atmospheric transport modeling did not significantly change the overall results. This indicates that further research efforts to reduce uncertainties should mainly focus on improving the descriptions of ecosystem processes. However, in areas close to emission sources, the atmospheric transport and the in-grid variability of deposition contributed significantly to the overall uncertainty. Critical load exceedances and their uncertainties in the Finnish areas adjacent to Russia and Estonia are strongly influenced by emissions and their uncertainties from these areas. This emphasizes the need for high-resolution modeling when developing protection strategies for natural areas in the vicinity of large emission sources.

The probabilistic calculation method presented above can be used for scenario analysis to investigate the ecosystem area protection probabilities with alternative reduction scenarios instead of the deterministic approach used in sections 4 and 5.

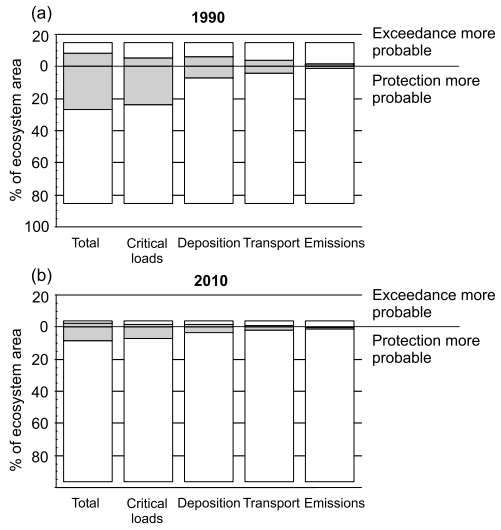

Fig. 13. The uncertainty of ecosystem protection level in (a) 1990 and (b) 2010, and the shares of module uncertainties in total uncertainty. In the leftmost bars, all modules have been assigned their uncertainties ("Total" = emissions + atmospheric transport + in-grid variability + critical load). In "Critical loads" only critical loads are assumed uncertain. "Deposition" comprises emissions, atmospheric transport and in-grid variability of deposition. The white top part of each bar represents the area where exceedance is certain (probability > 0.95). The gray part in each bar shows the area where it's uncertain whether there is protection or exceedance (probability between 0.05 and 0.95). The white part on the bottom of each bar gives the area where protection is certain (exceedance probability < 0.05).

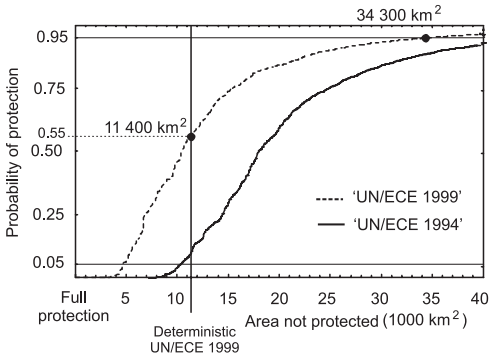

Fig. 14. The protection probabilities for the 'UN/ECE 1994' and 'UN/ECE 1999' scenarios (see section 5). The unprotected area estimated with the deterministic approach (11400 km^2) for the UN/ECE Gothenburg protocol ('UN/ECE 1999') is illustrated in the Figure.

Fig. 14 shows an example of scenario analysis results with the probabilistic method. The Finnish ecosystem area protection probabilities for 2010 with scenarios 'UN/ECE 1994' and 'UN/ECE 1999' (see section 5) are plotted. For the Gothenburg protocol and the EU NEC directive development, the areas at risk of acidification were estimated deterministically with the RAINS model. The area in Finland remaining at risk of acidification with the Gothenburg Protocol was calculated to be 11400 km^2 (Amann *et al.* 2000). Fig. 14 summarizes the estimates of protected areas of the probabilistic approach described here, and compares them with the corresponding deterministic estimate calculated with the RAINS model.

Fig. 14 illustrates that the estimates of areas at risk in Finland in 2010 increase rapidly if a bigger probability for protection is required. In the case of the Gothenburg protocol, if the modeling uncertainties described above are taken into account, there is a 55 % probability to protect the area (11400 km^2), which the deterministic approach estimates as protected. A conservative interpretation of the probabilistic method (protection probability greater than 0.95) gives a threefold area unprotected (34300 km^2) than the deterministic approach. With the 'UN/ECE 1994' scenario considerably larger areas would have remained at risk (with any probability level chosen as indicator). If a 95 % probability of protection is required, 53900 km^2 would have been at risk with the 'UN/ECE 1994' scenario.

