INTEGRATED MONITORING AND ASSESSMENT OF AIR POLLUTION

Doctors dissertation (DSc) 2009

NATIONAL ENVIRONMENTAL RESEARCH INSTITUTE
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Abstract:	Improved quality, better understanding of processes and optimisation of allocated resources, these are the main advantages of applying Integrated Monitoring and Assessment (IMA) in air quality management. The IMA is defined as the combined use of measurements and model calculations. The use of IMA is demonstrated with examples with different aims: to obtain data for air pollution in urban streets, to assess human exposure to traffic air pollution, and to assess atmospheric deposition of nitrogen compounds to marine and terrestrial ecosystems.
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Contents

Preface

This report is submitted as a thesis for the Danish doctoral degree in natural science. The report summarizes a significant part of the research I have taken part in during 1987 to 2008. During this time period, I have been working as researcher in what is today Department of Atmospheric Environment (ATMI), National Environmental Research Institute (NERI), University of Aarhus.

I have had the pleasure to work with many dedicated and highly skilled researchers during these 21 years. I will not be able to list and acknowledge all, but I owe special thanks to those that have played a special role for me and for the work that forms the basis of this thesis.

- Ruwim Berkowicz was my mentor at NERI in the first years, and gave me the basic training as researcher in atmospheric sciences. Ruwims work has been crucial for the development of the air pollution models at NERI.
- Hans Flyger, Head of Institute (until 1989). He encouraged me to start my PhD study.
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- Øystein Hov was a great inspiration and a dedicated supervisor on my PhD study.
- Jesper Christensen played an important role in the modelling work under the Marine Research Programme Sea90.
- Ole Raaschou-Nielsen, the Danish Cancer Society, involved me in traffic exposure assessment, which initiated our work in this area.
- Finn Palmgren took part in the analysis of traffic pollution in Danish urban streets.
- Mads F. Hovmand invited me to take part in the Background Air Quality Monitoring Programme (BOP), calculating dry depositions from concentrations of nitrogen compounds.
- Steen Solvang Jensen designed the AirGIS system for modelling human exposure to air pollution as a part of his PhD study.
- Henrik Skov took over the BOP, and together we extended the calculations to include nitrogen depositions to all Danish marine waters.
- Thomas Ellermann was the next to take over the BOP. Together we extended and refined the model calculations, and had many valuable discussions on the interpretation of the results.
- Lise Marie Frohn took part in developing routines for the modelling work under the BOP, and the environmental economy system EVA.
- Carsten Ambelas Skjøth took part in the modelling work under the BOP. He designed the routines for simulating ammonia emissions.
- Steen Gyldenkærne contributed with crucial knowledge about agricultural praxis for the routine for ammonia emissions.
- Per Løfstrøm developed OML-DEP in the Danish Ammonia Modelling System (DAMOS).
- Camilla Geels played a central role in the work with DAMOS and the procedures for regulating ammonia emissions from livestock farms.
- Matthias Ketzel took part in the particle modelling in OSPM and the exposure scenarios.
- Martin Hvidberg played a strong role in the work with AirGIS and the exposure scenarios.
- Lars Moseholm, our current Head of Department encouraged me to write this thesis, and has given me valuable points and basis for discussions.

I would like to thank Prof. Ole John Nielsen, Copenhagen University for moral support during writing of the thesis. Prof. Steffen Loft, Dr. Mette Sørensen, Dr. Zorana Andersen Copenhagen University, and Prof. Torben Sigsgaard and Prof. Herman Autrup, Aarhus University took part in the studies on health effects of air pollution. Dr. Gerrit de Leeuw TNO, Dr. Tim Jickells and Dr. Lucy Spokes, University of East Anglia, Dr. Heinke Schluenzen, Dr. Michael Schulz, Dr. Elke M.I. Meyer, Dr. Susanne Tamm, Hamburg University, Dr. Lise Lotte Sørensen, Risø (now NERI-ATMI), Dr. Britta Pedersen NERI-MAR took part in the air-sea exchange studies. Dr. Elisabetta Vignati took part in the Children cancer and the air-sea exchange studies. Finally I would like to thank the entire staff at NERI for a great working environment.

The report summarises work carried out within national and international research and advisory projects, as well as activities under the Danish air quality monitoring programmes (Table 1.1). The completion of three of the journal articles and the writing of this report was financially supported by a grant from the Carlsberg Foundation. Together with funding from NERI, this grant allowed me to dedicate the necessary time for writing this thesis report.

I dedicate this thesis to my wife Pernille, and my children Samuel and Johanna.

Ole Hertel, September 2009

Table 0.1 This thesis report summarises work carried out within a number of research and advisory projects as well as the Danish Air Quality Monitoring Programmes. The most important projects and programmes as listed in this table together with the time period they took place (or in which the activities are relevant in context of this thesis) and the source of funding.

Time period	Name/description	Funding
1989 - 1991	The Nordic Calculation Method for Vehicle exhaust (In Swedish: Den Nordiske Berekningsmetod for bilavgasser)	Nordic Council of Ministers (NMR)
1989 - 1994	The Danish Marine Research Programme Sea90	The Danish Environmental Protection Agency
1990 - 1995	Transport and transformation of nitrogen and sulphur compounds in the marine boundary layer – my PhD project	NERI and the Danish Research Academy. A travel grant was provided by the Nordic Council of Ministers
1990 - 2000	The Danish Urban Air Pollution Monitoring Programme (In Danish: Landsmåleprogrammet for luftkvalitet (LMP)	Danish Ministry of the Environment with funding from the Danish Law of Finances
1992 – 2004	Danish Environmental Research Programme (In Danish: Det Strategiske Miljøforskningsprogram): Centre for Air Pollution Processes and Models (1992-1996), Centre for Biochemical and occupational Epidemiology (1992 – 1997), Centre for Environment and the Respiratory System (In Danish: Center for Miljø og Luftveje - CML) (1998 – 2002), Centre for Transport Research on environment and health Impact and Policy (TRIP) (2000 – 2004)	An inter-ministerial environmental research programme with funding from the Danish Law of Finances
1994 - 2004	The Danish Background Air Quality Monitoring Programme (In Danish: Baggrundsovervågningsprogrammet for luftkvalitet (BOP))	Danish Ministry of the Environment with funds from the Danish Law of Finances
1995 - 1999	The Traffic Monitoring Program (In Danish: Trafik Overvågningsprogrammet (TOV))	Danish Ministry of Traffic with funds from the Danish Law of Finances
1993 - 1996	The Air-Sea Exchange Process Studies (ASEPS)	Office of Naval Research (ONR), Washington DC, USA
1997 - 2000	Atmospheric Nitrogen Input to the Coastal Ecosystem (ANICE)	Research grant from EU under the fifth framework programme
1999 - 2000	The Impact on NO_2 levels in Danish Urban Streets of introducing CRT particle filters on heavy duty vehicles	The Danish Road Safety and Transport Agency
1999 - 2004	The Marine Ecological response to Atmospheric nitrogen Deposition (MEAD)	Research grant from EU under the fifth framework programme
2000 - 2004	Centre for Environment Associated Cancer (In Danish: Center for Miljørelateret Kræft (CEMIK))	Danish Ministry of Interior and Health, Research Centre for Environmental Health
2004 - 2008	The Research Centre of Excellence AIRPOLIFE (AIR POllution in a LIFEtime perspective)	The Danish Research Council
2005 - 2008	The Ammonia and Odour projects in the Research Programme under the Danish Aquatic Action Plan III (In Danish: Forskningsprogram under Vandmiljøplan III (VMP-III)).	The Danish Ministry of Food and Agriculture with funding from the Danish Law of Finances
2008	Nitrogen load of nature areas in Eastern Jutland	Environment Centre Aarhus, Danish Ministry of the Environment
2008	Nitrogen load of natures on Zealand and Bornholm	Environment Centre Roskilde, Danish Ministry of the Environment

Dansk sammenfatning

Denne afhandling berører to centrale emner inden overvågning og regulering af luftkvalitet. Det første emne relaterer sig til helbredsmæssige effekter af luftforurening, og drejer sig om overvågning og vurdering af luftforurening fra trafikken samt bestemmelse af befolkningens udsættelse for luftforurening. Det andet emne relaterer sig til den atmosfæriske afsætning af kvælstofforbindelser til den terrestriske og marine natur. Til bestemmelse af miljøbelastningen inden for disse to områder er der udviklet og anvendt en række enkle operationelle luftkvalitetsmodeller.

Målet for denne afhandling har været at demonstrere at ″integreret overvågning og vurdering er et stærkt og effektivt redskab inden for operationel luftforureningsregulering″. I denne sammenhæng er ″integreret″ anvendt som udtryk for den rutinemæssige kombinerede anvendelse af målinger og modelberegninger. Jeg definerer her ″operationel″ som effektiv, pålidelig og realistisk. En sidegevinst ved integreret overvågning og regulering er nødvendigheden af et tæt samarbejde mellem modellører og målefolk. Det er min hypotese at ″modelberegninger er uundværlige til fortolkning af og som et tillæg til målinger i felteksperimenter og rutinemæssige overvågningsprogrammer″.

I forbindelse med vurdering af luftforurening fra trafik er der inden for det præsenterede arbejde udviklet to luftkvalitetsmodeller: ″Operational Street Pollution Model″ (OSPM) og ″Urban Background Model″ (UBM). OSPM repræsenterer fortsat state-of-the-art inden for operationelle modeller for luftkvalitet i bygader. Modellen har opnået en betydelig udbredelse i Europa, og er i dag et integreret element i det danske landsdækkende luftkvalitetsovervågningsprogram (LMP) for danske byområder. Inden for overvågningsprogrammet anvendes OSPM til kortlægning af luftforurening i bygader, hvor der ikke foretages målinger. Modellen anvendes imidlertid ligeledes til scenarieberegninger for at vurdere den fremtidige udvikling og til vurdering af effekten af forskellige reguleringer. Et eksempel er undersøgelser af betydningen af indførelse af CRT filtre på tunge køretøjer. Filtrene reducerer partikeludslippet, men kan føre til en øget andel af kvælstofdioxid i NO_x udslippet.

Trafikken udgør i dag den væsentligste kilde til befolkningens udsættelse for luftforurening i Danmark såvel som i en række andre lande. Dette har været baggrunden for at anvende OSPM til eksponeringsvurderinger. I de tidlige studier blev de nødvendige input data indsamlet enten manuelt eller gennem personlige henvendelser og spørgeskemaer udsendt til de lokale myndigheder. Denne metode er imidlertid ikke mulig, når der er tale om meget store kohorter. Disse problemer blev løst gennem udviklingen af AirGIS systemet. AirGIS gør brug af GIS-baserede værktøjer, digitale kort samt informationer fra de danske centrale registre. AirGIS er et unikt system og netop været anvendt på 200.000 adresser for 57.000 personer inden for Kræft, kost og helbredskohorten (etableret af Kræftens Bekæmpelse).

Inden for vurdering af den atmosfæriske afsætning af kvælstofforbindelser til naturen er der udviklet en Lagrangiansk transport-kemi model – Atmospheric Chemistry and Deposition (ACDEP) model. ACDEP blev udviklet inden for det marine forskningsprogram Hav90 og var gennem 10 år det anvendte værktøj inden for baggrundsovervågningsprogrammet til kortlægning af atmosfærisk afsætning af kvælstof og svovlforbindelse til hav og landområder i Danmark. ACDEP blev inden for en række EU projekter anvendt til beregninger af den atmosfæriske kvælstofbelastning af bla. Nordsøen og Østersøen samt til vurdering af den atmosfæriske belastnings betydning for algeopblomstringen i kystnære farvande. Den numeriske metode udviklet til løsning af kemiske reaktioner i modellen er internationalt blevet fremhævet som en metode med høj effektivitet i forhold til kravet til computer ressourcer. Med adgang til stadig mere regnekraft er det nu muligt at anvende tungere men også mere præcise modeller end ACDEP.

I nærheden af landbrugsbedrifter med husdyrhold kan den lokale afsætning af ammoniak udgøre en meget væsentlig del af den samlede atmosfæriske kvælstof afsætning. Dette har ført til udviklingen af DAMOS (Danish Ammonia Modelling System) til beregning af afsætningen kvælstof i områder med intensiv landbrugsproduktion. I den første udgave af DAMOS bestod systemet af en kombination af ACDEP og røgfanemodellen OML-DEP. OML-DEP er udviklet på baggrund af OML modellen, som i Danmark anvendes til bestemmelse af skorstenshøjder på afkast fra industri og kraftværker.

Lokale udslip af ammoniak har i Danmark en sæsonvariation, der i stort omfang styres af den lokale landbrugspraksis; en praksis som igen i stort omfang er dikteret af den danske lovgivning. De centrale registre over husdyrhold, dyrkningen af afgrøder på markerne samt placering af gårde og marker har gjort det muligt at udvikle en detaljeret opgørelse af danske ammoniak udslip. Metoden repræsenterer state-of-the-art på området, og i dag ved at blive implementeret i den internationalt anerkendte og den generelt i Europa meget anvendte EMEP model.

Lagrangianske modeller som ACDEP har deres begrænsninger i forhold til beskrivelsen af den atmosfæriske transport. Det har ført til et skifte fra ACDEP til den Eulerske DEHM (Danish Eulerian Hemispheric Model). Dette skifte er foretaget inden for såvel baggrundsovervågningsprogrammet som inden for DAMOS. DEHM anses i dag at udgøre state-of-the-art inden for beskrivelsen af den atmosfæriske transport.

DAMOS har været anvendt til vurdere belastningen med atmosfærisk kvælstof i en dansk region med relativt lave belastninger. Disse studier har givet mulighed for at vurdere effekten af lokale reguleringsindgreb. DAMOS har været anvendt til at vurdere effekten af at etablere bufferzoner omkring følsomme danske naturområder. Endvidere har beregninger med OML-DEP/DAMOS dannet grundlaget for revisionen af metoden til vurdering af lokale afsætning af kvælstof relateret til ændringer i husdyrproduktionen. En metode som nu anvendes af kommunerne ved vurderinger af ansøgninger fra landmændene om øget/ændret husdyrproduktion.

Integreret overvågning og vurdering (IOV) af luftforurening er i dag et veletableret koncept ved DMU-ATMI. De anvendte procedurer og metoder er resultatet af et arbejde, som er gennemført over de seneste 20 år. I anvendelsen af IOV på DMU-ATMI anvendes målinger til vurdering af:

- Aktuelle koncentrationer og/eller afsætninger af forureninger ved målestederne
- Sæsonvariationer i forureningsbelastning
- Langtidsudvikling i koncentrationer og/eller afsætninger af forurening
- Kildeallokering
- Validering og udvikling af modeller

Denne type af information kan ikke udledes af modelberegninger, idet sådanne beregninger er afhængige af pålideligheden af input data og anvendte procesbeskrivelser. Pålideligheden af såvel input data som procesbeskrivelser kan ændre sig over tid. Inden for IOV på DMU-ATMI anvendes modeller til at give information om:

- Den geografiske fordeling i forureningsbelastningen.
- Fordelingen mellem bidrag fra lokale og regionale kilder.
- Scenarier og prognoser, samt effekten af forskellige reguleringsstrategier.

Udviklingen og anvendelsen af IOV ved DMU-ATMI har øget kvaliteten i de gennemførte luftkvalitetsstudier og bla gjort det muligt at foretage:

- Kortlægning af luftkvalitetsniveauer i bygader, hvor der ikke foretages målinger.
- En vurdering af effekten af miljøzoner omkring centrale områder i byerne.
- Analyser af effekten på kvælstofdioxid forureningen af at indføre partikelfiltre.
- En fastlæggelse af udsættelsen for luftforurening for store befolkningskohorter
- En kobling af eksponeringsdata til diverse helbredsdata.
- En årlig kortlægning af atmosfærisk kvælstof afsætning til den samlede danske terrestriske og marine natur.
- En bestemmelse af kvælstofafsætningen fra enkelte gårde.
- Kildeallokering for lokal såvel som regional forurening i Danmark.

IOV er kommet til gennem en gradvis udvikling over en lang periode, og må betragtes som en betydelig succes i DMU-ATMI. Fordelene ved IOV er forbedret kvalitet af overvågningen og vurdering af luftkvalitet, samt en forbedret forståelse en de processer som er styrende for luftkvaliteten i Danmark. IOV betyder en mere optimal udnyttelse af ressourcerne, siden mere information trækkes ud af undersøgelserne og fortolkningen af målingerne forbedres.

IOV stiller krav til dokumentationer og validering af de værktøjer som indgår; Krav som på mange måder modsvarer de krav man stiller i forbindelse med målinger i felt- og overvågningsprogrammer. Modellerne ved DMU-ATMI er primært dokumenteret gennem internationale artikler, men også i tekniske rapporter.

Denne afhandling har demonstreret, at IOV er et stærkt og helt nødvendigt værktøj inden for miljøvurderinger af luftforurening. De viste eksempler har demonstreret at modelberegninger er uvurderlige til fortolkning af og som supplement til målinger i feltstudier og overvågningsprogrammer. Endvidere har IOV sidegevinst at modellører og målefolk kommer til at arbejde tæt sammen.

1 Introduction

Air pollution has a variety of negative effects on climate, human health and nature. Climate is affected by releases to the atmosphere of particles and trace gases that change the radiation balance. Adverse health effects in the population are the result of short-term as well as long-term exposure to air pollution. Nature is affected by atmospheric deposition of acid gases and aerosols that in certain areas leads to acidification of lakes and terrestrial ecosystems. Loss of biodiversity may be the long-term results of high atmospheric nitrogen depositions that lead to eutrophication of sensitive terrestrial and marine ecosystems. Exposure to ozone affects the growth of the vegetation, and makes it more vulnerable to other types of stress.

These different negative effects of air pollution are well-known and most of them have been explored for several decades. However, in recent years it has been clear that some of the effects may take place at much lower pollutant loads than previously believed. These findings are the results of an improved understanding of the governing physical, chemical and biological processes; an understanding that has been obtained as a result of the access to more accurate and detailed data as well as better tools for the analyses. Many pollutants are now measured with high accuracy at low concentrations and/or with high temporal resolution. Pollutants that could not previously be measured are now in some cases included in routine monitoring programmes. The increasing available computer power has made it possible to develop air pollution models with still higher spatial and temporal resolutions. These models are very useful as advanced tools in the analysis of air pollution measurements.

The environmental problems addressed in this thesis concern two important topics in air pollution management. The first topic relates to health effects associated with air pollution, and concerns the monitoring and assessment of air pollution from traffic as well as assessment of human exposure to air pollution. The second topic relates to eutrophication of nature, and concerns the monitoring and assessment of the atmospheric deposition of nitrogen compounds to terrestrial and marine ecosystems. The modelling part of these studies is based on the application of strongly parameterised air quality models.

1.1 Policy context

In the late 1970ties and early 1980ties there was a growing concern in the population for the various environmental problems associated with anthropogenic emissions. A number of NGO's – like WWF, Danish NOAH and Green Peace - concerned with environmental issues were established, and they quickly received many members. The political parties defined environmental policies to protect nature, natural resources and health. The Ministry of the Environment was formed in 1971, and in the following years the parliament launched a number of national strategies to reduce anthropogenic pollutant loads of health and environment. Among the important issues on the environmental agenda in these years were the increasing eutrophication problems observed in nature in the Northern countries. At this time, focus in Denmark was mainly on the aquatic ecosystems; As a result of large nutrient inputs, many of the Danish lakes and streams were more or less dead, and turnovers were becoming still more frequent in the Danish coastal waters. In the cities there was a growing concern in the public about the health problems related to the traffic pollution from the increasing car fleet.

In 1982 the Danish Air Quality Monitoring Programme (LMP) was launched as the first nation-wide urban air quality monitoring programme in Denmark. The intension was to monitor levels and trends in gaseous and particulate air pollution in the major Danish cities in order to protect human health. Since the initiation in 1982, the programme has been significantly revised three times and now consists of monitoring stations at kerb side and in urban background in the four largest cities: Copenhagen, Aarhus, Aalborg and Odense. Since the beginning of the 1990'ties the measurements have been closely linked to modelling activities concerning kerb side and urban background pollution.