6.4 Concluding remarks

The purpose of this uncertainty study was to develop a framework for the quantitative assessment of uncertainties and to estimate the relative importance of the model parts in the total uncertainty to guide further research and decision-making.

The presented examples illustrated that small differences in the areas with critical load exceedances estimated with deterministic integrated assessment models do not fully portray the risks to the ecosystems and can be misinterpreted with respect to the expected benefits of the emission reductions. The approach presented allows to display the differences in the probability of ecosystem protection between reduction scenarios in order not to underestimate the ecosystem areas at

risk, analogously to (Hirst *et al.* 2000, Barkman *et al*. 1995). It should be utilized in the design of ambitious and costly reduction policies to enable decision-makers to balance the risks of exceedance and costs involved in various emission reduction scenarios.

In assessing the role of long-range transport of pollutants in the total uncertainty of integrated assessment modeling, it should be remembered that the model parameter uncertainties used were derived for an earlier version of the EMEP model. For more reliable estimates, the uncertainties of the current model version should be investigated.

In addition, the impacts of local variability in deposition and in critical load values were not profoundly assessed due to the preliminary scope of the study. These should be investigated more closely and involve comparison with deposition measurements as well.

7 Conclusions

This thesis presented the development and applications of regional and local scale models for use in integrated assessment of air pollution effects in conjunction with large-scale models. The model applications included studies of regional environmental side benefits of global CO_2 control strategies, which have become topical with the UN/FCCC process and its Kyoto protocol. These strategies have significant potential for reducing air pollution as well. The model applications presented in this study contributed to identifying the problem characteristics and have supported environmental policy development at international, national and regional levels.

The development of the regional nitrogen deposition model DAIQUIRI for integrated assessment purposes in this study was successful, as the model was found to perform well in estimating long-term average nitrogen deposition in forest-dominated areas. Long-term trends and the average level of deposition estimated with DAIQUIRI were comparable with the monitored deposition levels and trends. For areas dominated by other surface types, DAIQUIRI overestimated nitrogen deposition, which is however of less importance in acidification research in Finland. The long-term levels and source attribution of deposition are the most relevant parameters in the integrated assessment modeling of acidification and eutrophication of forest soils. For the mid-nineties the regional nitrogen deposition modeled with DAIQUIRI resulted in 9 % to 19 % (depending on the region compared) larger estimates of areas with acidity critical load exceedances than when using the European scale nitrogen deposition model. This demonstrates that domestic deposition should be estimated with regional or local scale models in order not to underestimate or misallocate the impacts of domestic emissions. The computationally simple technical structure of DAIQUIRI facilitates various applications and scenario studies in support of environmental policy development.

The method development and testing for estimating the impact of urban NO_x levels on ozone concentrations indicated that urban NO_x levels are a significant explanatory factor in the differences between urban and surrounding rural ozone concentrations. Correlation coefficients between measured daytime ozone values in the study area were found to improve from 0.64 (correlation between urban and surrounding rural measurements) to 0.85, on the average. The average correlation between daytime large-scale model estimates and urban site measurements increased from 0.37 to 0.58 with the method tested. The results indicated that the urban NO titration effect could be satisfactorily represented in integrated assessment models with the method studied.

The synergies between control strategies for CO_2 and acidification and ozone formation in the case of the UN/FCCC Kyoto protocol and the air quality targets of the EU were assessed with the help of linked models. With two alternative energy scenarios reflecting the Kyoto targets for CO_2, reductions of sulfur and NO_x emissions between 12 % and 22 % and 8 % to 12 %, respectively, were estimated by 2010 in the EU-15 with the present emission control legislation. The European countries' official energy projections in the 1990s, assuming continued growth in fossil fuel consumption, would partly offset the environmental improvements achieved by technical controls and would demand application of advanced and expensive emission control technologies in some countries to achieve the agreed reduction targets. With structural changes in energy production and consumption, the most expensive air pollutant control measures in the problem regions could be avoided. The further emission reductions required for achieving the EU acidification and tropospheric ozone control targets for 2010 would cost 35–43 %

less under the studied energy scenarios fulfilling the Kyoto protocol, which is due to the lower activity level, energy saving measures and cleaner energy forms used. The results demonstrate that it is important to take into account the significant potential for synergy between global greenhouse gas emission control strategies and regional air quality in the future development of cost-efficient mitigation policies for the multiple pollutants.