In 1987 came the first Danish Aquatic Action plan (VMP-I). The goal was a 63.000 tonnes reduction in the nitrogen load of Danish marine waters. The implementation of the action plan created a need for monitoring the development in the nitrogen loads of the Danish waters, and for developing tools for evaluating the needs for further actions. In 1989 the Marine Monitoring Programme (VMOP, later NOVA and now NOVANA) was established, and in 1990 the Danish EPA initiated the Danish Marine Research Programme Sea90. The NOVA programme included an atmospheric component for monitoring ambient concentrations and depositions of nitrogen compounds to the Danish marine waters. Similarly the Sea90 research programme had an atmospheric component for improving the understanding of the air-sea exchange processes of nitrogen compounds, and for the development of a mathematical model for mapping atmospheric nitrogen deposition to Danish marine waters.

1.2 Science context

In the late 1970ties and early 1980ties, ATMI build a strong expertise in atmospheric transport and dispersion modelling. A significant effort was put into the development of the OML plume model and its meteorological pre-processor. Most plume models at that time were based on calculations for discrete stability categories, and a subsequent statistical handling of the results. The OML model was based on hour by hour calculations using time series of dispersion parameters computed on the basis of atmospheric stability parameters derived from hourly meteorological data.

Furthermore, ATMI developed strong expertise in numerical handling of stiff systems of partial differential equations – e.g. solving large and complex equation systems with high precision and with high computational efficiency. This expertise was e.g. established during the 1980ties in the development of the long-range transport model – The Danish Eulerian Model (DEM). ATMI had limited expertise in modelling atmospheric chemistry, and this was part of the background for establishing a close cooperation with Norwegian modellers in these years.

1.3 Objective

Some air pollution models are mainly aimed at performing studies to improve our understanding of the governing physical and chemical processes in the atmosphere; whereas other models are strongly parameterised and aimed at application in monitoring and assessment studies in relation to environmental management. Model calculations may be used in assessments of the pollution loads of nature as well as in assessment of human exposure to air pollution. The models may furthermore be used to extend the geographic coverage of the air pollution data obtained from measurements. In addition they can provide information about source-receptor relationships, and they may be used for scenario studies in the evaluation of abatement strategies or for providing prognoses for the future trends in pollution loads. All these types of model applications are demonstrated in this thesis work.

The aim of the thesis is to demonstrate that "integrated monitoring and assessment is a strong and efficient tool in operational air pollution management". In this context the word "integrated" is used for the steady combined use of measurements and model calculations. I define "operational" as efficient, reliable and realistic. The trade off in integrated monitoring and assessment is that modellers and experimentalists have to work closely together. It is my hypothesis "that model calculations are indispensable for use in interpretation of, and as an extension to measurements in field experiments and routine monitoring programmes".

I will support the above stated hypothesis through a series of examples of results obtained in research and advisory projects carried out over the past 20 years.

1.4 Outline of the thesis

This thesis report consists of a main body of five technical chapters and an appendix containing twelve scientific papers (one conference proceeding and eleven peer-reviewed journal articles). The twelve scientific papers are listed in Table 1.1, whereas the five main chapters are shortly outlined below.

The descriptions of integrated monitoring and assessment in the five main chapters (and supported by papers I - XI), is further supported by the conceptual paper (Paper XII) on Integrated Monitoring within the Danish Air Quality Monitoring Programmes.

1.4.1 Air pollution from traffic

The first main chapter (Chapter 2) concerns the assessment of air pollution from traffic. Focus is here on the urban environment where traffic usually constitutes the main source of enhanced air pollution levels in local hot spots (typically inside the trafficked streets). This chapter addresses the following environment questions:

A.1. How is the distribution between pollution contributions from local traffic in the single street, from sources in the urban area in general and from sources in more remote areas (including long-range transport)?

A.2. Do we comply with current and future air quality guidelines for urban streets?

A.3. What are the impacts of certain emission reduction strategies concerning traffic air pollution?

A central part of this work concerns the development of a model for air pollution concentrations in urban streets (described in detail in Papers I and II). A specific monitoring programme is designed for following the trends in traffic air pollution concentrations and emissions. The impact on nitrogen dioxide concentrations of using particle filters on diesel vehicles is investigated in a scenario study. The chapter furthermore addresses the following more technical questions:

B.1. How good is the performance of the simple parameterised street pollution model?
B.2. To what extend do we have access to the necessary input data at the demanded level of quality?
B.3. To what extend do we have access to air pollution measurements for testing the model for various pollutants and various types of streets?

1.4.2 Human air pollution exposure

The second main chapter (Chapter 3) concerns the assessment of human exposure to air pollution. Focus is also on traffic pollution that constitutes the main source of human air pollution exposure in most of the industrialised countries today. This chapter addresses the following environmental questions:

A.4. Does the exposure to air pollution constitute a significant health hazard for the population?
A.5. What is the actual range in air pollution exposures of the population?
A.6. To what extent is it possible to reduce the air pollution exposure by selecting a low exposure route through the city?

The street pollution model is tested as a tool in a couple of human air pollution exposure studies (supported by Papers III and V). A GIS based model system is developed for generating some of the necessary input data (described in more detail in Paper IV). The exposure model system is applied in a scenario to investigate the importance for the human air pollution exposure of following various routes through the city (described in detail in Paper VI). The chapter furthermore addresses the following technical questions:

B.4. Is it possible based on the street pollution model to construct a human exposure model system suitable for application to large cohorts in epidemiological studies?
B.5. Do we have access to the necessary input data for applying such systems in larger epidemiological studies?

1.4.3 Atmospheric nitrogen deposition

The third main chapter (Chapter 4) concerns the assessment of atmospheric deposition of nitrogen compounds to nature. Nitrogen is an important nutrient for the flora in terrestrial and marine ecosystems.

In this chapter focus is on the tools and methods in assessment of atmospheric nitrogen deposition. To some extend this chapter is an introduction to the two following chapters. The chapter describes the development of a strongly parameterised model for assessment of atmospheric deposition of reactive (and bio-available) nitrogen compounds (described in detail in Paper VII). A model system combining regional scale and local scale transport is developed for use in assessment of atmospheric nitrogen deposition in relation to regulation of ammonia from local livestock farms. The chapter addresses the following more technical questions:

B.6. How well does the combined regional scale and local scale model system work?
B.7. How important is the spatial and temporal variation in ammonia emissions for a proper assessment of the local atmospheric nitrogen deposition?

1.4.4 Nitrogen deposition on regional scale

The fourth of the main chapters (Chapters 5) concerns the assessment of atmospheric nitrogen deposition on regional scale. This chapter addresses the following environmental questions:

A.7. Does the atmospheric nitrogen deposition play a significant role in eutrophication – including algae blooming – of the marine waters?
A.8. To what extend are terrestrial ecosystems threatened by atmospheric nitrogen deposition?
A.9. How important are the Danish emissions for the atmospheric nitrogen deposition, and is the long-range transport contribution alone sufficiently high to exceed critical loads of Danish nature?
A.10. How large is the atmospheric nitrogen deposition to the Baltic Sea and the North Sea, respectively?

The chapter describes the assessment of atmospheric nitrogen deposition in the marine research programme. The nitrogen depositions to the Baltic Sea and the North Sea are computed. The stepwise integration of model calculations in the Background Air Quality Monitoring Programme

(BOP) is outlined. The chapter addresses the following technical question:

B.8. How well does the parameterised model describe the transport and deposition of atmospheric nitrogen compounds?

1.4.5 Nitrogen deposition on local scale

The fifth of the main chapters (Chapter 6) concerns the assessment of atmospheric nitrogen deposition on local scale. This concerns especially the agricultural emissions of ammonia from livestock production. The chapter addresses the following environmental questions:

A.11. Is it possible at regional scale (i.e. in a Danish county) with moderate loads to reduce the atmospheric nitrogen loads below critical loads through regulation of local sources?

A.12. Are buffer zones with restricted ammonia emissions around local nature areas an efficient way to regulate the atmospheric nitrogen load?

A suggested procedure for assessment of atmospheric nitrogen deposition from local livestock farms is outlined. The chapter is supported by the review on local scale modelling of atmospheric nitrogen deposition provided in Paper XI. The chapter furthermore addresses the following technical question:

B.9. What is needed in order to develop an easy to apply assessment system for use in regulation of ammonia emissions from livestock farms?

1.4.6 Conclusions and perspectives

The main conclusions from the thesis work are outlined in Chapter 7. The various questions addressed in this introduction are answered on basis on the presented examples from research and advisory projects. At the end of the report, the perspectives in integrated monitoring and assessment at NERI are outlined in Chapter 8.

Table 1.1 The scientific papers included in this thesis and attached in the appendix to the report. In the report these papers are referred in the same way as other references, but in addition the number of the paper is indicated in the text as" (Paper xx)".

Paper I: Berkowicz, R., Hertel, O., Sørensen, N.N. and J.A. Mikkelsen, 1997. Modelling Air Pollution from Traffic in Urban Areas –Pp 121-141, In: R.J. Perkins and S.E. Belcher (Eds.) Flow and Dispersion through Groups of Obstacles, 249 p., Clarendon Press, Oxford 1997.

Paper II: Berkowicz, R., Palmgren, F., Hertel, O., and Vignati, E., 1996. Using measurements of air pollution in streets for evaluation of urban air quality - meteorological analysis and model calculations. Science of the Total Environment, 189/190, 259-265

Paper III: Raaschou-Nielsen, O., Hertel, O., Vignati, E., Berkowicz, R., Jensen, S. S., Larsen, V.B., Lohse, C., and Olsen, J.H., 2000. Evaluation of an air pollution model with respect to use in epidemiologic studies; comparison with measured levels of Nitrogen Dioxide and Benzene. Journal of Exposure Analysis And Environmental Epidemiology, 10, 4-14.

Paper IV: Jensen, S. S., Berkowicz, R., Hansen, H. S., and Hertel, O., 2001. A Decision-support GIS tool for Management of Urban Air Quality and Human Exposures. Transportation Res. Part D: Transport and Environment, 6(4), 229-241.

Paper V: Hertel, O., de Leeuw, F.A.A.M., Raaschou-Nielsen, O., Jensen, S.S., Gee, D., Herbarth, O., Pryor, S., Palmgren, F., and Olsen, E., 2001. Human Exposure to Outdoor Air Pollution. IUPAC Technical report. Pure and Applied Chemistry, 73(6), 933-958.

Paper VI: Hertel, O., Hvidberg, M., Stausgaard, L., and Storm, L., 2008. A proper choice of route significantly reduces air pollution exposure – A study on bicycle and bus trips in urban streets. Science of the Total Environment, 389(1), 58-70.

Paper VII: Hertel, O., Christensen, J., Runge, E.H., Asman, W.A.H., Berkowicz, R., Hovmand, M.F., and Hov, Ø., 1995. Development and Testing of a new Variable Scale Air Pollution Model - ACDEP. Atmospheric Environment, 29, 11, 1267-1290.

Paper VIII: Hertel, O., Ambelas Skjøth, C., Frohn, L. M., Vignati, E., Frydendall, J., de Leeuw, G., Schwarz, U., and Reis, S., 2002. Assessment of the Atmospheric Nitrogen and sulphur Inputs into the North Sea using a Lagrangian model. Physics and Chemistry of the Earth, 27(35), 1507-1515.

Paper IX: Hertel, O., Ambelas Skjøth, C., Brandt, J., Christensen, J., Frohn, L. M., and Frydendall, J., 2003. Operational Mapping of Atmospheric Nitrogen Deposition to the Baltic Sea. Atmospheric Chemistry and Physics, 3, 2083-209.

Paper X: Ambelas Skjøth, C., Hertel, O., Gyldenkærne, S., and Ellermann, T., 2004. A dynamical emission parameterisation part II: Implementation in ACDEP and test of performance. Journal of Geophysical Research, 109, D06306.

Paper XI: Hertel, O., Ambelas Skjøth, C., Løfstrøm, P., Geels, C., Frohn, L.M., Ellermann, T., and Madsen, P.V., 2006. Modelling nitrogen deposition on local scale – a review of state-of-the-art. Environmental Chemistry, 3(5), 317-337.

Paper XII: Hertel, O., Ellermann, T., Palmgren, F., Berkowicz, R., Løfstrøm, P., Frohn, L.M., Geels, C., Ambelas Skjøth, C., Brandt, J., Christensen, J., Kemp, K., and Ketzel, M., 2007. Integrated Air Quality Monitoring - Combined use of measurements and models in monitoring programmes. Environmental Chemistry, 4(2), 65-74.

2 Air pollution from traffic

The air pollution in an urban environment is the result of local emissions as well as contributions from pollution transport from both nearby sources inside and remote sources outside the city (Figure 2.1). The size of the city domain and the emission density governs the urban area's contribution to the local pollution level (Berkowicz, 2000a). The distribution between contributions from different source types and source areas inside and outside the urban area to a given site vary between pollutants. Furthermore, the pollutant level has a temporal variation which is a function of variations in local releases as well as in the meteorological parameters governing the transport and dispersion conditions (Berkowicz et al., 1997b) (Paper I).

Pollution released from tall sources is most of all transported out of the urban area before being dispersed down to ground level. Industries, power plants and other sources with releases from tall chimneys contribute only rarely to the local pollutant concentrations at ground level inside the urban areas. Pollution from tall sources contributes therefore primarily to the more regional pollution.

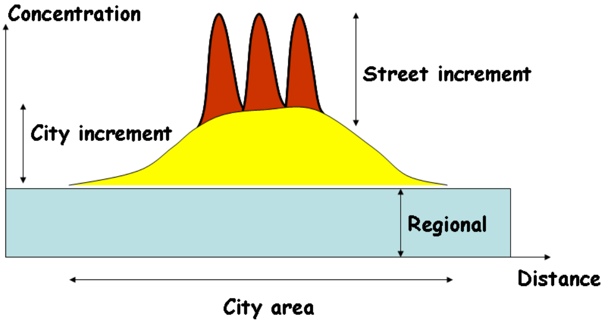


Figure 2.1 A schematic illustration of the air pollutant contribution from regional transport, the city area and the street traffic. The relative magnitude of the various contributions depends on the considered pollutant and the actual dispersion conditions (governed by the meteorology).

Pollutant emissions related to vehicle transport, local domestic heating and smaller industries have low release heights, for which the emission is not diluted as efficiently as it is usually the case for emissions released from tall sources. Thereby, these sources contribute significantly to the pollutant concentrations at ground level. Furthermore, these releases take place close to where the population reside, and they therefore contribute significantly to human air pollution exposures. The emissions from road traffic follow in general a fixed pattern through out the day and through out the week. However, due to variations in the meteorological conditions, this is not necessarily the case for the resulting pollution levels. In general, the dilution increases with wind speed, especially in urban areas where the highest concentrations generally appear at low wind speeds (below 2 m/s).

2.1 Air pollution in urban streets

The trafficked streets are the hot spot areas in the urban environment (Figure 2.1), which is illustrated in the ranges shown in Tables 2.1 and 2.2. Besides the emissions that are taking place inside the street, the air pollution in an urban street is to a high extent governed by the physical conditions surrounding the street. The special airflow inside streets and around buildings may result in very different concentrations on different locations in the street. This is illustrated for a street canyon in Figure 2.2. Street canyons are characterised by the presence of tall buildings on both sides of the street. When the wind blows perpendicular to the street, the pollution concentrations may be up to 10 times higher on the leeward side of the street compared to the windward side (Berkowicz et al., 1996) (Paper II).

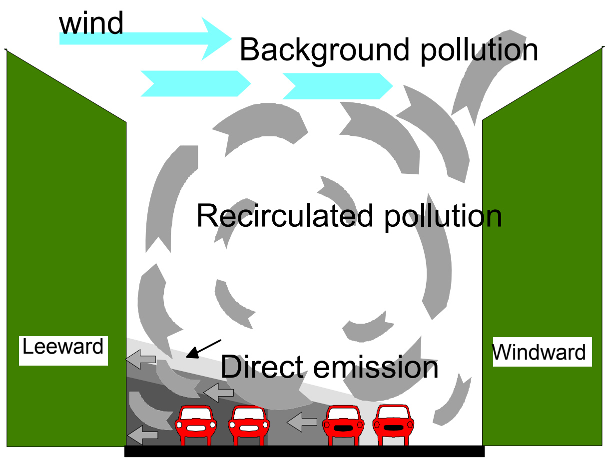


Figure 2.2 Illustration of the flow and dispersion inside a street canyon. In the shown situation, the wind above roof level is blowing perpendicular to the street. Inside the street canyon a vortex is created, and the wind direction at street level is opposite to the wind direction above roof level. Pronounced differences (they may be up to a factor of 10) in air pollution concentrations on the two pavements is the result of these flows. Source: (Berkowicz, 1998).

Street canyons have in general higher pollution concentrations compared with more open streets or street sections with a similar traffic density. The open streets are generally windier due to the lack of buildings to provide a shield for the wind. In the street canyon the pollution is recirculated instead of being transported away from the street. The dispersion due to traffic induced turbulence has been found to be very important for the urban street pollution levels at low wind speeds (Hertel and Berkowicz, 1989c; Kastner-Klein et al., 2000). The higher the driving speed of the vehicles in the street, the quicker the pollution is mixed with 'clean air' of the surroundings. This also means that the relationship between traffic intensity and air quality is non-linear.

Urban air quality monitoring programmes are designed to follow trends in the general pollutant levels as well as in hot spots – typically the most trafficked streets. The programmes are naturally constrained by the available resources. Measurements are therefore often limited to one urban background site and a limited number (in the Danish cities one or two) of street stations. The impact of the complex flow conditions in the urban streets combined with large variations in traffic means that huge differences in pollutant levels may be observed between nearby streets and in some cases even between street sections of the same street (Berkowicz et al., 1997b) (Paper I). Measurements performed at one or two street stations in a monitoring programme may therefore not reflect the worst case conditions in the urban area. In this context, street pollution models are useful tools in the assessment of air quality in those streets where measurements are not carried out. These models are at the same time useful in air quality management for scenario studies for predicting the future pollution loads or for looking at the impact of various regulations of traffic or pollutant emissions. The development of the Operational Street Pollution Model (OSPM) is the result of a wish for access to such a tool for use in urban air quality management.

2.2 Air pollution levels in Denmark

Denmark is placed in the region between the polluted central European areas and the much less polluted Scandinavia (Grønskei, 1998). The air pollutant levels are therefore generally in the low end of the range for those pollutants that have a significant long-range transport component. However, the local hot spots especially in trafficked urban streets may still reach significant levels.

For annual mean concentration of particle mass, the long-range transport component in Denmark is in the order of 20µg/m^3, the contribution from the urban area only in the order of 1 to 2µg/m^3 (Palmgren et al., 2005), whereas the contribution from traffic in a busy street may be in the range of 5 to 10µg/m^3. The picture is very different when NO_x is considered. The contribution from rural background is in the order of 10 to 15µg/m^3, from sub-urban areas 1 to 5µg/m^3, from urban background 10 to 25µg/m^3 and from traffic in the single streets 50 to 100µg/m^3.

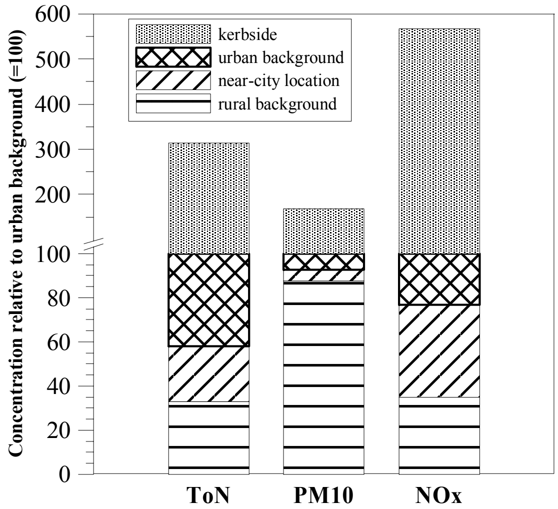


Figure 2.3 Comparison of average concentrations of total particle number (ToN), particle mass (PM10) and NO_x at rural, near-city, urban and kerbside stations relative to urban background levels in the Copenhagen area. The concentration bars are stacked so that only the additional contributions are marked with the pattern shown in the legend. Note that the scale of the vertical axis changes at 100. Source: (Ketzel, 2004), and also presented in (Hertel et al., 2007a) (Paper XII).

These figures are obtained from measurements in the Danish urban air quality monitoring programme, and represent typical annual mean values from recent years (2004 to 2007). If the similar analysis is carried out for particle number concentrations, the distribution is somewhat similar to what is obtained for NO_x. Such a picture has been derived from model calculations based on careful analysis of measurements (see Figure 2.3).

Table 2.1 Measured ranges in annual mean pollutant concentration in the period 2000 to 2007. Unit: (µg/m^3) except for particle number. For NOx the concentration is in (µg NO_2/m^3). Values are derived from measurements within the Urban Air Quality Monitoring Programme. Source: www.dmu.dk

Pollutant	Rural	Sub-urban	Urban	Street
PM	20 - 25	20 - 25	20 - 25	20 - 40
NO_x	10 - 13	13 - 16	25 - 45	75 - 175
NO_2	8 - 11	11 - 13	15 - 25	30 - 40
Particle[a]	-	-	10.000	40.000
CO[b]	-	-	450 - 475	450 - 1000

[a] Number concentration, one years measurements at H.C. Andersen's Boulevard, [b] 8-hours running average on annual basis

The tables 2.1 and 2.2 show for the recent years, the typical range in observed pollutant concentrations at rural, sub-urban, urban background, and street sites. The annual mean NO_x concentrations are a factor of two to three higher in the urban background compared with the rural levels. For NO_2 this range is slightly smaller, and for PM the gradient is only significant when the extreme levels are considered (in this case the 98 percentile). The range between rural and street level concentrations is up to an order of magnitude for NO_x and a factor of four to five for NO_2. For PM there is almost no gradient between the levels in the urban background and the rural sites.

Table 2.2 Measured ranges in 98 percentiles of pollutant concentration in the period 2000 to 2007. Unit: (µg/m^3) except for particle number. For NOx the concentration is in (µg NO_2/m^3). Values are derived from measurements in the Urban Air Quality Monitoring Programme. Source: www.dmu.dk

Pollutant	Rural	Sub-urban	Urban	Street
PM[b]	45 - 55	45 - 58	50 - 60	60 - 90
NO_x	5 - 15	40 - 60	80 - 160	350 - 450
NO_2	30 - 45	35 - 45	50 - 70	80 - 120
Particle [c]	-	-	-	130.000
CO[a]	-	-	1700 - 2100	2000 - 5500

[a] 8-hours running average on annual basis, [b] 95 percentile,
[c]Number concentrations, one years measurements at H.C. Andersen's Boulevard and H.C. Ørsted Institute, Copenhagen.

In the 1990ties, the nitrogen oxide and carbon monoxide pollution from traffic was higher compared with the present levels (Figure 2.4). This improvement is mainly due to the introduction of catalytic converters on gasoline driven vehicles. Annual mean NO_x levels were typically about 25% higher, whereas street level CO was 50 to 100% higher in the most trafficked streets. The range in air pollution exposure was similarly larger in the 1990ties compared with the current situation.

In recent years, it has been seen that the air quality in Danish streets in some cases still exceeds the limit values. Measurements from the urban air quality monitoring programme have shown that NO_2 levels at three streets in 2005 exceeded the EU limit values for 2010 of 40µg/m^3 (Kemp et al., 2006). The NO_2 levels at H.C. Andersen's Boulevard furthermore exceeded the EU limit + margin of tolerance[1]. For PM_{10} limit levels in Denmark one station in 2005 (Kemp et al., 2006) and two stations in 2004 (Kemp et al., 2005) exceeded limit value + margin of tolerance.

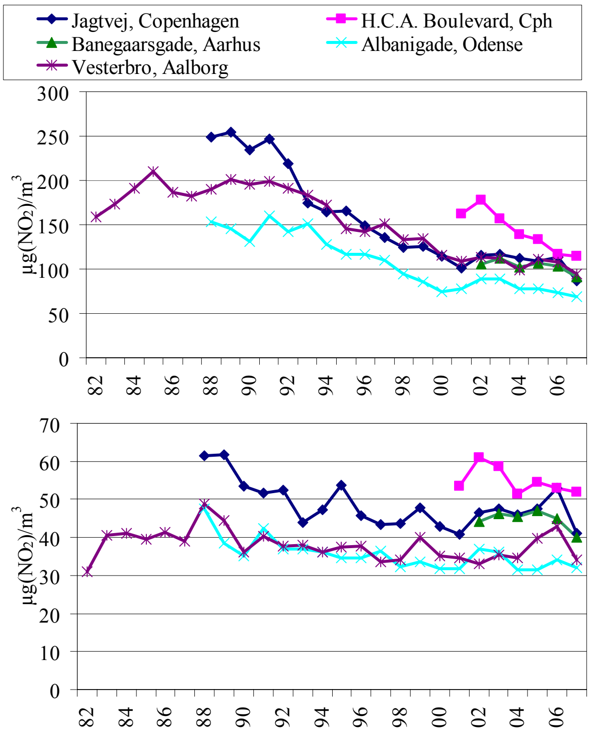


Figure 2.4 The measured trend in annual mean concentrations of NO_x (upper plot) and NO_2 (lower plot) (both shown in µg(NO_2)/m^3) at the street stations in the largest Danish cities Copenhagen, Aarhus, Odense and Aalborg. Upper plot shows NO_x and lower plot the NO_2. The plots include measurements from the time period 1982 to 2005. Source: (Kemp et al., 2006; Hertel et al., 2007a) (Paper XII).

2.3 Modelling urban background

The air pollution level in the urban background is the result of both regional pollution and contributions from the city itself. In many of the studies carried out in Copenhagen and the three other cities in the Danish urban air quality monitoring programme, urban background concentrations have been obtained from measurements performed at a station in the city. The measured levels have then been used to determine the contribution from traffic in the single street (I will return to this later), and as input to the calculations of the street pollution levels. Naturally, this method cannot be applied for assessment of the impact of emission regulations, or when calculations are prepared for estimating pollution concentrations for time periods before the monitoring was established. For calculating urban background concentrations in such situations, a simplified Gaussian dispersion model - the Urban Background Model (UBM) – is developed (Hertel and Berkowicz, 1990; Berkowicz, 2000a).

[1] The EU Daughter directive defines limit values to be complied with no later than 2010. It the time period up to 2010 a margin of tolerance between the previous limit values and the limit values of 2010 is defined.

The first version of the UBM is developed in the 1990ties in connection with a system for forecasting the development in NO_2 levels during periods with warnings of enhanced air pollution levels. This first model version is developed on basis of analyses of Danish and Dutch measurements from urban areas (Hertel and Berkowicz, 1990). The strength of the UBM is that it demands limited input data, it is relatively fast, and it is easy to apply. The UBM is now a routine tool in the NERI air pollution forecasting system THOR (Brandt et al., 2001a), as well as in the human exposure modelling system AirGIS (Jensen et al., 2001b) (Paper IV). The GIS (Geographical Information Systems) based AirGIS system is described in more detail in a later section.

2.4 Modelling street pollution

The principles behind the OSPM are developed in the late 1980ties (Hertel and Berkowicz, 1989b). However, the OSPM has been improved and extended over the years. Various improvements of the methodology and specific parameterisations applied to the model are now described in several subsequent papers (Berkowicz et al., 1997a; Berkowicz, 2000b).

In a street canyon, the contribution from the traffic emissions that take place inside the street (street contribution - c_s) is added to the pollution present in the air that enters from roof level (urban background - c_b).

$$c = c_b + c_s \tag{2.1}$$

The pollution emitted from traffic in the street is advected by the wind vortex towards the leeward side of the street (See Figure 2.2). For the windward side of the street, the impact of emissions in the street is only from the air that has recirculated inside the canyon. In order to consider both contributions in OSPM, the street contribution is calculated by a combination of a plume model for the direct contribution, and a box model for the recirculating part of the pollutants in the street. It is assumed that the traffic emissions are evenly distributed across the street, and that the emission field may be treated as a number of infinitesimal line sources aligned perpendicular to the wind direction at street level. The cross wind diffusion is disregarded. For the contribution from recirculation, the canyon vortex is assumed to have the shape of trapeze. For more details about the OSPM, see (Berkowicz et al., 1997b) (Paper I).

2.4.1 Low wind speed conditions

The wind speed and the wind direction are the two most important parameters for the pollutant transport and dispersion inside the street. This has e.g. been demonstrated in a comparison of pollutant levels in Milan and Copenhagen (Vignati et al., 1996). At very low wind speeds there is not sufficient energy in the wind to drive the vortex. Below a wind speed of 2 m/s, the recirculation is therefore disregarded in the OSPM. At moderately low wind speeds, the wind direction tends to fluctuate; a tendency that increases with decreasing wind speed. This is accounted for after an analysis of measurements from St. Olav's gate in Oslo (Hertel and Berkowicz, 1989c). In Oslo, low wind speed conditions are very frequent compared with Copenhagen. The average wind speed is thus about 2 m/s in Oslo, which may be compared with about 5 m/s in Copenhagen of. The street canyon ventilation velocity is determined by the turbulence at the top of the canyon. The ventilation velocity determines how quickly the pollution is removed from the street canyon. At low wind speeds this canyon ventilation velocity is governed by the traffic induced turbulence, and also this is accounted for in the model parameterisation (Hertel and Berkowicz, 1989b; Hertel and Berkowicz, 1989c; Berkowicz et al., 1997a). In a resent paper various formulations of traffic induced turbulence is tested in the OSPM, by comparing the results to measurements (Solazzo et al., 2007).

2.4.2 Nitrogen oxide chemistry

$$\begin{array}{l}
\mathbf{NO + O_3 \rightarrow NO_2 + O_2} \\
\mathbf{NO_2 + hv \rightarrow NO + O} \\
\mathbf{O + O_2 \rightarrow O_3} \\
\hline
\mathbf{NO + O_3 \rightarrow NO_2 + O_2} \\
\mathbf{NO_2 + hv \rightarrow NO + O_3}
\end{array} \tag{2.2}$$

The distribution between the harmless NO and the airway irritant NO_2 is therefore to a high extent determined by the fast reaction between NO and ozone (O_3) and the similarly fast photo dissociation of NO_2 back to NO and O radical. The O radical is quickly reforming O_3 in reaction with

2.4.3

The OSPM is originally developed for describing nitrogen oxides in urban streets. Nitrogen oxides (NO_x) are mainly emitted as nitrogen monoxide (NO) (typically 90% to 95%) and to less extend as nitrogen dioxide (NO_2) (typically 5% to 10%). The residence time in the street is short (few minutes) and only very fast reactions have time to take place inside the street.

O_2. It is a good approximation to assume O_3 to be the product of the photo dissociation of NO_2 (see Equation 2.2). An analysis of measurements from urban streets show that NO_2 concentrations may thus be predicted by accounting for only these two reactions (Palmgren et al., 1996).

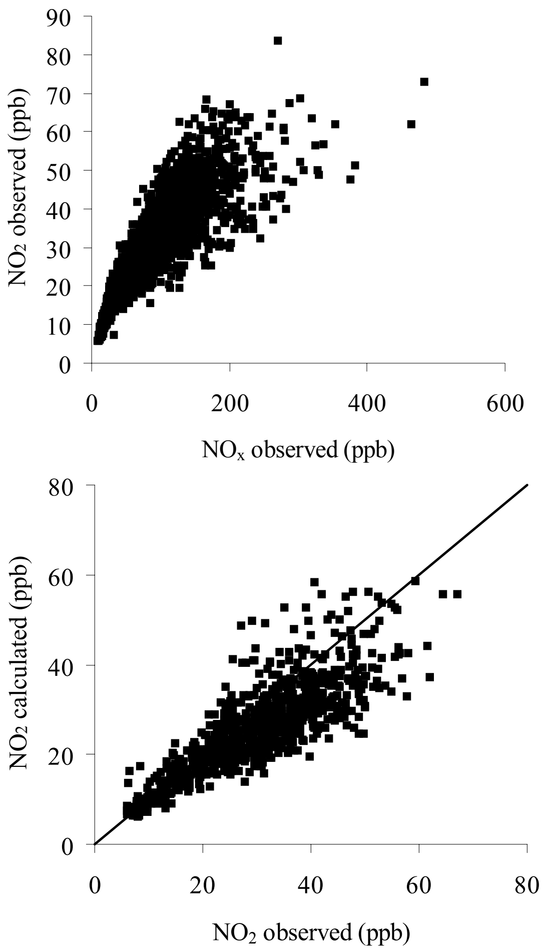


Figure 2.5 The chemistry of NO_x in urban streets. Upper plot: the relationship between NO_2 and NO_x. For NO_x concentrations below about 20 ppb, all NO_x is in the form of NO_2, since the air contains sufficient O_3 for converting all NO to NO_2. For higher NO_x concentrations, only the direct emission of NO_2 contributes to further increase in the NO_2 concentrations. Lower plot: Comparison between observed and calculated hourly mean concentrations of NO_2. All data are from Jagtvej in Copenhagen in 2003, and calculations performed with the OSPM. Only working days during daytime (800-1600) are included. Correlation coefficient (R^2) = 0.7. Source: (Hertel et al., 2007b).

A chemical sub-model for describing these two reactions is developed and implemented in the OSPM in the later 1980ties (Hertel and Berkowicz, 1989a; Palmgren et al., 1996) and later also in the UBM as well as in the OML plume model (Olesen, 1995). The concentration of NO_2 in the urban areas (street as well as urban background) is thus strongly governed by the long range transported O_3. Model calculations are in good agreement with observations, even with this simplification of the NO_x chemistry (Figure 2.5). In a recent study it has been applied for studying primary emissions of NO_2 in London (Carslaw and Beevers, 2004).

2.4.4 Other types of streets

The first version of OSPM is developed on basis of detailed analyses of air pollution measurements carried out during campaign studies at Vesterbrogade in Copenhagen (Hertel and Berkowicz, 1989b). Vesterbrogade is a typical urban street canyon, and the first parameterisations are adapted to the dispersion conditions in street canyons. Later, the parameterisations are improved after analyses of pollution measurements from other street canyons in Denmark and abroad (Hertel and Berkowicz, 1989c; Berkowicz et al., 1996). However, the aim is to develop a more general model tool for application in any type of urban street. The OSPM is therefore during the 1990ties further developed to account for other types of urban streets (Berkowicz et al., 1997a). These street types include streets with buildings on only one side of the street, streets with openings in the building facade along the street, streets with buildings of different heights along the street, and even fully open streets with no buildings at all. This is handled in OSPM by specifying a general building height along the street, and then specifying wind sectors in which the building height deviate from the general building height. In these sectors the wind vortex flow is affected.

2.4.5 Input data for the model

Various types of input data are needed for the OSPM calculations of air pollution in the streets (see the simple sketch in Figure 2.6). These input data include meteorological parameters, urban background concentrations, emission data, traffic data and street configuration data.

The street configuration concerns the specific characteristics of the street and its surroundings, which are parameters that to a large extent are governing the flow conditions in the street. The street configuration include: street width, the general building height around the street, the street orientation, sectors with buildings different from the general building height. The input files may be edited manually, or they can be prepared within the user interface of the WINOSPM.[2]

[2] The OSPM may be downloaded in a trial version at the webpage http://ospm.dmu.dk. This web-page also contains lists of relevant publications concerning the model etc.

There are various ways to categorise urban streets. However, in the following the categorisation used in the Dutch CAR model (den Boeft et al., 1996) is adopted due to its simplicity. Furthermore, this categorisation is also used in our Danish Children Cancer Study, which is described in a later section of this thesis. The Dutch categorisation split the streets into five classes of streets A to E, where the first three classes are for streets with no or only low buildings and the last two are for streets with tall buildings on one or two sides (See Table 2.3).

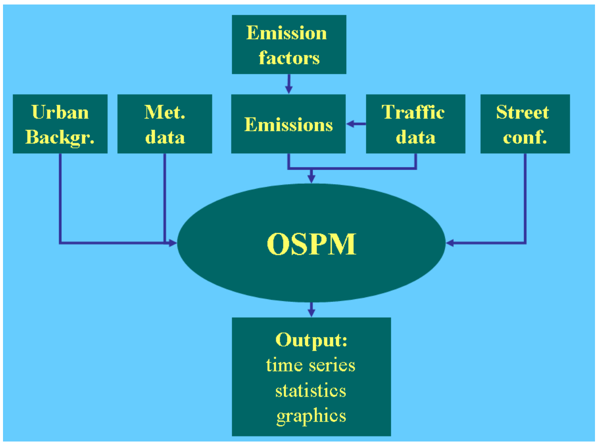
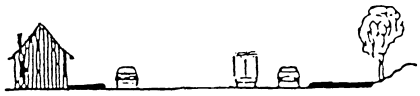
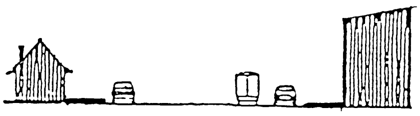
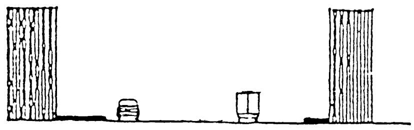
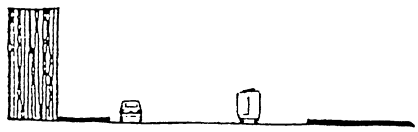


Figure 2.6 Sketch to illustrate the data flow in the OSPM. The input data include meteorological parameters (wind speed, wind direction, temperature and global radiation), traffic data (temporal variation in number of vehicles and driving speed distributed on categories), emission factors (temporal variation distributed on vehicle categories) and street configuration (street width, general building height, street orientation, sectors where the building height deviate from the general building height).

2.5 Validation studies of OSPM

The OSPM is applied in a large number of Danish as well as foreign air pollution studies. A summary of the OSPM validation studies found in literature is shown in Table 2.4. The street types are here categorised according to the classification used for the Dutch Car model (Table 2.3), and the model performance crudely grouped into: good, fair and poor. In addition various information about: applied input data, studied pollutants, place where the study is carried out, and references to where the studies are published, are also given in Table 2.4.

Table 2.3 Street categories as they are used in the Dutch CAR model (Eerens et al., 1993; den Boeft et al., 1996). This categorisation is applied in Table 2.4.

Category	Description
A	A house in an open street. The house can be situated close to or far from the street. There may be other houses nearby. 
B	Low scattered houses, this category include: • A row of low houses on one side of the street. Almost no houses on the other side. • Villas on both sides with space (gardens) in between. • Low houses with open front areas (parking lots, gardens etc.) 
C	Low houses on one side of the street and tall houses on the other side of the street. 
D	Tall houses on both sides of the street 
E	Tall houses on one side of the street. Almost no houses on the other side. 

The significant number of validation studies has the advantage that the OSPM is validated by comparison to measurements from a wide number of monitoring sites in urban streets. A Danish example of such a comparison of OSPM calculations and measurements performed in Jagtvej, Copenhagen is shown in Figure 2.7. Whereas the plot in Figure 2.5 shows a comparison for the chemically active NO_2, the plot in Figure 2.7 shows NO_x values (sum of NO and NO_2).

Table 2.4 Monitoring sites in urban streets in Denmark and abroad for which the OSPM calculations have been compared to measurements. Categories according to those used for the Dutch CAR model (see Table 2.3). BNZ is short for benzene. The "Ttraffic" column indicate the source of traffic data.