The model applications for Finland indicated that there has been a 60 % decrease in the area at risk of acidification from 1990 to 1995 and that the declining trend is expected to continue due to the recent international emission reduction agreements within the UN/ECE and the EU. The positive trend in the nineties can partly be attributed to the rapid decline in Finnish sulfur emissions. Other important elements are the EU legislation concerning acidifying emissions and the decline in acidifying emissions from the former centrally planned economies. Analysis of Finnish and European energy scenarios meeting the Kyoto greenhouse gas reduction targets demonstrated that implementation of the Kyoto protocol (with the present emission legislation) in the EU-15 could bring up to 8 % more reduction of ecosystems at risk of acidification in Finland than the recent UN/ECE protocol.

The uncertainty analysis performed in this study indicated that critical loads dominate the uncertainty of acidification integrated assessment modeling in Finland. The estimates are becoming more robust, as the general level of deposition is decreasing. In areas affected by major nearby emission sources, also uncertainties in emissions and deposition are significant. These can be reduced by using individual point-source emission data and meso-scale deposition modeling developed in this work in contrast to emissions aggregated to large grids used in international reporting and deliberations.

The probabilistic modeling approach of acidification control strategies presented in this study allows to display the differences in the probability of ecosystem protection between reduction scenarios. It can be used to quantify the uncertainties related to the estimates and to communicate them to policy-makers. This is of growing importance, as areas at risk are diminishing and as reduction costs are increasing rapidly with the exhaustion of cheap technical emission control options. Probabilistic approaches should be used in the design of ambitious and expensive reduction policies to enable decision-makers to balance the risks of adverse environmental impacts and the costs of emission reduction measures.

The following topics for further research can be identified:

- The description of nitrogen deposition to waters and coastal areas should be improved in DAIQUIRI to allow its use for the assessment of control strategies against eutrophication and acidification of surface waters.
- The method for estimating the impacts of local NO_x emissions on urban and sub-urban ozone should be further developed with widely applicable ways of deriving the required parameters and tested in a larger variety of locations.
- Multiple global and regional environmental problems should be considered simultaneously. In addition to the examples of the synergies of CO_2 reductions and acidification and ozone formation presented in this study, this holds also for fine particles causing adverse health effects. An effective mitigation of climate change will require profound changes in the global energy production and consumption structures in the next decades. If realized, the mitigation measures will have large impacts on the emissions of all air pollutants.
- The impacts of climate change on the dispersion and transformation of air pollutants and on the tolerance of ecosystems against air pollution are important topics of future research.

Yhteenveto

Tässä tutkimuksessa kehitettiin ilmansaasteongelmien alueellisen ja paikallisen mittakaavan mallintamista. Kaukokulkeutuvista ilmansaasteista johtuvat happamoituminen, alailmakehän korkeat otsonipitoisuudet ja rehevöityminen ovat laaja-alaisia ongelmia Euroopassa. Tässä työssä kehitettyjä malleja voidaan käyttää kaukokulkeutuvien ilmansaasteiden päästörajoitusten yhdennettyyn arviointiin yhdessä euroopanlaajuisten mallien kanssa. Tutkimuksessa tehtiin myös mallisovellutuksia kansallisen ja kansainvälisen päätöksenteon tueksi.

Työssä kehitetty alueellinen typpiyhdisteiden laskeumamalli DAIQUIRI tuotti samankaltaisia laskeuma-arvioita kuin mittaukset ja yksityiskohtaisempi ilmanlaatumalli. Testien perusteella

DAIQUIRI:n todettiin soveltuvan hyvin typpilaskeuman arviointiin metsävaltaisille alueille. Rannikoilla ja peltovaltaisilla alueilla DAIQUIRI yliarvioi laskeumaa. DAIQUIRI kuvaa Suomen omien päästöjen vaikutuksia yksityiskohtaisemmin kuin Euroopan laajuisessa päästörajoitustyössä käytetyt karkeammat mallit, jotka aliarvioivat päästöjen aiheuttamaa lähilaskeumaa ja niiden ympäristövaikutuksia. Typpilaskeuman alueellinen mallintaminen antoi 9 % – 19 % euroopanlaajuista mallia suurempia arvioita alueista, joilla happamoittavan laskeuman kriittiset kuormitukset ylittyvät.