Cat.	Pollutant	Res.	Traffic	Emis factors	Calc. year	Meteorology	Perf.	Street name	Reference
D	NO_x	Hour	Manual	Fit to conc.	1985 - 86	Airport	Good	Vesterbrogade, Copenhagen, DK	(Hertel and Berkowicz, 1989b)
D	NO_x	Hour	Automatic	Local factors	1984 - 86	Mast on roof	Good	St. Olavs Gate, Oslo, Norway	(Hertel and Berkowicz, 1989c)
D	NO_x/CO	Hour	Manual	Fit to conc.	1989	Airport	Both good	Jagtvej & Bredgade, Copenhagen, DK	(Hertel and Berkowicz, 1991)
D	NO_x/NO_2/CO	Hour	Manual	Fit to conc.	1993 - 94	Mast on roof	All good	Jagtvej & Bredgade, Copenhagen, DK	(Berkowicz et al., 1996)
E	NO_x/NO_2	Hour	Manual	Fit to conc.	1993 - 94	Mast on roof	Fair/fair	H.C. Andersens Bld., Copenhagen, DK	(Berkowicz et al., 1996)
A - E	NO_x/BNZ	Week[1]	Estimate[2]	OSPM	1994 - 95	Mast on roof	Good/fair	204 streets in Copenhagen area, DK	(Raaschou-Nielsen et al., 2000)
A - E	NO_x/BNZ	Week[1]	Estimate[2]	WINOSPM	1994 - 95	Mast on roof	Good/fair	204 streets in Copenhagen area, DK	(Berkowicz et al., 2008)
D	CO	Hour	Automatic	WINOSPM	1994	Mast on roof	Fair	Göttinger straße, Hannover, Germany	(Aquilina and Micallef, 2004)
D	CO	Hour	Automatic	WINOSPM	1994 - 95	Mast on roof	Good	Jagtvej, Copenhagen, Denmark	(Aquilina and Micallef, 2004)
D	CO	Hour	Unknown	WINOSPM	1995	Mast on roof	Fair	Schildhorn Straße, Berlin, Germany	(Aquilina and Micallef, 2004)
D[3]	NO_x/CO	Hour	Evenly[4]	OSPM[5]	1995	Mast on roof	Both fair	Qianmen Dajie street, Beijing, China	(Fu et al., 2000)
D	CO/NO_x/NO_2	Hour	Automatic	LIISA95	1997	Mast on roof	All good	Runeberg St., Helsinki, Finland	(Kukkonen et al., 2000; Kukkonen et al., 2001; Kukkonen et al., 2003)
D	NO_x	Hour	Modelled	MIMOSA	1998	Mast on roof	Good	Plantin en Moretuslei, Antwerp, Belgium	(Mensink et al., 2006)
D	CO/BNZ	Hour	Automatic	COPERT II/own	1998	Masts & Airport	Both fair	Bld Voltaire, Paris, France	(Vardoulakis et al., 2002)
D	CO/BNZ	Hour	Automatic	COPERT II/own	1999	Masts & Airport	Both fair	Rue de Rennes, Paris, France	(Vardoulakis et al., 2002)
D	CO/NO_2/NO_x/BNZ	Week	Estimate[9]	COPERT III	1999 - 01	Unknown	All good	Rue Crébillon, Nantes, France	(Gokhale et al., 2005)
D	CO, NO_2, NO_x	Hour	Estimate[9]	COPERT III	1999 - 01	Unknown	All good	Rue de Strasbourg, Nantes, France	(Gokhale et al., 2005)
D	CO, NO_2, NO_x	Hour	Estimate[9]	COPERT III	1999 - 01	Unknown	All good	Boulevard Victor Hugo, Nantes, France	(Gokhale et al., 2005)
C	CO, BNZ	Week	Unknown	IMPACT[6]	2001	Masts & Airport	Fair/ poor	Avenue Leclerc, Paris, France	(Vardoulakis et al., 2005)
D	NO_x, Part.no	Hour	Manual	WINOSPM[7]	2001	Mast on roof	All good	Jagtvej, Copenhagen, DK	(Ketzel, 2004)
D	NO_x, BNZ	Hour	Manual	Fit to conc.	2000 - 01	Mast on roof	Both good	Pestelya, St. Petersburg, Russia	(Ziv et al., 2002)
E	NO_x, $PM_{2.5}$	Day	Manual	WINOSPM	2003	Mast on roof	Fair/ poor	H.C. Andersens Bld., Copenhagen, DK	(Hertel et al., 2008) (Paper VI)
D	NO_x, PM_{10}	Day	Manual	WINOSPM	2003	Mast on roof	Good/poor	Jagtvej, Copenhagen, DK	(Hertel et al., 2008) (Paper VI)
D	CO, PM_{10}	Hour	Auto./man.	Local factors	2003	Mast on roof	Good/good[8]	Stratford Road, Birmingham, UK	(Vardoulakis et al., 2007)
D	NO_x/NO_2/CO/PM_{10}	Hour	Auto./man.	Local factors	2003	Airport	All fair	Marylebone Road, London, UK	(Vardoulakis et al., 2007)
D	NO_x/NO_2/PM_{10}	Hour	Manual	COPERT III	2004	Mast	All fair	3 streets hist. centre, Thessaloniki, Gr	(Assael et al., 2008)
D	CO/NO_x/$PM_{2.5}$	Hour	Automatic	COPERT III/fit	2004	Mast	Good	Rue du Revarel, Rouen, Fr	(Ghenu et al., 2008)

[1]NO_2 measured as 14 days and benzene as weekly samples – in both cases by passive samplers.[2]All street configuration data obtained from questionnaires fill in by local municipalities.[3]The monitoring station placed in crossing between two streets. Calculations performed for both streets and the results selected depending on wind direction. [4]The diurnal traffic obtained from counts provided by local authorities. The traffic assumed evenly distributed over the day (crude assumption).[5]The emission factors for European cars applied since no Chinese factors available.[6]Benzene emissions obtained from relationship to CO in ambient air.[7]Emission factors for particle numbers derived from relationship between particle number and NO_x concentrations in ambient air.[8]PM_{10} is dominated by the contribution from urban background and this explains a significant part of the good model performance. [9]Traffic data obtained from local authorities.

The NO_2 concentrations are to a large extent governed by the regional O_3 concentrations (see section 2.4.2). Applying measured O_3 concentrations for the calculations, the NO_2 may have a high correlation as a result of the dependency of O_3. The comparison for NOx is thus a more direct test of the models ability to describe the dispersion conditions in the urban streets.

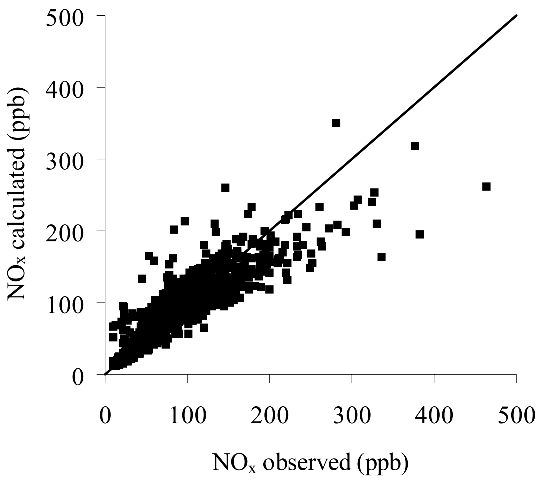


Figure 2.7 Comparison between observed and calculated hourly mean concentrations of NO_x for the monitoring station at Jagtvej, Copenhagen in 2003. Calculations performed with the OSPM. Only data for working days during daytime (800-1600) are included. Correlation coefficient (R^2) = 0.8. Source: (Hertel et al., 2007b).

2.5.1 Geography and street type

Most of the OSPM validation studies are naturally carried out for streets in the city of Copenhagen, and the studies found in literature are all except one (from Beijing, China) from European cities (in Norway, Finland, Germany, Belgium, England, Russia, Greece and France).

With very few exceptions, the performed validation studies all concern comparisons to measurements from monitoring stations in street canyons. This is probably because street canyons are where the highest air pollution levels are expected to appear, and the various research-groups all aim at using dispersion models for the air pollution hot spots in the city. However, this is a weakness in relation to having the OSPM performance tested for other types of streets. Especially since the performance of OSPM seems less good in the few studies performed for other types of streets.

2.5.2 Traffic, meteorology and emission data

In about one third of the validation studies, traffic data is obtained from automatic counting. For the remaining two thirds of the studies, traffic data is obtained from manual counts, or from a combination of the two. In several of the studies, it is emphasised by the authors that the model performance depends strongly on the quality of the traffic and the emission data.

The meteorological data is generally obtained from a local mast placed in the urban area, and only in a couple of cases from nearby airports. In a few studies, data from a local mast and a nearby airport data are combined in order to obtain a full time series.

In the first OSPM studies performed at NERI in the later 1980ties and early 1990ties, the emission data is obtained from fitting the model results to measured concentrations. In the published validation studies, the emission module from WINOSPM is often applied, but there are also many studies where local emission data have been applied.

2.5.3 Pollutants

The earliest OSPM validation studies are performed for the gaseous pollutants CO, NO_x and NO_2. In most of the studies, the model performs well for the traditional gaseous traffic pollutants. For benzene, the performance is poor in several of the validation studies. However, the uncertainties in ambient benzene measurements are rather high when passive samplers are applied (as in many of these studies), and the benzene emission factors are even more uncertain. It should here be noted that the OSPM performs well for the Russian study where the benzene (and NO_x) emission factors are generated from an inverse modelling approach, but also for Rue Crébillon in Nantes, France for which emissions are obtained from COPERT III.

Only one validation study from Jagtvej (Ketzel, 2004) is focusing on modelling particle number concentrations. Like in the case of some of the benzene studies, the emission factor for the particle number is generated from analysis of the relationship to NO_x in ambient air concentrations. Particle mass is modelled in a couple of studies. The relatively poor model performance obtained for particle mass is due to the complex emission processes. Only a fraction of the particle mass is emitted with the vehicle exhaust, since a large part is related to resuspension of dust and wear of road material, tires, breaks etc.

2.6 Assessment of traffic pollution

In this section a couple of OSPM assessment studies and a traffic pollution monitoring programme are presented. These studies are evaluated on the background of the actual development in traffic pollutant concentrations in urban streets obtained from the Urban Air Pollution Monitoring Programme.

2.6.1 Testing of derived emission factors

In 1991 OSPM is applied in a research project "Air pollution from individual and collective traffic" carried out for the Danish EPA and the Road Data Laboratory (Hertel and Berkowicz, 1991). The aim is to look at the current and future situation in urban streets using emission factors derived in another part of the project. Comparisons to measurements of ambient air NO_x concentrations from Bredgade in Copenhagen show a good agreement for the current situation.

The modelled air pollution concentrations for the current situation are compared with similar calculations representing the future situation when all gasoline cars are assumed to have catalytic converters. It is concluded that NO_x and CO concentrations are expected to decrease by a factor 2.4. However, it is also concluded to be highly uncertain to what extend NO_2 levels will decrease in the future. This uncertainty is due to the dependency of NO_2 concentrations on long-range transported O_3. These conclusions are later seen to match the reality pretty well; since Denmark now has difficulties to comply with EU limit values for NO_2, despite the decrease in NO_x concentrations in streets of Copenhagen and other larger cities in the country.

2.6.2 The traffic pollution monitoring programme

In 1995 a monitoring programme for traffic air pollution (the TOV programme) is carried out for the Danish ministry of Traffic as cooperation between NERI, the Danish Meteorological Institute, The Road Data Laboratory, and the Municipality of Copenhagen. In the TOV programme, additional air quality measurements are established to supplement those carried out within the Urban Air Quality Monitoring Programme. Automatic traffic counts are established at Jagtvej. An important part of NERI's responsibility in the TOV programme is to apply the OSPM in the analyses of the measurements from urban streets (Hertel et al., 2000). The basic idea is here to study trends in pollutant emissions from traffic in Danish urban streets.

The concentration of the non-reactive or slowly reacting pollutants may be expressed as (Equation 2.3):

$$\begin{aligned} c &= F(met) \times Q + c_b \\ Q_t &= \frac{c_t \quad c_b}{F_t(met)} \end{aligned} \qquad (2.3)$$

Where *Q* is the emission, *F(met)* is a function that describes the dispersion in the street (depending on meteorological conditions), c_b is the contribution from other sources than traffic in the street (the urban background), and the index *t* refers to the actual time on the day. A method for removing outliers (the result of errors in modelling or measuring the pollutants) is developed for the analysis. Given that detailed traffic counts distributed on categories are available, it is furthermore possible to derive emission factors for the different vehicle categories (Palmgren et al., 1999).

Table 2.5 The measured trend (in %) in annual mean concentrations and estimated trend in annual mean emissions for NO_x, benzene and CO at Jagtvej during the time period 1993 to 1999. All figures are relative to the year 1994, which is set to 100%. Source: (Hertel et al., 2000).

	NO_x		Benzene		CO	
	Conc	Emis	Conc	Emis	Conc	Emis
1993	118	101				
1994	100	100	100	100	100	100
1995	93	92	80	90	69	72
1996	99	83	90	77	48	38
1997	71	73	68	69	31	30
1998	60	64	55	60	21	21
1999	67	62	76	69	24	19

The pollutant concentrations vary with the meteorological conditions, and this is the case even when annual mean concentrations are considered (Table 3.5). Over the time period 1993 to 1999, the ambient air pollutant concentrations at Jagtvej decrease by more than 50%, whereas the decrease in emissions is estimated to 38%. For benzene the decrease in concentrations and emissions are 24% and 31%, respectively. The largest decrease is observed for CO where concentrations decrease 89% and the emissions 86%. The increase in the number of gasoline vehicles with catalytic converters is the main explanation for the observed decreases in ambient air pollution concentrations and emissions.

2.6.3 Trends in nitrogen dioxide levels

Measurements show that at Jagtvej in Copenhagen over the time period 1988 to 2005, the annual mean ambient air concentrations of NOx have decreased by a factor of 2, whereas NO_2 is reduced only by about 25% (Table 2.6). This may be compared with the previously mentioned estimate from 1991 in the project on "Air pollution from individual and collective traffic", where NO_x concentrations in Bredgade are expected to decrease by a factor of 2.4 whereas the NO_2 concentrations are believed to decrease only moderately.

In fact the NO_2 levels in Danish urban streets in 2007 still exceed the EU limit value for 2010 of 40 µg $(NO_2)/m^3$ (Kemp et al., 2008). Different abatement strategies to reduce the ambient NO_2 levels in urban streets are now being discussed. In this context a new scenario study using the OSPM is carried out.

This study investigate the impact on NO_2 levels in urban streets of using the more recent technologies in SCR filters on heavy duty vehicles (Palmgren et al., 2007).

Table 2.6 The development in NOx and NO2 concentrations over the period 1988 to 2005. Values are obtained from the measurements within the Urban Air Quality Monitoring Programme. Source: www.dmu.dk.

	1988	2005
NO_x	180	87
NO_2	62	47

2.6.4 Impact of particle filters on NO_2 levels

In 1999 a scenario study is carried out in order to evaluate the impact on NO_2 levels in urban streets of introducing CRT (Continuous Regenerating Trap filters) particle filters on heavy duty vehicles (Hertel and Berkowicz, 2000). In Swedish test stand studies, a direct NO_2 emission of more than 30% of the total NO_x emission is measured on a van and a bus equipped with CRT filters. This gives rise to concern in relation to complying with the EU limit values for NO_2 concentrations in urban streets. Implementation of CRT filters with such a high direct emission of NO_2 is thus likely to keep NO_2 levels in Danish urban streets at a high level.

The case study is setup for Odense, since an experiment with CRT filters on busses and heavy duty vehicles is planned. UBM and OSPM are applied for studying the current and future situation at background and street level, respectively. The results show that the annual mean limit value of 40 μg/m^3 for NO_2 is expected to be exceeded. The limit value will be exceeded for a direct NO_2 emission from heavy duty vehicles in the range of 20 to 30% of the total NO_x emission.

2.6.5 Impact of environmental zones

A way to quickly reduce the health impacts of traffic pollution is to establish environmental zones around the larger cities. Inside these zones restrictions are put on the car fleet i.e. heavy duty vehicles are only allowed to enter the zone if they are equipped with particle filters. Since October 1st 2006 particle filters have been a demand on newly registered heavy duty vehicles that have to comply with the Euro-IV norm (European Commission Directive 98/69/EC). However, it takes time before the car fleet has been renewed, and the environmental zone is therefore a faster way to reduce emissions in the close vicinity of where people reside.

The impact on the health effects of introducing particle filters on heavy duty vehicles in Denmark is estimated as a part of the Danish particle study (Palmgren et al., 2001). I will return to this Danish study in the next chapter, but the results of the particle study indicated that implementation of particle filters on heavy duty vehicles might lead to a reduction in the range of 22 to 1250 incidences of premature death together with a series of other adverse health effects (Palmgren et al., 2001; Raaschou-Nielsen et al., 2002). The uncertainty of the impact assessment is very high, but still the results indicate that particle pollution in Denmark is associated with significant health effects.

The impact on air quality of establishing environmental zones around the larger Danish cities is been investigated in couple of studies (Jensen et al., 2001a; Palmgren et al., 2005). These studies show that the local contribution to PM_{10} is moderate. The annual mean urban background concentration in 2001 in Copenhagen is estimated to about 23μg/m^3 with a contribution from local traffic of about 1μm/m^3. The use of particle filters on heavy duty vehicles is estimated to reduce the local contribution to PM_{10} about 40%; from about 1μg/m^3 to about 0.6μg/m^3. This is then estimated to reduce the number of premature death by about 2.4 incidences per year. The number of incidences of chronicle and acute bronchitis is estimated to be reduced by about 10, asthma attacks by about 115, and the number of days with reduced physical activity by about 1300, respectively. For Aalborg and Vejle, the expected reduction in premature death is estimated 0.5 and 0.2 incidences, respectively.

2.7 Conclusions

The aim of this chapter was to address how IMA has contributed to new knowledge and improved methodologies in the assessment of traffic pollution in urban streets. The introduction addresses environmental and more technical questions. In the following I will go through these questions one by one.

A.1. How is the distribution between air pollution contributions from local traffic in the single street, from sources in the urban area in general, and from sources in more remote areas (including long-range transport)?

The range in air pollution concentrations vary strongly from one pollutant to another. For Danish levels of particle mass in ambient air, the long-range transport component dominates the urban background levels. For particle mass there is therefore almost no gradient between the levels in the urban background and the rural sites. Our estimates point at a local contribution to the annual mean urban background on the order of 1μg/m^3, which may be compared with a long-range transport component in the order of 22μg/m^3.

For NO_x and especially for particle number concentrations in the urban streets there is a strong signal from local traffic, and the concentrations quickly decrease with distance to the sources. The annual mean NO_x concentrations are a factor of two to three higher in the urban background (25 – 45µg(NO_2)/m^3) compared with the rural levels. For NO_2 this ratio between urban and rural background is slightly smaller and for PM the gradient is only significant when the extreme levels are considered i.e. the 98 percentile. The relationship between rural and street pollution level is up to an order of magnitude for NO_x, and a factor of four to five for NO_2.

A.2. Do we comply with current and future air quality guidelines for urban streets?

The measurements in Danish urban streets show that the air pollution levels in some cases exceed the EU limit value for 2010 for NO_2 and particle mass. OSPM calculations performed on 138 streets in Copenhagen have shown that this is true for a wide range of urban streets, and that further actions are probably needed in order to reduce NO_2 levels in Danish urban streets sufficiently to comply with EU limit values in 2010.

These studies have demonstrated the strength in applying well-tested operational models for carrying out air pollution scenario studies.

A.3. What are the impacts of certain emission reduction strategies concerning traffic air pollution?

The introduction of catalytic converters on gasoline driven vehicles reduces the NO_x emissions by about 90%. OSPM calculations performed in 1991 indicated that NO_x concentrations were expected to decrease by about a factor of two after full implementation of catalytic converters on all gasoline driven vehicles in the Danish car park, whereas NO_2 concentrations were expected remain almost unchanged. Measurements performed over the time period 1988 to 2005 have shown that NO_x concentrations have decreased by a factor of two, whereas NO_2 only is reduced by about 25% over the same time period.

Laboratory studies in the late 1990ties showed that the direct NO_2 emission from heavy duty vehicles would increase to 20 or even 30% of the total NO_x emission by introduction of CRT filters. Without particle filters, it is in the range of 5 to 10%. OSPM calculations performed in 1991 show that NO_2 levels may exceed guideline values after the introduction of CRT filters on the heavy duty vehicles.

Introduction of environmental zones around the larger cities is a way to quickly reduce the exposure of the population to traffic pollution. A survey on international literature performed in 2001 shows that mainly particle pollution is believed to be associated with adverse health effects. Analyses of Danish PM_{10} levels show that the local contribution to urban background is only about 1µg/m^3, compared with a total PM_{10} concentration of about 23µg/m^3 (annual mean). A 40% reduction of the local contribution is considered possible as a result of introduction of particle filters. In Copenhagen this would results in a reduction of the incidences of premature death of about 2.4 per year. The number of incidences of chronicle and acute bronchitis is estimated to be reduced by about 10, asthma attacks by about 115, and the number of days with reduced physical activity by about 1300 incidences.

B.1. How good is the performance of the simple parameterised street pollution model?

The OSPM is widely used, and generally accepted as state-of-the-art in parameterised operational models for air pollution in urban streets. A literature survey has shown that the model has been validated in studies performed in eight different countries – mainly in Europe. Generally a good model performance has been obtained for the traditional gaseous traffic pollutants, especially when high quality traffic data and emission factors are available. Less good performance is obtained for calculated ambient air particle mass where the emission is considerably less well defined. High quality of input data has been shown to be crucial for the performance of OSPM; this especially applies for traffic and emission data.

B.2. To what extend do we have access to the necessary input data at the demanded level of quality?

In many of the international studies there is no access to local emission factors, and these have therefore been estimated crudely. Similarly there is often a lack of good quality traffic data based on counts in the specific street and distributed on vehicle categories. In the Danish traffic monitoring project, emission factors were successfully derived from inverse modelling. The study showed, however, that the quality of traffic data was a weak point in the analysis. As a result, the inverse modelling was given up after a couple of years.
Recently diurnal traffic data has been collected for the entire Danish road network (I will return to this in the next chapter). Data is therefore now available for assessment of traffic pollution in Denmark.

B.3. To what extend do we have access to air pollution measurements for testing the model for various pollutants and various types of streets?

Most OSPM validation studies in literature – national as well as international - have been performed for street canyons. Generally there is a need for studies for other street types for assessment of traffic pollution in urban streets. This is especially important when the model is intended to be used for mapping of air pollutant levels of an entire urban area. This is the case in a number of applications for assessment of human air pollution exposure that I will return to in the next chapter. A strengthening of the international cooperation may be a way to overcome this obstacle of getting access to street pollution measurements from other street types than street canyons. Many European cities have long time-series from a number of street stations.

2.8 Fingerprints on science and environmental management

OSPM represent state-of-the-art in operational models for air quality in urban streets. Seen in international perspective, the OSPM is to my knowledge the most widely used model of its kind. During the 1990ties, at least two research groups (one in France and one in England (Buckland, 1998)) have developed their own versions of the model on basis of the descriptions provided in technical reports published from NERI.

The VLUFT system (Tønnesen, 2000) is developed by the Norwegian Institute for Air Research (NILU), and used for mapping air pollution in streets[3]. The VLUFT is based on a combination of the American Highway2 for handling air pollution from highways and the OSPM is also for this system handling air pollution in urban streets.

The SIMAIR system is developed by the Swedish Meteorological and Hydrological Institute (SMHI) to be used by the Swedish municipalities for mapping air pollution from in urban streets[4]. This system is again using OSPM for computing air pollution in urban streets.

The British Nomogram method called AEOLIUS for calculating air pollution in urban streets was derived from OSPM calculations (Buckland and Middleton, 1999).

SMHI has similarly developed a nomogram method for use in Sweden and again based on OSPM calculations (Foltescu and Gidhagen, 2001).

A literature survey has shown that the OSPM has been validated in comparison to measurements in 8 different European countries as well as in China (see Table 2.4). In the Netherlands, the Dutch CAR model (den Boeft et al., 1996) has been applied for a number of years. The CAR model is an empirically derived calculation method. Such empirical methods have their limitations in relation to perform scenario studies, and researchers at RIVM in the Netherlands are currently considering substituting CAR with the OSPM in their future air pollution assessments (Joost Wesseling, RIVM, 2007, personal communication). The model is currently being applied in Turkey (Hicran Altug, Anadolu University, Dep. of Env. Eng., 2008, personal communication), and recently an application of the model for the centre of Manhattan, New York in the US has been published (Zhou and Levy, 2008).

The OSPM is a routine tool within the Danish Urban Air Quality Monitoring Programme (LMP). The model is used for scenario studies as well as for mapping air pollution levels in streets where measurements are not carried out. OSPM is a part of the NERI air pollution forecasting system THOR (Brandt et al., 2001a), which to my knowledge is the only current system for producing 4-days air pollution forecasts from hemispheric scale and down to the scale of single streets.

OSPM has been applied in various European projects for assessment of air pollution from traffic in European cities, including the Auto Oil II project for assessment traffic generated air pollution in urban areas (Skouloudis and Suppan, 2000). The analytical solution for the NO, NO_2 and O_3 chemistry in urban streets developed for the OSPM has been implemented also in the OML plume model (see Chapter 6), and the method inspired to the development of the Eulerian Backward Interative (EBI) solver developed for the ACDEP model (see Chapter 4). OSPM and UBM are core parts of the AirGIS system for human air pollution assessment (see Chapter 3). The work with the development of the OSPM had a strong impact on the design during the revision of the Urban Air Quality Monitoring. The combination of street stations and urban background stations makes it possible to separate the contribution from traffic in the single street from the general contribution from the urban area.

[3]www.nilu.no/index.cfm?ac=topics&text_id=7368&folder_id=4314&view=text&crit=vluft

[4]www.smhi.se/cmp/jsp/polopoly.jsp?d=5481&a=16769&l=sv&price=true

3 Human exposure to air pollution

Human exposure to ambient air pollution has been associated with severe health effects in many investigations. The most classical example is the London smog (smoke and fog) episode in 1952, during which the mortality rate in the city increased dramatically (Wilkins, 1954). During this episode, the concentrations of particulate matter and sulphur dioxide in London were respectively 56 and 7 times above the normal level at that time. It has been estimated that the episode caused a premature death of 12,000 people.

However, ambient air pollution may cause adverse health effects even at much less extreme conditions. Studies of long-term exposure to air pollution (especially particles) suggest an increased risk of chronic respiratory illness, and of developing various types of cancer. Similarly has higher prevalence of bronchitis, acute cardiovascular decease, asthma and other symptoms been associated with short-term exposure to enhanced air pollution concentrations during episodes.

The population is generally most at risk in urban areas where many people spend a significant part of their time. At the same time the pollution levels are often high in the urban area, due to the high density of pollution sources, and to the poor dispersion conditions resulting from the presence of building obstacles (Berkowicz et al., 1997b) (Paper I).

Over the last couple of decades, the emission from traffic has become the major local source of air pollution in most of the larger European cities. This development is the result of legislation concerning emissions from power plants and industries, together with a steady growth in road traffic. Traffic exhaust fumes contain pollutants such as NO_X, volatile organic compounds (VOC's), carbon monoxide (CO), and particulate matter (PM). In recent years an increasing focus has been on determining the possible association between adverse health effects in the population and various traffic related air pollutants. A significant part of this emphasis has been on particulate matter (Kunzli et al., 2000; Pope and Dockery, 2006).

The most important health effects of air pollution are airway and cardiovascular disease and cancer. The mechanisms involved are thought to be related to inflammation and oxidative stress. In Danish as well as other human exposure studies there has been shown systemic effects of traffic-related particles in terms of decreased endothelial function important for the development of cardiovascular disease as well as oxidative stress-induced DNA damage (Vinzent et al., 2005; Mills et al., 2005). In order to improve our understanding of air pollution related health effects there is a need for detailed exposure assessments of high quality.

3.1 Exposure assessment

Human exposure refers to an individual's contact with a pollutant concentration (Zartarian et al., 1997). However, the assessment of human exposure to air pollution may be carried out in a number of different ways (Hertel et al., 2001) (Paper V):

- Categorical classification
- Application of biomarkers
- Personal exposure monitoring
- Analysis of monitoring network data
- Application of exposure model

Often exposure assessments are based on ambient air pollution data from routine monitoring stations that in some cases may be representing very local conditions, especially when these stations are placed at street level. However, strong relationships are found between measurements in urban background and various adverse health effects (Brunekreef and Holgate, 2002). These associations indicate that urban background is at least in some cases a good proxy for air pollution exposure. In a recent Danish study, strong associations were found between PM_{10} and hospital admissions due to cardiovascular and respiratory disease in the elderly (age≥65), and asthma in children (age 5–18) in Copenhagen, Denmark (Andersen et al., 2007).

Figure 3.1 Equipment for personal monitoring of air pollution. To the left: Radiello passive samplers for collecting nitrogen dioxide (available for other pollutants: **http://www.radiello.it/english/index_en.html**) (Cocheo et al., 1996). To the right: portable active ultrafine particle counter **http://www.tsi.com/documents/3007.pdf**.

The categorical classification is a crude indirect method based on simple exposure indicators such as type of residence, type of job, presence of indoor sources etc. This method is generally considered too crude to be used in epidemiological studies. However, in a number of studies simple exposure proxies are used with success and shown to be associated with adverse health effects. Two examples of such proxies for air pollution exposure are "distance to trafficked road" that has been used in a number of studies (van Vliet et al., 1997; Hoek et al., 2002; Tonne et al., 2007), and "traffic density in the region" which is used in at least one study (Lipfert et al., 2006).

Small portable personal exposure samplers sensitive enough for measuring ambient concentrations are available for a number of pollutants (Figure 3.1). These monitors may be integrated samplers that collect pollutants over a certain period of time and then are returned for laboratory analysis, or they may be continuous monitors that use a self-contained analytical system on location. Both systems may be passive as well as active monitors. Passive diffusion samplers have been widely used in exposure studies, since they are inexpensive and easy to operate (see references in (Hertel et al., 2001) (Paper V)). I will in the following sections return to studies where NO_2 and benzene concentrations have been sampled with passive samplers. Recently an inexpensive new system of automatic monitors has been applied in London for a continuous online monitoring on people travelling in the city. These data are present online on the web[5]. NERI has initiated cooperation with these British researchers. The uncertainty concerning the methodology concerns the precision in the measurements, which is not yet well investigated.

Biomarkers are in this context samples from the human body that provide information about either internal dose or markers of health effects (Heinrich-Ramm et al., 2000). They may e.g. be blood or urine samples analysed for the contents of a pollutant or its degradation products in order to provide information about exposure/dose. However, blood samples may also be analysed for DNA damage in white blood vessels as a measure of effects. Biomarkers have been widely used in the exposure studies referred later in this report.

The use of Land-use regression (LUR) methods is becoming frequent in exposure studies (Jerrett et al., 2005). This is a result of the increasing information stored in digital databases, the substantial computer power that is now available, and the widespread use of Geographical Information Systems (GIS). The basic idea behind LUR is to use information about surrounding physical, land-use and traffic characteristics to generate an exposure proxy for an area. Such an exposure proxy may e.g. be a combination of different simple exposure proxies and generated by the use of a statistical model. The exposure proxy will therefore be strongly adapted to the considered region. In a recent review of intra-urban air pollution exposure models, the authors argue that the use of LUR is relatively inexpensive to implement compared with the more sophisticated air quality models (Jerrett et al., 2005).

Models may serve as useful tools in indirect estimation of human exposure, and may in some cases be the only option. An example: lifetime exposure cannot be measured directly. The models may e.g. be applied for pollutant calculations in various micro-environments where the study subject spends time, or calculations may be performed for a few selected sites for which the pollutant levels are used as proxies for the total exposure. In the following I will discuss a number of Danish studies where the OSPM is used in exposure modelling. A more detailed description of some of these studies is provided in (Hertel et al., 2001) (Paper V). In most of these studies, pollution level at the front door is used as proxy for the air pollution exposure. However, in some of the studies the study subject is tracked along a route through the city.

3.2 The bus driver project

Working in certain occupations is associated with high exposure to ambient air pollution. People with such occupations have a higher risk of pollution related adverse health effects compared with the rest of the population. This counts especially for people who stay in this occupation during a significant part of their life. As an example, it has been found that bus drivers in Copenhagen have a higher prevalence of several forms of cancer when compared to reference groups (Soll-Johanning et al., 1998).

The bus driver project focus on the air pollution exposure of bus drivers in Copenhagen, and use a group of postmen as reference. The aim of the project is to test whether the OSPM may be used for the assessment of occupational air pollution exposure of bus drivers (Wilhardt et al., 1996), e.g. in the epidemiological studies carried out on the cohort of bus drivers in the Greater Copenhagen area. NO_2 measurements are performed using passive diffusion samplers inside a bus, outside a bus, on a postman, and on the bicycle of the postman, during a three-week campaign period in Copenhagen. Calculations with OSPM are then performed for 22 selected street sections along the bus route and the mail route of the postman. Traffic data is obtained from the municipality, and street configuration data is obtained by studies on location.

[5] http://www.escience.cam.ac.uk/mobilesensing

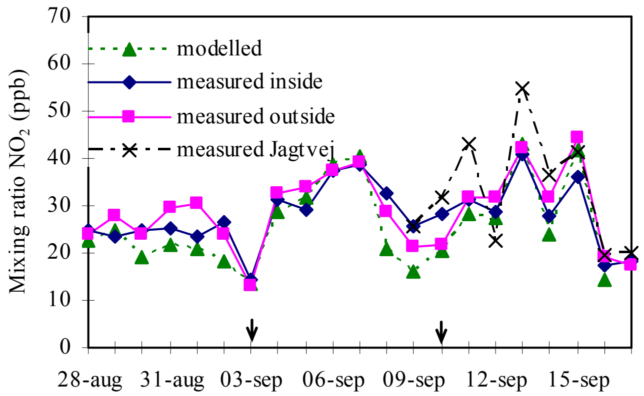


Figure 3.2 Average nitrogen dioxide mixing ratios over a whole working day of the Copenhagen bus driver (8 hours) for the campaign period 28 August – 17 September 1995. The figure shows measurements for the entire working day of the bus driver obtained inside and outside the bus, and results obtained from OSPM calculations representing the average concentrations along the bus route. Average concentrations for the working hours at the monitoring station Jagtvej (one of the streets on the bus route) are shown as well. Arrows on the time axis indicate Sundays. Source (Hertel et al., 2001) (Paper V).

The OSPM reproduce the measurements well (Figure 3.2), and this indicate that the model may serve as a useful tool for exposure assessment in occupational epidemiology studies of bus drivers and postmen (Hertel et al., 1996; Hertel et al., 2001) (Paper V).

3.3 The Children Cancer project

This study is initiated in 1994 (Raaschou-Nielsen et al., 1996). The basic idea is to investigate the possible association between development of cancer in childhood and exposure to air pollution. Exposure at the front door is taken as indicator of the personal exposure of the child. Input data for the OSPM calculations (street configuration and traffic data) is obtained from a questionnaire sent to the local authorities.

The information in the questionnaire is digitised, and a program is developed for interpretation and subsequent generation of input files for the OSPM (Vignati et al., 1997). The system is evaluated against a series of NO_2 and benzene measurements obtained from 204 addresses (Raaschou-Nielsen et al., 2000) (Paper III). The placement of these addresses is depicted in Figure 3.3. A reasonably good agreement between model calculations and measurements is obtained (See Figure 3.4), and the system is subsequently applied for calculations for almost 20,000 addresses of Danish children in the time period 1960 to 1991.

The children cancer study show no association between air pollution exposure and development of childhood cancer, except for lymphoma (restricted to Hodgkin's disease) for which a boarder line significant relationship is found (Raaschou-Nielsen et al., 2001).

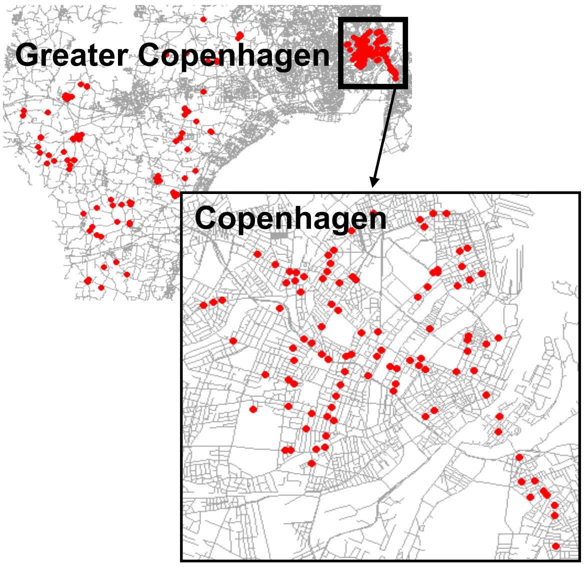


Figure 3.3 The 204 addresses in Greater Copenhagen (103 inside the city and 101 in rural areas) used in the Children cancer project for testing performance of OSPM (Raaschou-Nielsen et al., 2000). Measurements of NO_2 and benzene were performed using passive diffusion samplers.

Generation of input data is a major task when air pollution models like OSPM are applied for exposure assessment for large cohorts in epidemiological studies. Starting with the Children Cancer project, OSPM is used in a number of epidemiological studies. In these studies calculations are performed for thousands of addresses, and collecting street configuration data and traffic data turned out to be a very time consuming task. It is estimated that the municipalities use between two and three man years to fill in the questionnaires in the Children Cancer project. It is unlikely that we can ask for their assistance in providing information in even larger exposure studies to come. This is the motivation for developing the AirGIS system

3.4 The AirGIS system

The AirGIS system (Jensen et al., 2001b) (Paper IV) is based on UBM and OSPM, and the system makes use of GIS, digital maps and data from available Danish registers. The AirGIS system uses available technical and cadastral maps (buildings, roads, address points, property limits) and available Danish national administrative databases on buildings, cadastres and populations. The various available digital maps and registers are applied depending on the purpose of the application and the approach taken to generate the required data. Data on building heights may be obtained in different ways:

a) Object heights above sea level of buildings minus terrain heights.
b) Heights obtained from the Building and Dwelling Registry using property limits to geo-code buildings.
c) Heights obtained from Digital Elevation Models.

Figure 3.4 Illustration of the 2½-dimensional GIS-based landscape model applied for generation of street configuration data in the AIRGIS. The calculation procedure is built on digital maps and data from Danish national registries. The parameters that are determined include; general building height in the area, building height in wind sectors, width and orientation of the street and distances to street intersections. Source: (Jensen et al., 2001b) (Paper IV).

For population exposure assessment, population data can be linked to an individual address using the Central Population Register. The AIRGIS system contains a so-called 2½ dimensional landscape model (Jensen et al., 2001b). The landscape model performs an interpretation of digital maps and information from the available databases, which leads to generation of street configuration data for use as input data in the OSPM calculations (Figure 3.2).

Figure 3.5 shows two different comparisons of OSPM calculations performed for the 204 addresses (see Figure 3.3) where measurements are performed in the Children cancer study. The first set of calculations is the original OSPM runs performed on basis of information from the questionnaires sent to the local authorities. The second set of calculations is based on input data generated from the 2½-dimensional landscape model under the AirGIS system. It is seen that the correlation coefficient is slightly smaller for the second set of calculations, but the AirGIS system may be applied to the entire country without the need to obtain more information from local authorities.

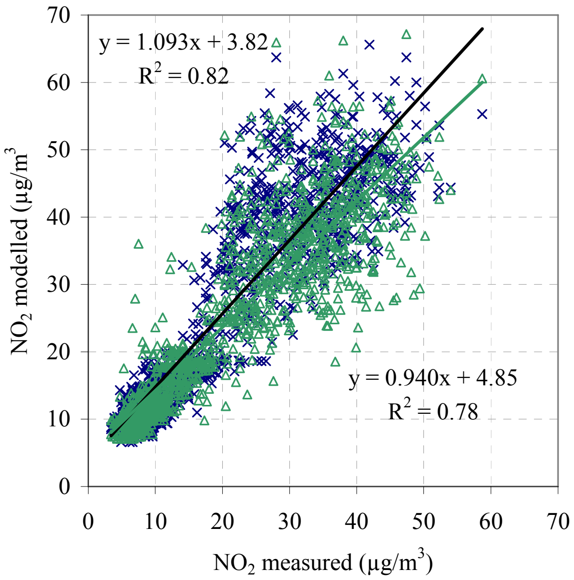


Figure 3.5 Scatter plot for monthly averages of NO_2 modelled versus measured at the 204 addresses (See Figure 4.1) within the Children cancer project. The scatter plot shows two sets of model data: the original questionnaire based OSPM calculations (**x**), and results obtained with the AirGIS system (**Δ**) where the 2½ dimensional model has been applied for generating the street configuration data. Source: (Berkowicz et al., 2008)

Obtaining information about traffic on the road network is one of the key obstacles for generating the needed input data for the OSPM calculations in the AIRGIS system. The administration of the Danish road network has until recently been subdivided into three different levels of administration: municipalities, counties and state. Data for the county and state road network are available in digital form from the Danish Road Directorate, but the Danish municipalities do not have a common database for traffic. However, it turns out that many of the municipalities have traffic databases for their local road network. Most of this traffic data is now – after a long process – entered into the AirGIS system as a result of cooperation between NERI, the Danish EPA, and the Danish Cancer Society.