Työssä kehitettiin myös metodiikkaa arvioimaan kaupunkialueiden NO_x-päästöjen vaikutuksia otsonitasoihin. Työn tarkoituksena oli löytää metodi, joka soveltuisi liitettäväksi euroopanlaajuisiin otsonin yhdennettyihin arviointimalleihin. Korrelaatiokerroin kaupunkialueen ja ympäröivän maaseudun otsonimittausten välillä parani keskimäärin 0,64:stä 0,85:een testatulla metodilla. Korrelaatiokerroin euroopanlaajuisen mallin tulosten ja kaupunkialueen otsonimittausten välillä puolestaan parani keskimäärin 0,37:stä 0,58:aan.

Mallisovelluksissa analysoitiin Kioton pöytäkirjan toteuttavien energiaskenaarioiden vaikutuksia happamoittaviin ja otsonia muodostaviin ilmansaasteisiin sekä Euroopan että Suomen mittakaavassa. Kioton pöytäkirjaa vastaavilla energiaskenaarioilla EU-maiden rikkipäästöt vähenisivät 12–22 % ja NO_x-päästöt pienenisivät 8–12 % (riippuen päästökaupan oletuksista) vuoteen 2010 mennessä verrattuna tilanteeseen ilman CO_2 -rajoitustoimia. EU:n happamoitumisen ja otsoninmuodostumisen välitavoitteet vuodelle 2010 voitaisi Kioton pöytäkirjan täyttävillä energiaskenaarioilla saavuttaa 35–43 % pienemmillä teknisten rajoitustoimien kustannuksilla verrattuna tilanteeseen ilman CO_2 -rajoitustoimia.

Mallisovellutukset Suomelle osoittivat että happamoitumisen haitallisille vaikutuksille alttiina olevien ekosysteemien pinta-ala vähentyi Suomessa tuntuvasti 1990-luvulla. Positiivisen kehityksen odotetaan jatkuvan vuonna 1999 allekirjoitetun YK:n Euroopan Talouskomission Göteborgin pöytäkirjan myötä Suomessa siten, että vuonna 2010 happamoitumiselle alttiina olevien ekosysteemien pinta-ala olisi noin 25 % vuoden 1990 tilanteesta. Syynä positiiviseen kehitykseen ovat olleet Suomen rikkipäästöjen väheneminen noin 65 %:lla 1990-luvun aikana ja samanaikaisesti vähentynyt kaukokulkeuma. Vuonna 1995 Suomen omien päästöjen aiheuttama osuus rikin kokonaislaskeumasta ylitti 20 % ainoastaan suurien päästölähteiden läheisyydessä. Suomen typenoksidi- ja ammoniakkipäästöt sen sijaan eivät ole vähentyneet vastaavasti. Vuonna 1995 Suomen omien päästöjen osuus typenoksidien kokonaislaskeumasta oli 20–50 % (paikasta riippuen) Etelä- ja Keski-Suomessa, ja ammoniakin osalta vastaava osuus oli 20–70 %. Kioton pöytäkirjan toteuttaminen EU:ssa toisi myös Suomelle hyötyjä happamoittavan laskeuman ja kriittisten kuormien ylityksen pienenemisessä. Vähenemä voisi olla noin 8 % enemmän kuin Göteborgin pöytäkirjan päästörajoituksilla.

Työssä kehitettiin myös yhdennettyjen arviointimallien epävamuusanalyysin metodiikkaa ja käytettiin sitä happamoitumisen yhdennetyn arviointimallin epävarmuuksien arviointiin Suomen kohdalla. Analyysi osoitti, että kriittiset kuormitukset ovat Suomessa merkittävin epävarmuutta tuova osatekijä. Lähellä merkittäviä päästölähteitä myös päästöjen, niiden sijainnin ja ilmakulkeutumisen epävarmuudet ovat merkittäviä. Niitä on voitu vähentää tässä työssä kehitetyillä aluellisen tason malleilla ja pistekohtaisilla päästötiedoilla. Kehitettyä laskentametodiikkaa voidaan käyttää myös skenaarioanalyysiin, jossa voidaan verrata eri päästörajoitusvaihtoehtojen vaikutuksia ekosysteemien suojelun todennäköisyyteen.

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