The advantages of GIS based systems for use in modelling of human exposure to air pollution are widely accepted, and many GIS based systems are currently under development. Recently the basic ideas behind the AirGIS system have been used as outset for the development of an air pollution exposure system for the Macao Peninsula (Tang and Wang, 2007).

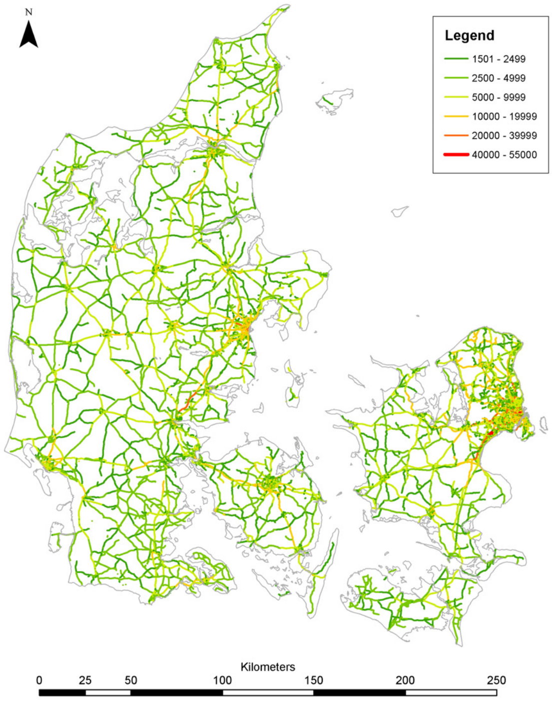


Figure 3.6 Illustration of the traffic data on the Danish road network obtained from the municipalities in the cooperation between the Danish Cancer Society, the Danish EPA and NERI. Only roads with an annual diurnal traffic (ADT) more 1500 vehicles per day have been included in the plot, but all streets in the country are included (although highly uncertain for the smallest streets).

3.5 Studies designed for validation of the AirGIS system

The HOTGAP project is a human air pollution exposure study designed for studying the relationship between air pollution exposure and various biomarkers for health effects (Sørensen et al., 2003c), and for providing data to be used in validation of the AirGIS system. Measuring campaigns of five weeks duration are carried out in autumn, winter, spring and summer during 1999 and 2000.

50 students living and studying in central Copenhagen are recruited for the study. The students carry a backpack with equipment for measuring $PM_{2.5}$ and black carbon (Figure 3.7). This equipment contains a cyclone driven by a pump to separate the fine fraction particles from the course fraction. Radiello passive samplers are placed on the outside of the backpack for measuring NO_2 and BTX (Benzene, Toluene and Xylene). Measurements are performed over 48 hour time periods. Morning blood samples are collected at the end of each 2-day campaign and 24-hour urine samples are collected on the second day of the study period.

The backpack contains a GPS for determining the position of the students. The GPS positions are stored on a Palm computer. In addition to the personal monitoring, measurements are performed inside (in bedroom) and outside (at front door) the home addresses for 30 of the study subjects.

Figure 3.7 Personal monitoring of air pollution exposure in Copenhagen. Upper left corner: Radiello samplers for passive sampling of NO_2. Upper right corner: the equipment in the backpack: cyclone and pump for monitoring $PM_{2.5}$, GPS and Palm computer for tracking position of the students, and FM corrector for higher precision in GPS data. Lower left corner: a student arranging the sampling equipment on a bicycle for sampling at front door. Lower right corner: one of the students carrying the backpack with the equipment. On the backpack is placed a Radiello sampler for NO_2 sampling. Foto: Ole Hertel.

The results of the campaign measurements show that outdoor pollutant concentrations are not always a good predictor of personal exposure (Sørensen et al., 2005a). The results for NO_2 show that the relationship between personal exposure and front door concentration depends upon the season, with a stronger association in the warm season compared with the cold season. For $PM_{2.5}$ and black carbon the same tendency is observed. These findings imply that personal exposure to $PM_{2.5}$, black carbon and NO_2 rely on many factors in addition to the outdoor levels and that information about season or outdoor temperature and residence exposure may be used to improve the accuracy of exposure estimates.

The routes of the students are constructed from GPS data and a digitising of the routes drawn on printed maps. The AirGIS system is applied for computing NO_2 exposures of the students (Figure 3.8).

3.6 Health outcomes of air pollution exposure

Two types of studies concerning health effects of air pollution are carried out as a part of the presented work (Table 3.1). In the first type of studies, dose-response relationships obtained from literature are used to estimate overall health outcomes in the Danish population at current pollutant levels. In the second type of studies, time series of pollutant levels – in some cases measured and in other cases modelled – are linked to observed health outcomes; with the aim of determining the relationships for the Danish population.

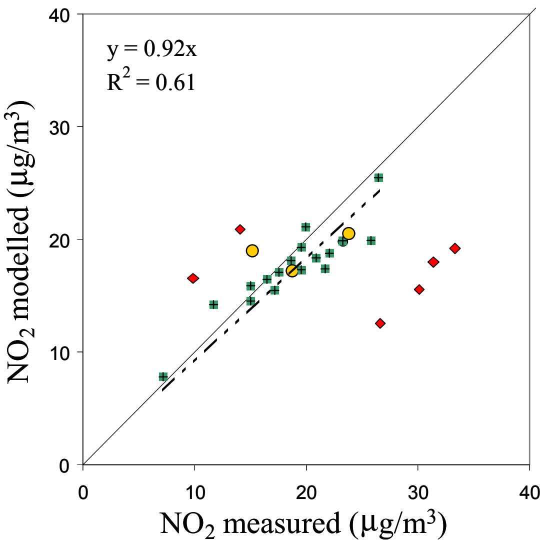


Figure 3.8 Comparison of NO2 exposures for the 48 hours a study subject was traced. Dots marked with ◆ are significantly over- or underestimated values, dots marked with ○ are points with likely errors, and dots marked with ■ are believed to be correctly measured and with good agreement between measurements and model results. Source: (Jensen et al., 2004b).

In the country wide survey on health effects of PM exposure based on the work in a WHO study, American dose-response functions are applied to estimated urban background levels in Danish cities derived on address level for the entire Danish population. The air pollution levels are obtained from analysis of measurements from the urban monitoring programme, and from the use of simple down scaling of the air pollution levels based on the size of the urban area in question relative to the cities in the Danish monitoring programme from which measurements are available. The pollution concentration at address levels is then crudely taken as a proxy for the personal exposure of the residents. According to this study about 5000 premature death, 5,000 incidences of chronic obstructive airways decease, 17,000 incidences of acute air-ways decease, 200,000 asthma attacks and close to 3 million days of illness in the Danish population may be associated with exposure to particle pollution (Palmgren et al., 2001; Raaschou-Nielsen et al., 2002).

Recently a couple of Danish time series studies have been presented. Daily hospital admissions among children and elderly in Copenhagen have been linked to ambient particle levels (Andersen et al., 2007). Associations have been found between ambient PM_{10} levels and three health outcomes, with the strongest association for asthma. The relative risk (RR) for a change of one inter-quartile range in PM_{10} (14µg/m³) with a 4-day lag has been determined to 2.7% (95-% confidence interval 1.3%-4.2%).

The HOTGAP study has several aims. One aim is to provide data for testing the performance of the AirGIS system. The other aim is to determine possible health effects of air pollution exposure in the urban environment. Biomarkers are related to the measured air pollution exposure. The analysis of the association between biomarkers and personal exposure suggest that even moderate exposure to PM may induce oxidative DNA damage (Sørensen et al., 2003b; Sørensen et al., 2003c; Sørensen et al., 2003d; Sørensen et al., 2003e) and that personal $PM_{2.5}$ is more important in this aspect than the ambient $PM_{2.5}$ background concentration.

In a couple of studies exposure estimates have been obtained from OSPM and AirGIS calculations. Until now none of these have shown associations between health effects and air pollution exposure. Calculations have just been made for the 200,000 addresses in the large Danish cancer, diet and health cohort of 50,000 people. Preliminary analyses indicate that a stronger association between air pollution and health effects (unpublished), than what has previous been obtained using measurements from the Danish monitoring programme LMP as proxy (Andersen et al., 2007). However, the analyses are still ongoing and no final conclusions have yet been drawn.

3.7 Exposure scenario studies

Besides providing exposure estimates for use in epidemiological studies, air quality models may be used in various types of exposure scenario studies; studies designed for analysing the impact of various strategies to reduce human air pollution exposure. An example of such a study is given in (Hertel et al., 2008) (Paper VI). In this study it is investigated to what extend a proper choice of route through the city may reduce the exposure to air pollution.

Table 3.1 Selected Danish studies of health effects of air pollution.

Study	Cohort and design	Exposure assessment	Pollutants	Time period	Association or determined health effect	Reference
The children cancer study	Case-control study: 2,500 childhood cancer incidences and 5,000 controls.	Front door pollutant level computed by OSPM using data from questionnaires filled in by municipalities.	NO_2 and benzene	1960-90	No association found, except for a boarder significant association to the rare Hodgkin's disease	(Raaschou-Nielsen et al., 2001)
The HOTGAP study	50 students in four campaigns. Air pollutants linked to biomarkers in blood and urine.	Personal monitoring over 48 hours. Biomarkers of internal dose, biologically effective dose and susceptibility.	$PM_{2.5}$ Carbon black, NO_2, and transition metals in PM sample	1999-00	DNA damage analysed in terms of 8-oxodG in lymphocytes in blood. 11% increase in 8-oxodG per 10μg/m^3 increase in $PM_{2.5}$. 1.9% & 2.2% increase in 8-oxodG per 1μg/L increase in vanadium and chromium, respectively. Lipid peroxidation analysed by malondialdehyde (MDA). 3.7% increase in MDA per 10μg/m^3 increase in $PM_{2.5}$.	(Sørensen et al., 2003b; Sørensen et al., 2003d; Sørensen et al., 2005b)
The Danish particle study	Dose-response functions from literature applied on the entire population; 5.5 million people.	1: Measured urban background scaled by size of city. 2: Distance to trafficked road.	1: PM_{10} .... 2: simple traffic pollution proxy.	2000	1: About 5,000 premature death, 5,000 chronic obstructive airways decease (AWD), 17,000 acute AWD, 200,000 asthma attacks, and close to 3 million days of illness. About 1/3 associated with traffic pollution. 2: about 1500 premature death.	(Raaschou-Nielsen et al., 2002; Palmgren et al., 2005)
The Odense time-series study	Time-series study on the population in Greater Odense area. Include 4158 hospital admissions for asthma & bronchitis.	Measurements from urban background and street in Odense.	TSP, O_3, NO, NO_2, NO_x, and SO_2	1994-99	10μg/m^3 increase in PM_{10} (estimated from TSP) increase hospital admission for respiratory disease by 0.99%. For NO_2 and SO_2 the figures are 1.23% and 1.49%, respectively. Values found for a lack time of 3 days. The Funen only association for PM_{10} & SO_2. No association is found for O_3.	(Sigsgaard et al., 2007)
The RAV pilot study	Case-control pilot study. 33 asthma cases 29 controls	Front door and urban background calculated with the AirGIS system (OSPM/UBM)	NO, NO_2, and CO	1993-02	No association is obtained.	(Hansen et al., 2007)
The Panum bicycle study	Cross-over study on 10 male and 5 female healthy non-smoking subjects.	Bicycling in ambient air in central Copenhagen and in clean air in laboratory.	UFP, PM_{10}, NO, NO_2, and CO	2003	Four fold increase in oxidative DNA damage after bicycling in traffic compared with bicycling indoor.	(Vinzents et al., 2005)
The EXPLUS study	Time-series study on 411 children in the COPSAC study; followed from birth to 18 month.	Measured urban background from HCØ, and street pollution Jagtvej and HCA. Only children from central Copenhagen included.	CO, NO_x, NO_2, O_3, $PM_{2.5}$, PM_{10} and UFP	1998-01	1ppm increase in CO at Jagtvej and HCA increase air-way symptoms (AWS) by 89% and 211%, respectively. Doubling in NO_x at Jagtvej and HCA increase AWS by 37% and 113%, respectively. Boarder significant for CO, NO_x, NO_2 & PM_{10} at HCØ and AWS; 1μg/m^3 increase in PM_{10} at HCØ increase AWS by 1%.	(Andersen et al., 2005)

In this case it is not crucial that the model provide accurate estimates of the exposure of the study subject at one particular hour or day. However, it is important that the model calculations reflect long-term differences between streets with high and low pollutant levels in order to provide a realistic assessment of the long-term differences between the two routes. Comparisons between calculations and measurements from Jagtvej and H.C. Andersen's Boulevard indicate that the model performs well at these sites, especially when long-term pollutant levels are considered (Hertel et al., 2008) (Paper VI). It is therefore reasonable to assume that the long-term differences in exposure are well reflected in the calculations.

The results show that the accumulated air pollution exposure for the low exposure route is between 10% and 30% lower for the primary pollutants (NO_x and CO). However, the difference is insignificant and in some cases even negative for the secondary pollutants (NO_2 and $PM_{10}/PM_{2.5}$). Considering only the contribution from traffic along the travelled streets, the accumulated exposure is between 54% and 67% lower for the low exposure route for both primary and secondary pollutants. The bus is generally following highly trafficked streets, and the accumulated exposure for all pollutants along the bus route is between 79% and 115% higher than the high exposure bicycle route (the short bicycle route). Travelling outside the rush hour time periods reduces the accumulated exposure between 10% and 30% for the primary pollutants, and between 5% and 20% for the secondary pollutants.

The study indicates that a web based route planner for selecting the low exposure route through the city might be a good service for the public. In addition the public may be advised to travel outside the rush hour time period. In general bicyclists have the change to select less exposed routes compared with buses and cars and in addition they get the exercise.

3.8 Conclusions

The aim of this chapter is to address a number of environmental and more technical questions related to human exposure to air pollution. These questions are outlined in the introduction.

A.4. Does the exposure to air pollution constitute a significant health hazard for the population?

A number of exposure assessment studies have confirmed that air pollution is associated with significant health effect in the Danish population. Dose-response functions from American epidemiological studies applied to estimated exposure levels for the Danish population point at an effect that include 5,000 incidences of premature death, the development of 5,000 chronic air-way deceases, 17,000 acute incidences of air-way decease, 200,000 asthma attacks, and 3 million days of restricted activity due to illness. About 1/3 of the effect has been attributed to traffic pollution. Application of a simple proxy (distance to trafficked road) show about 1500 incidences of premature death may be attributed to traffic based pollution; which is well in line with 1/3 of the effect associated with exposure of the population to PM_{10}.

Studies linking observed air pollution levels to biomarkers of health effects show a positive association between $PM_{2.5}$ and pre-stages of cancer in the form of oxidative damage of DNA (expressed by 8-oxodG in blood). As an example was found an 11% increase in 8-oxodG per 10μg/m^3 increase in $PM_{2.5}$.

A.5. What is the actual range in air pollution exposures of the population?

In the previous chapter I have presented the gradients in air pollution concentrations in Denmark. The annual mean NO_x concentration is a factor of 2 to 3 higher in the urban background compared with the rural areas. For NO_2 this range is slightly smaller and for PM the gradient is only significant when the extreme levels are considered (in this case the 98 percentile). The ratio between rural and street level air pollution is up to an order of magnitude for NO_x and a factor of 4 to 5 for NO_2. For PM there is almost no gradient between urban background and rural sites. However, the range in air pollution exposure depends not only on the ambient levels, but also on the time-activity pattern of the considered person or cohort. The air pollution exposure is generally high when a person is commuting in urban streets. Compared with the time people spent indoors, travelling is a minor activity and therefore usually of little impact on the long-term accumulated exposure. Urban background pollutant levels at the address are therefore generally considered as a better proxy for the personal exposure than the pollution level at the front door.

In the 1990ties, the traffic generated pollution was higher compared with the present levels. This is mainly due to the introduction of catalytic converters on gasoline driven vehicles. Annual mean NO_x levels were typically about 25% higher, whereas street level CO was 50 to 100% higher in the most trafficked streets. The air pollution exposure near trafficked streets was similarly larger in the 1990ties compared with the current situation.

Although ambient concentrations in Denmark for many of the air pollutants are significantly lower than what is observed in central Europe as well as in many third world countries, there is still a significant spatial and temporal gradient in many of the pollutants. In combination with the unique Danish register databases, this gradient form the best possible basis for carrying out time series and cohort studies of the associations between exposure and health effects.

A.6. To what extent is it possible to reduce the air pollution exposure by selecting a low exposure route through the city?

The scenario studies show that the accumulated exposure to traffic pollution may be reduced by approximately 30% when a low exposure route is chosen as an alternative to the shortest bicycle route through the city. The study also shows that travelling outside rush hours may reduce exposure by about 20% compared with travelling during the rush hour time periods.

Travelling in urban streets of high traffic density includes being exposed to high concentrations over short periods of time. A typically duration will be 10 to 30 minutes and it will take place many times over a year if the route represent the travel between home and working place. Some of the recent health effect studies of air pollution have shown that exposure to high levels of traffic pollution in Copenhagen may lead to significantly enhanced oxidative damage of DNA in blood (Sørensen et al., 2003d; Vinzents et al., 2005; Bräuner et al., 2007). These and other studies on health effects of short-term air pollution exposures, underline the importance of reducing the peak exposures to air pollution.

B.4. Is it possible based on the street pollution model to construct a human exposure model system suitable for application to large cohorts in epidemiological studies?

The first air pollution calculations for larger epidemiological cohorts in Denmark were performed as a part of the Children Cancer study in cooperation with the Danish Cancer Society. In the Children Cancer project, OSPM calculations of front door air pollution levels for about 19,000 addresses were based on information provided by questionnaires filled in by the municipalities. However, this was a very time consuming procedure. The AirGIS system has made it possible to carry out model calculations for even very big cohorts. Recently model calculations have been made for about 200,000 addresses of the 57,000 persons in the Diet, Cancer and Health cohort.

Until now no associations have been found between health effects and modelled air pollution exposure levels. When the results for the Diet, Cancer and Health cohort have been analysed it will be time to evaluate whether the AirGIS system provides the right set of information for such epidemiological studies. However, the comparisons to measurements indicate that the system reproduces the observed levels reasonably well.

B.5. Do we have access to the necessary input data for applying such systems in larger epidemiological studies?

One of the main obstacles has until now been access to traffic data necessary for calculating urban background and street pollution concentrations. Just recently NERI has in cooperation with the Danish EPA and the Danish Cancer Society established a traffic database containing diurnal traffic for the entire Danish road network. These data form the basis for the recently completed calculations for the Diet, cancer and health cohort.

Very limited information is currently available concerning time-activity pattern in the Danish population. Such information would improve the exposure assessments significantly, although data in some case have been obtained for specific cohorts in the form of questionnaires.

3.9 Fingerprint on science and environmental management

Denmark is in a unique position for carrying out health assessments of air pollution exposures. This unique position has at least two main reasons:

- No other country has access to so extended central registers with health data covering the entire population – diagnoses on hospital admissions, causes of death etc.
- No other country has – at least to my knowledge – a complete inventory with traffic information on all streets as well as digital images of all building in the country.

The AirGIS system makes it possible to perform individual air pollution exposure assessment on address level even for very large cohorts. Recently calculations have been performed for about 200,000 addresses of the 57,000 people belonging to the Danish diet, cancer and health cohort established by the Danish cancer society. Preliminary analyses indicate that the association between address level exposure and health effects is more significant than what has previously been found when measurements from the Danish monitoring programme LMP has been applied as proxy for the air pollution exposure.

4 Atmospheric nitrogen deposition

The Haber-Bosch process for anthropogenic fixation of nitrogen (N), developed in the beginning of the 19th century, was a tremendous break-through for food production world-wide, and thereby also for mankind in general. However, this method for producing mineral fertiliser was at the same time the starting point for a significant change in the global N budgets (Galloway, 1998). It is now clear that biologically active N compounds in high amounts can have significant negative impact on the environment. The negative impact of enhanced N deposition was first documented in the Netherlands when damages were found on trees in forests in the vicinity of stables with high ammonia (NH_3) emissions. It is now common to talk about a global N imbalance of the environment, and it has been documented that deposition of biologically active N compounds may have significant negative impact on both terrestrial and marine ecosystems (Nosengo, 2003). Even a small increase in the N supply may in some cases have significant negative impact (Galloway et al., 2004).

The Dobris assessment in the beginning of the 1990'ties showed that the critical loads and levels for atmospheric N compounds were exceeded over large parts of Europe (EEA, 1995). The Third Assessment showed considerable improvements were obtained during the 1990ties concerning the pressures on European nature. However, the critical loads were still exceeded for more than half of European ecosystems (EEA, 2003).

In Denmark, the impact of atmospheric N on terrestrial and marine ecosystems is known to be significant. Episodes of oxygen decificts in bottom waters, in worst case situations followed by subsequent death of fish and bentic fauna, are frequent phenomena, appearing almost every year, in the inner Danish waters (Bach et al., 2005). These episodes are strongly linked to the anthropogenic N loads, of which current estimates have shown that about 30% of the bioavailable N arise from atmospheric loadings (Spokes et al., 2006). More than 70% of Danish terrestrial ecosystems have an atmospheric deposition that exceed the critical loads (Bach et al., 2005). For the most sentitive Danish terrestrial ecosystems, calculations have shown that even the atmospheric background (far from local NH_3 sources) N deposition exceeds critical loads (Hertel et al., 2003a). These model calculations show that Danish sources currently contribute by 40 - 45% of the background N deposition over land (Ellermann et al., 2006).

The current deposition to Danish land surfaces is in the order of 10 to 30 kg N/ha/year (Ellermann et al., 2006). A Swedish group has computed the development in atmospheric nitrogen deposition over the time period 1800 and up to 2000 (Figure 4.1). It is seen that in 1800, the deposition of NH_x has been about 10% of the value for the late 1990ties, and for NO_y (NO_x plus reaction products) the similar figure is about 3%. Using these curves, the pre-industrial atmospheric N deposition to Danish terrestrial ecosystems may be estimated to have been in the range between 1 to 2 kg N/ha/year.

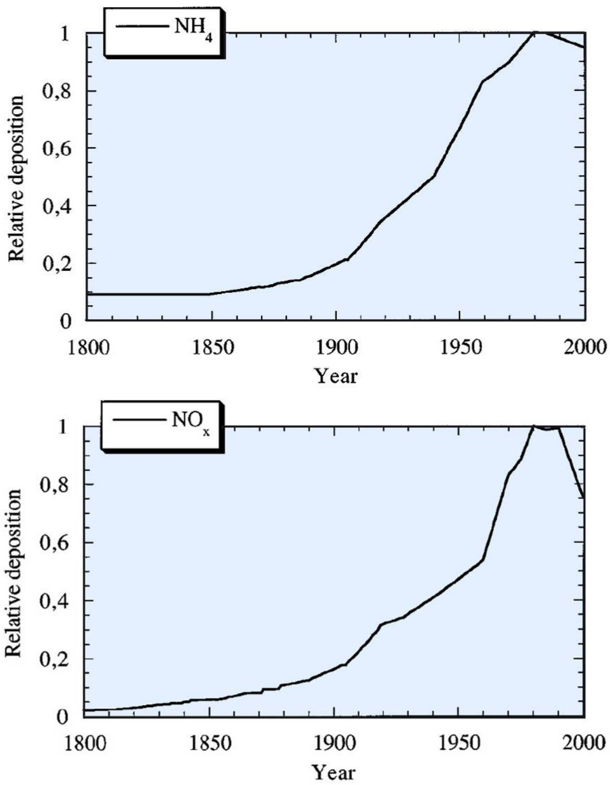


Figure 4.1 Atmospheric nitrogen deposition curves derived from historical emission inventories and model calculations. The curves represent relative depositions to terrestrial ecosystems over the time period 1800 to 2000. Source: (Alveteg et al., 1998).

The sensitive terrestrial ecosystems have critical loads in the range of 5 to 10 kg N/ha/year (see table 6.1). These systems are therefore currently under substantial pressure from the atmospheric anthropogenic loadings. I will return to this in the sections to come.

4.1 Deposition of reactive N compounds

The atmospheric deposition of reactive (and biologically active) N compounds arises from two different groups of compounds in the atmosphere (Figure 4.2). The first group is ambient NH_x and consists of gas phase NH_3 and particulate ammonium (NH_4^+). Agricultural activities leading to the emission of NH_3 constitute the far most dominant source of NH_x in the atmosphere (Skjøth et al., 2004) (Paper X). The other group consists of the nitrogen oxides (NO_y). These compounds are formed mainly from free nitrogen (N_2) in combustion processes related especially to traffic, power production, and industry.

Near agricultural sources, the dry deposition of NH_3 is often the largest contribution to the atmospheric N deposition. The fraction of locally emitted NH_3 that is deposited in the nearby area of the source depends on the type of surface, the atmospheric stability and the concentration of acid gases and aerosols in the atmosphere. The latter include sulphuric acid (H_2SO_4) and nitric acid (HNO_3) that take part in fast reactions with NH_3 and form aerosol-bound NH_4^+.

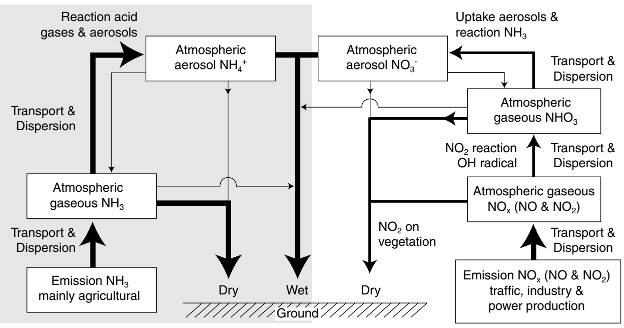


Figure 4.2 Main path ways of reactive N compounds in the atmosphere. The left side of the figure: the atmospheric path ways of NHx (gas phase ammonia (NH_3) and particle phase (NH_4^+)) compounds, and the right: path ways of the NO_y (NO_x and reaction products) compounds. NH_3 is emitted mainly from agricultural sources. In the atmosphere the N compounds are subject to transport and dispersion, but also scavenging by dry deposition and by transformation to particle bound NH_4^+ in reactions with acid gases and particles. Aerosol bound NH_4^+ has generally a long lifetime in the atmosphere and may be transported long distances (>1000km). The NH_4^+ containing particles are mainly removed by wet deposition. Nitrogen oxides (NO_y) are emitted as NO_x (nitrogen monoxide (NO) and nitrogen dioxide (NO_2)), where also these compounds are subject to transport and dispersion. NO_2 may be dry deposited to the vegetation, but is mainly scavenged from the atmosphere by reaction with OH radical in the formation of nitric acid (HNO_3). HNO_3 has a very short lifetime in the atmosphere, since it is quickly scavenged by uptake in particles, reaction with NH_3 or dry deposition (it sticks to any surface – ambient particle as well as ground). Uptake in ambient particles or formation of new particles by the reaction with NH_3 leads to particle bound nitrate (NO_3^-). Also NO_3^- containing particles (some times the same particles that contain NH_4^+) are mainly scavenged from the atmosphere by wet deposition. Particle phase NH_4^+ and NO_3^- may under certain circumstances be released back to gas phase NH_3 and HNO_3. Source: (Hertel et al., 2006b) (Paper XI)

The NO_y compounds are emitted as NO and NO_2 (see also section 2.3.2). The NO_2 dry deposits to land surfaces (actually to vegetation), but with a limited rate. The NO_y compounds are therefore mainly removed from the atmosphere after chemical conversion. NO_2 is converted to HNO_3 with a typical reaction rate of about 5% per hour (Hertel, 1995). HNO_3 dry deposits very fast, but it is also quickly taken up on aerosol surfaces and converted to long-range transported nitrate (NO_3^-). Thus, the biologically active N compounds take part in a series of chemical reactions in the atmosphere that have to be described in a transport-chemistry model for atmospheric N deposition.

The majority of the long-range transported N compounds in the atmosphere is in the form of particulate NH_4^+ and nitrate (NO_3^-). The particulate N compounds are mainly associated with particles in the size range with an aerodynamic diameter slightly smaller than 1 μm (Pakkanen et al., 1996), which is the size range with the smallest dry deposition velocity (Ruijgrok et al., 1995). Particles in this size range are mainly removed from the atmosphere by wet deposition processes. A very significant part of the atmospheric N deposition in background areas is thus related to wet scavenging of particle-bound N compounds (Hertel et al., 2003a).

A transport-chemistry model aimed at calculating atmospheric N deposition to nature has to describe the governing processes for both NO_y and NH_x compounds in the atmosphere. This is the aim in the development of the ACDEP model.

4.2 The Atmospheric Chemistry and DEPosition (ACDEP) model

The Atmospheric Chemistry and DEPosition (ACDEP) model (Hertel et al., 1995) (Paper VII) is developed within the Danish EPA's Marine Research Programme Sea90. The aim is to develop a tool for mapping atmospheric N deposition to Danish marine waters. The ACDEP is a Lagrangian trajectory model that originally is based on the one-dimensional (1DIM) model (Hertel et al., 1994), which was developed for describing the chemical degradation in the atmosphere of biogenic sulphur compounds emitted from the sea.

In the ACDEP model transport, chemical transformation and deposition processes are computed following an air parcel along 96-h back-trajectories. The air parcel is divided into 10 vertical layers from ground and up to 2 km height (Figure 4.3). Transport of the entire air parcel is assumed to follow the σ-level 0.925 wind (approximately 800 m above ground) disregarding wind shear (wind turning with height). The concept behind ACDEP captures the most important boundary layer processes, although wind shear with height is disregarded (Stohl, 1998). The advantages of ACDEP as an easy to apply model for atmospheric deposition calculations are generally accepted, and in 2001 a version of the model – named ACDEP-Asia – is developed by a Japanese group and applied for calculating sulphur deposition in Asia (Carmichael et al., 2001). During transport, the air column in the ACDEP model receives emissions from the sources it pases, and vertical mixing, chemical transformation, and dry and wet deposition take place. A specific parameterisation of the dry deposition of N compounds to the sea surface is developed (Hertel et al., 1995) (Paper VII).

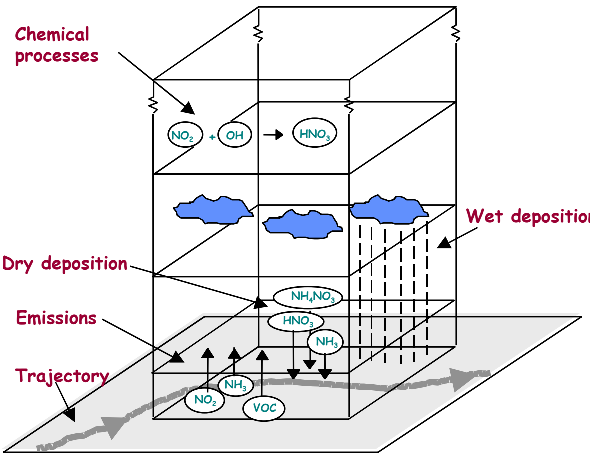


Figure 4.3 Sketch to illustrate the principles behind the Lagrangian Atmospheric Chemistry and DEPosition (ACDEP) model. An air package represented by a vertical column is advected along a 96-h back-trajectory to a selected receptor point. During the transport the air column receives emissions of air pollutants from sources, the pollutants are mixed in the vertical by turbulent diffusion, chemical transformations take place and the pollutants are deposited by dry and wet deposition. Source: (Ellermann et al., 1996).

The ACDEP model contains a full chemical mechanism of 35 species and 80 chemical reactions based on the widely used CBM-IV (Gery et al., 1989a; Gery et al., 1989b). The CBM-IV is originally developed for describing photochemical smog and includes therefore the chemistry of nitrogen oxides and the most important hydrocarbons. In ACDEP, the CBM-IV mechanism is extended with NH_3 and NH_4^+ chemistry to account for all the reactive N compounds in the atmosphere. The applied chemical solver – the Eulerian Backward Iterative (EBI) method - is developed for handling the chemical mechanism (Hertel et al., 1993). The EBI method is considered to have the best accuracy/speed ratio among a variety of commonly applied solvers (Huang and Chang, 2001). The description of wet deposition processes is based on state-of-the-art parameterisations at the time the model is developed. In the first version of the ACDEP model, the trajectory programme from the EMEP model is applied, but later a new and more flexible trajectory programme has been constructed (Skjøth et al., 2002). The overall principles behind the model are illustrated in the simple sketch in Figure 4.3.

The ACDEP is a parameterised model where certain processes are simulated using simple approximations and procedures (like in the case of the OSPM). An example is the parameterisation of the horizontal dispersion which is indirectly modelled (Figure 4.4) by performing an averaging of the emissions which is depending on the distance to the receptor point.

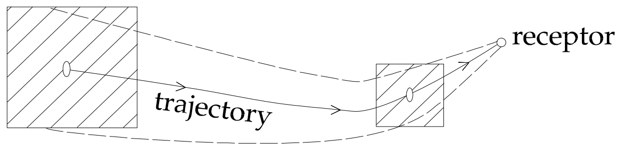


Figure 4.4 Sketch illustrating the simplified indirect parameterisation of horizontal dispersion in the ACDEP model. The full line curve is the trajectory of the one-dimensional model column. The patterned squares represent the areas over which emissions are averaged. The sides of the square are defined as 1/10th of the travelled distance along the trajectory to the receptor point. Source: (Hertel et al., 1995) (Paper VII).

It is a good assumption that a plume in average has a width of one tenth of the travelled distance (Hanna et al., 1982). In the handling of the emissions received by the model column in ACDEP, an average is taken over a square where the sides in the square are one tenth of the travelled distance. This is done in order to take into account emissions from the area that due to the horizontal dispersion will give contributions to the pollution at the receptor point. This is a rather crude simplification since the dispersion of a plume depends not only on the travel distance but also on the meteorological conditions. However, this simple parameterisation is shown to work well in the ACDEP model (Hertel, 1995) (Paper VII).

In the first mappings of atmospheric N depositions to the open Danish marine waters, a 2-layer version of the ACDEP model is applied. This version of ACDEP is used for reducing the calculation time, since the computer resources are limited. For receptor points placed some distance from NH_3 sources, only little difference is obtained between the results with the 2-layer and the 10-layer version of ACDEP (Ellermann et al., 1996). Later when computer resources increased, only the 10-layer version of the ACDEP is applied.

4.3 Validation of the ACDEP model

The ACDEP model has routinely been validated by comparisons with measurements from the Danish background monitoring programme as well as with measurements from the EMEP programme at various monitoring sites all over Europe. Table 4.1 shows the comparison between modelled and "observed" atmospheric N depositions at the monitoring stations in the Danish background monitoring programme. The "measured" dry deposition is here obtained by multiplying calculated dry deposition velocities to measured ambient air concentrations. The model calculations are obtained with the ACDEP after the implementation of the new NH_3 emission module (see Section 4.6). The results are for the year 2003, but similar comparisons have been made in the background monitoring programme over the years 1999 to 2004. In 2003, the comparison shows that for the land stations the modelled total deposition is within the range -20% to +30% from the "observed" deposition. The model tends to overestimate the wet deposition, and this is believed mainly to be due to uncertainties in the precipitation amounts. For Anholt the model overestimates the NH_3 concentrations and this result in a 40% overestimation of the dry deposition. Based on the comparisons over the years, it has been estimated that the total deposition to the open marine waters is determined with an uncertainty of ±30% to 40% and the deposition to land surfaces with an uncertainty of ±50% to 60%.

The results of the ACDEP calculations are shown to be highly sensitive to the initial concentrations. In first ACDEP calculations performed under the Danish EPA's Marine Research Programme Sea90, the air column is given an initial concentration of 1 ppbv for the long lived aerosol compounds ammonium nitrate (NH_4NO_3) and ammonium sulphate ($(NH_4)_2SO_4$). These initial concentrations are chosen since they give the best model fit to measurements from the Danish monitoring stations, when the results are evaluated by comparison to one years measurements for the year 1990. Up to 50% of the concentration of the aerosol compounds at the receptor points over Denmark is found to be related to the initial concentration given to the air column at the beginning of the trajectory (Hertel, 1995). Since the history of the air mass is important for the realistic initial concentration, and this in general must be considered to be significantly lower for an air mass arriving from the North Sea compared with an air mass arriving from the European continent, the procedure is in 1994 modified so that the air column is initialised using calculated concentrations from the Danish Eulerian Model (DEM) (Zlatev et al., 1992).

An analysis of the calculated atmospheric N deposition shows that the results are very sensitive to the amounts of precipitation. The coarse resolution in meteorological input (150 km x 150 km) obtained from EMEP is thus one of the major uncertainties in the ACDEP calculations in the 1990ties. Especially the resolution and quality of precipitation data are shown to be crucial (Skjøth et al., 2002).

The air pollution forecasting system THOR (Brandt et al., 2001a; Brandt et al., 2001b) is established in 1999, and in full operation in 2000. The THOR system is based on a series of the air pollution models developed at NERI. Four times a day the THOR system produces a 3-days air pollution forecast. Meteorological data are provided from calculations with the weather forecasting models Eta and MM5. Thereby the THOR system produces meteorological data on a 39 km x 39 km grid. The Eta and MM5 calculations are based on coarse resolution meteorological data obtained from NCEP in the US. Air pollution calculations are performed with DEOM on 50 km x 50 km grid. The air pollution and the meteorological data produced by the THOR system is applied to the ACDEP calculations in the mapping of N deposition. The higher resolution in the meteorological input data are seen to significantly improve the model performance (Skjøth et al., 2002).

Table 4.1 Comparison of "observed" and modelled N depositions to Danish marine and terrestrial ecosystems in 2003. Unit: kg $N/km^2/year$ (divide by 100 to convert to kg N/ha/year). Observed (obs) depositions are constructed from measured wet depositions and dry depositions computed from measured diurnal mean air concentrations (filter pack measurements) multiplied by calculated dry deposition velocities. Dry deposition velocities are based on actual meteorological data. Depositions modelled with the ACDEP after implementation of the new NH_3 emission module. Source (Ellermann et al., 2004).

	Dry deposition		Wet deposition		Total deposition			NHx fraction		Wet dep. fraction	
	obs	model	obs	model	obs	Model	Diff (%)	obs	model	obs	model
Water											
Anholt	77	242	598	771	675	1013	40	47	44	89	76
Keldsnor	270	329	611	659	881	988	11	63	50	69	67
Land											
Anholt	398	600	598	771	996	1371	32	39	30	60	56
Frederiksborg	560	627	724	1079	1284	1706	28	42	37	56	63
Keldsnor	1048	745	611	659	1659	1404	-17	55	36	37	47
Lindet	1360	1035	1005	1414	2364	2449	4	64	59	42	58
Tange	974	947	749	1169	1723	2116	20	60	57	43	55
Ulborg	537	742	736	1008	1272	1750	32	51	53	58	58

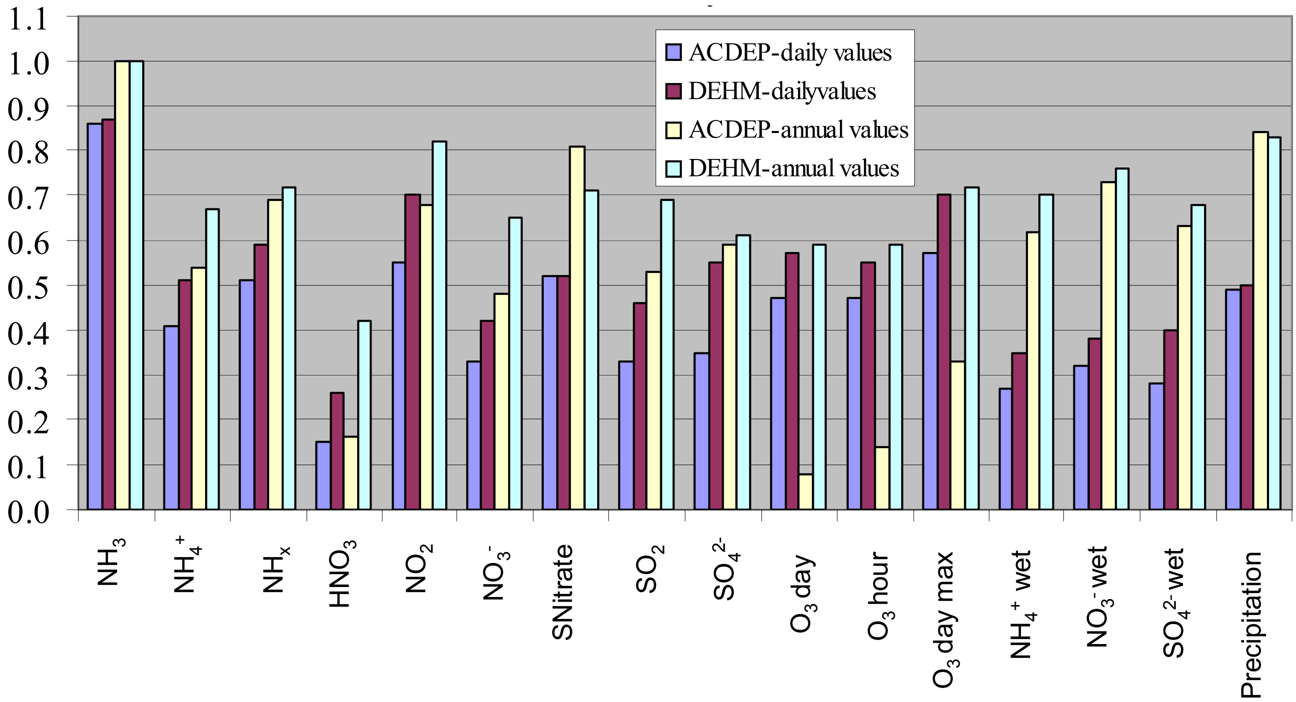


Figure 4.5 Correlation coefficients for corresponding sets of modelled (ACDEP & DEHM) and measured concentrations of reduced N (NH_3, NH_4^+ & the sum NH_x), nitrogen oxides (HNO_3, NO_2 & SNitrate (sum of HNO_3 & $NO3^-$), sulphur (SO_2 & SO_4^{2-}), diurnal, hourly and daily maximum O_3 (O_3 day, O_3 hour & O_3 day max), and wet deposition of ammonium, nitrate and sulphate (NH_4^+ wet, NO_3^- wet & SO_4^{2-} wet) and precipitation. Data for 208 EMEP monitoring stations in year 2000. Source: (Frohn et al., 2007).

Table 4.2 Comparison of ACDEP, DEHM and OML-DEP to current state-of-the-art in transport-chemistry modelling with focus on atmospheric N deposition. Extended from (Hertel et al., 2006b) (Paper XI)

Process	Long-range transport			Local scale	
	State-of-the art	**ACDEP**	**DEHM**	**State-of-the-art**	**OML-DEP**
Emissions	Inventories for Europe on 17km x 17km resolution. For parts of the domain with dynamic seasonal variation in emissions.	Inventories for Europe on 17km x 17km resolution. For parts of the domain with dynamic seasonal variation in emissions.	Inventories for Europe on 17km x 17km resolution. For parts of the domain with dynamic seasonal variation in emissions.	Inventory on single farm level with dynamical seasonal variation and distributed on contributions from stables, manure application, evaporation from crops etc.	Inventory on single farm level with dynamical seasonal variation and distributed on contributions from stables, manure application, evaporation from crops etc.
Transport	Eulerian 2-way nested grid models with 17km x 17km or even 5km x 5km resolution in the inner nest.	Lagrangian model using 96h back-ward trajectories. Advection of a vertical column of 10 vertical grid cells along the trajectory.	Eulerian 2-way nested grid models with 17km x 17km or 5km x 5km resolution in the inner nest (depends on application).	For process studies CFD modelling provide detailed information e.g. about flow around buildings. Most local scale models are Gaussian plume models.	Gaussian plume model. A research version contains an improve description of the transport.
Aerosol phase	Dynamical aerosol phase chemistry in size bins. Nucleation, condensation, evaporation, aerosol phase chemistry.	Aerosol phase handled as gas phase compounds. Indirect simulation of aerosol phase processes in simple 1st order reactions.	Aerosol phase handled as gas phase compounds. Indirect simulation of aerosol phase processes in simple 1st order reactions.	First order transformation between gas phase NH_3 and aerosol phase NH_4^+.	First order transformation between gas phase NH_3 and aerosol phase NH_4^+.
Gas phase	Explicit chemical mechanisms including most important species.	Carbon Bond Mechanism IV (CBM-IV) extended by the NH_x chemistry. The applied mechanism contains 35 species and about 80 reactions.	Explicit chemical mechanism with 63 chemical species and about 120 chemical reactions.	First order transformations.	First order transformations.
Wet deposition	Full wet phase chemistry in cloud and rain droplets and subsequent scavenging of rain droplets.	Incloud and below cloud scavenging coefficients.	Incloud and below cloud scavenging coefficients.	Not included. Of limited importance on the short time scale for the local scale processes.	Not included.
Dry deposition	Resistance method with detailed seasonal variation in surface resistances and accounting for current meteorological conditions. Handling of bi-directional flux of NH_3, NO_2 and a few other compounds.	Resistance method without seasonal variation in surface resistance. Account for current meteorological conditions. Special module based on Slinn and Slinn for handling conditions over sea. No handling of bi-directional flux.	Resistance method including seasonal variation in surface resistances based on the parameterisation implemented in the EMEP model (Simpson et al., 2003). No handling of bi-directional flux.	Resistance method with detailed seasonal variation in surface resistances and accounting for current meteorological conditions. Handling of bi-directional flux of NH_3, NO_2 and a few other compounds.	Resistance method including seasonal variation in surface resistances based on the parameterisation implemented in the EMEP model (Simpson et al., 2003). No handling of bi-directional flux.

4.4 The transition from ACDEP to DEHM

Lagrangian models are computationally efficient when calculations are performed for a limited number of receptor points. However, a general problem concerning the application of Lagrangian models is that the uncertainty of the trajectories increases significantly with distance to the receptor point (Stohl, 1998). In ACDEP, 96 h back-trajectories are applied, but shorter trajectories could in principle have been applied. Lagrangian model have been shown to have difficulties in describing the dispersion conditions when a front is passing (Stohl et al., 1998).

In Eulerian models, the high demand for computer resources has previously been an obstacle concerning high resolution. However, with the increasing available computer power this has become less problematic (see the comparison between Lagrangian and Eulerian models shown in Table 4.2).

The increasing computer resources available together with the considerations about how well the two models describe atmospheric transport, has been the background for substituting ACDEP with DEHM – the Danish Eulerian Hemispheric Model (DEHM) (Christensen, 1997; Frohn et al., 2001; Frohn et al., 2002). DEHM is a further development of DEM and DEOM, but with a number of improved parameterisations and use of nested grid.

Before substituting ACDEP with DEHM as the routine tool within the background monitoring programme, a series of comparisons between the two models are analysed (Frohn et al., 2007). In this comparison, the coarse resolution version of the DEHM model is applied (one nest with a resolution of 50km x 50km). The results from the two transport-chemistry models are compared with measurements from a number of selected EMEP monitoring stations over Europe. The comparisons show that the DEHM achieve better correlation coefficients for all measured pollutants (16 in total) when daily mean values are analysed (Figure 4.5). For the annual mean values 15 out of 16 pollutants are better reproduced by DEHM compared with the ACDEP calculations.

One of the difficulties for the ACDEP model concerns the annual mean O_3 concentration. The model does not contain a seasonal variation in surface resistance applied in calculation of the dry deposition velocities. The applied values for the surface resistance means that dry deposition of O_3 in the winter season is significantly overestimated, which is making the O_3 concentrations too low in this season. This affects the NO_3^- concentration as a result of a too low conversion rate from NO_2 to HNO_3 (producing NO_3^-). The observed differences are thus not only a result of the description of atmospheric transport. However, the conclusion from these comparisons is that the DEHM model performs generally better than the ACDEP model. These results supported the move from ACDEP to DEHM within the background monitoring programme.

4.5 The Danish Ammonia Modelling System (DAMOS)

The Danish Ammonia Modelling System (DAMOS) is a combination of a long-range-transport model and a local scale model describing the contribution from NH_3 emissions released from single farms (Hertel et al., 2006a) (Figure 4.6). In the first version of DAMOS, the long-range transport component is computed by the ACDEP model. However, in the most recent version of DAMOS, the DEHM has substituted the ACDEP model. A comparison to current state-of-the-art of ACDEP, DEHM and OML-DEP is given in Table 4.1.

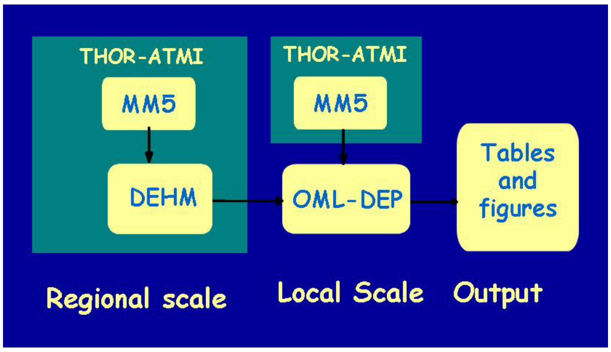


Figure 4.6 Sketch to illustrate the Danish Ammonia Modelling System (DAMOS) after the DEHM model has substituted ACDEP for the long-range transport component. Source: Based on (Hertel et al., 2006b) (Paper XI) and (Hertel et al., 2006a).

OML-DEP the Gaussian local scale transport-deposition model is developed from the OML-model (Olesen, 1995), which is used in the regulation of chimney heights on polluting industries, power plants etc. in Denmark (Danish EPA, 2001). For the handling of dry deposition of NH_3, the OML-DEP is making use of the surface depletion method from (Horst, 1977). The model uses a pseudo first order reaction velocity for the conversion of NH_3 to NH_4^+ (Asman et al., 1989).

The DEHM calculations are performed for the entire Northern Hemisphere with 2-way nesting in several sub-domains; the outer domain using a 150km x 150km resolution. For Europe a 50km x 50km resolution is applied, and for Denmark and nearby areas a 16.67km x 16.67km resolution is used. These calculations are based on the meteorological model MM5 (Grell et al., 1994). The local scale model OML-DEP is applied for a 16km x 16km domain that covers the nature area of interest for which detailed deposition mapping is needed (see the model performance depicted in Figure 4.7).

The DEHM background concentrations of NH_3 and sulphur dioxide are obtained for each hour by using interpolation between up to three grid cells upwind from the OML-DEP domain. OML-DEP calculations are performed for 40 x 40 receptor points evenly distributed over the domain and each representing a 400m x 400m area. The dry deposition velocities of NH_3 are in DEHM and OML-DEP performed with the same module. This dry deposition module is based on the parameterisations of the EMEP model (Simpson et al., 2003). The NH_3 emissions are computed using the parameterisations with high spatial and temporal resolution (Skjøth et al., 2004; Gyldenkærne et al., 2005) (see section 4.6). The high resolution in the inventories has shown to be very important for the model performance (see the discussion in (Hertel et al., 2006b) (Paper XI)).

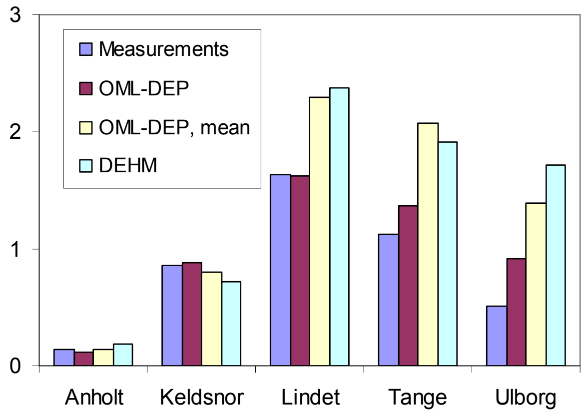


Figure 4.7 Measured and calculated NH_3 concentrations (µg NH_3-N/m^3) at five Danish rural stations in 2005. The DEHM calculations represent the mean value for the 16.67 km x 16.67 km cell where the station is placed. For OML-DEP two sets of calculations are shown: One (OML-DEP, Mean) is the average for the 16 km x 16 km domain covering the main part of the grid cell of the DEHM receptor net. The other (OML-DEP) is for the 400 m x 400m grid cell where the station is placed. Measurements of NH_3 on Anholt and Tange are performed with filter pack samplers, whereas measurements at Keldsnor, Lindet and Ulborg are performed using the denuder method. Source: (Ellermann et al., 2006; Hertel et al., 2007a) (Paper XII).

4.6 High resolution emission inventories

The analyses performed within the Sea90 project show that the quality as well as both the temporal and spatial resolution of the applied emission inventories is crucial for the quality of the calculations of atmospheric N deposition (Hertel, 1995). In these early ACDEP calculations, a highly simplified diurnal and seasonal variation in pollutant emissions is applied. Sinus functions developed for application in the original Lagrangian version of the EMEP model (Hov et al., 1994) are here adopted in the ACDEP. Comparisons of model calculations with measured ambient air concentrations of NH_3 show that the sinus function does not reflect the emission pattern for Danish conditions (Figure 4.8). This is the background for developing a new dynamical NH_3 emission model for Denmark and nearby surrounding areas (Skjøth et al., 2004) (Paper X) (Gyldenkærne et al., 2005).

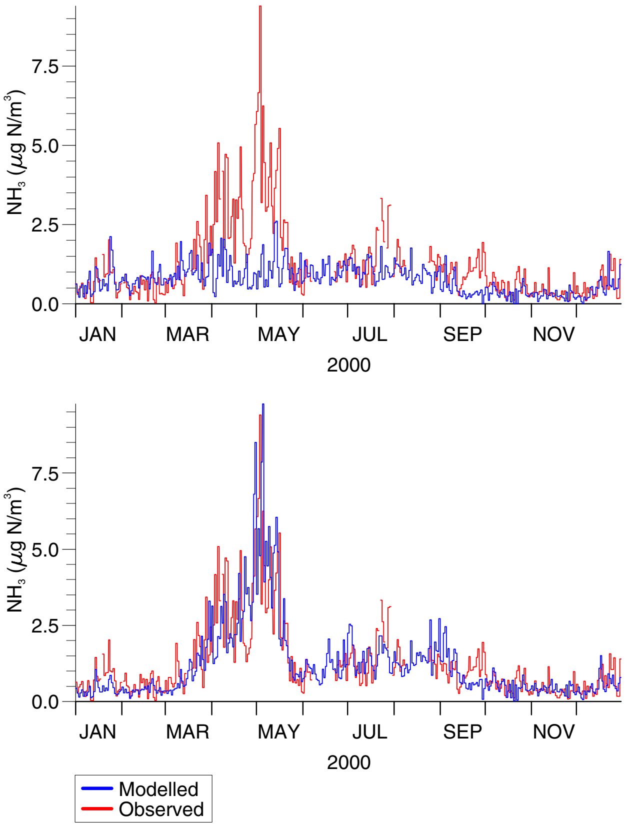


Figure 4.8 Comparison of diurnal mean ambient air NH_3 concentrations for Tange in 2001. ACDEP model calculations performed using the original emission model for NH_3 (upper figure) and the new high temporal and spatial resolution emission model for NH_3 from Danish sources (lower figure). Model calculations are shown in blue and measurements in red. The correlation coefficient between model results and measurements improve from 0.28 to 0.67 when the new emission module is implemented. After refinement of the emission model an even better performance is obtained. Source: (Skjøth et al., 2004) (Paper X).

The dynamic NH_3 emission model is constructed in such a way that the total annual emission is preserved, and only the seasonal distribution is modified. This is a great advantage when the emission module is applied in various models. This makes it possible to perform calculations preserving the official emission inventories from EMEP (www.-emep.int) and CORINAIR. The emission model is shown to work well in the ACDEP model (Figure

4.8). Subsequently the emission model has been implemented in both models under the current version of the DAMOS system (DEHM and OML-DEP), which is now applied in the Background Air Quality Monitoring Programme as routine tool for mapping local N deposition (Ellermann et al., 2006).

It is shown that the dynamic NH_3 emission model may be used in the analysis of the impact of changes in Danish agricultural praxis on local ambient air NH_3 concentrations (Skjøth et al., 2006) due to changes in agricultural praxis as a result of changes in Danish legislation.

The developed dynamical NH_3 emission model is currently under implementation in the EMEP model (Fagerli et al., 2007).

4.7 Conclusions

The aim of this chapter is to introduce the assessment of atmospheric nitrogen deposition to marine and terrestrial ecosystems.

B.6. How well does a combined regional scale and local scale model system work?

The ACDEP reproduces reasonably well the "observed" N depositions to Danish land and sea surfaces. However, a model inter-comparison showed that the DEHM generally performs better than ACDEP. This has motivated a shift from the ACDEP to DEHM as the routine tool for mapping the regional N depositions within the DAMOS system as well as in the Danish background monitoring programme (BOP). Calculations performed with OML-DEP within the DAMOS system has showed that a local scale model is needed for mapping of local atmospheric N depositions to natural and semi-natural ecosystems in Denmark. Even 10 to 15 kilometres away from agricultural sources, the influence on local NH_3 concentration and depositions is significant. The comparisons to measurements from Danish monitoring stations have shown that the DAMOS system reproduces very well the observed NH_3 concentrations.

B.7. How important is the spatial and temporal variation in ammonia emissions for a proper assessment of the local atmospheric nitrogen deposition?

The analyses have shown that NH_3 concentrations in Denmark are strongly influenced by the local emissions. Changes in the Danish legislation have forced the farmers to apply manure during growth season for the crops. These changes are strongly reflected in the ambient air concentrations of NH_3 concentrations over Denmark (Skjøth et al., 2006). Analyses within the Danish background monitoring programme has shown that application of the local scale OML-DEP model significantly improves the results compared with the regional scale DEHM. In order to map depositions to local nature areas with high precision, a local scale model has to be applied.

4.8 Fingerprints on science and environmental management

The Eulerian Backward Iterative (EBI) method is developed for solving the stiff system of ordinary differential equations describing atmospheric chemistry in transport-chemistry models. The EBI has been considered to have the best accuracy/speed ratio among a variety of commonly applied solvers (Huang and Chang, 2001) (in these tests it is referred to as the Hertel solver). The EBI has been selected as standard method by the US-EPA.

A Japanese research group has used ACDEP as frame for developing the ACDEP-Asia model for calculating sulphur deposition in Asia (Carmichael et al., 2001). The concept behind the ACDEP model has been described as the best way of capturing the most important boundary layer processes in the Lagrangian frame (Stohl, 1998).

The registers of Danish livestock animals, budget on animal waste as well as on the production of crops are unique. These registers have made it possible to derive the most detailed NH_3 emission inventory currently available. The parameterisation of the seasonal variation in the NH_3 emissions in Denmark are state-of-the-art in this area (Pinder et al., 2007). There is substantial interest for this parameterisation from other research groups in Europe, and the NH3 emission module is currently under implementation in the EMEP model (Fagerli et al., 2007).

5 Regional scale nitrogen deposition

The coastal seas contain some of the most valuable resources on the planet, and have a number of different uses. They are used for fisheries, recreation, waste disposal, as well as for providing mineral resources. The ecosystems in the coastal waters are in many areas under tremendous pressure from anthropogenic pollution loads (Nixon, 1995; Jickells, 1998). Increased incidences of harmful algae blooms and other eutrophication phenomena are linked to increased inputs of nutrients to the coastal sea. As a result, oxygen deficits and subsequent death of fish and benthic fauna have become frequent phenomena in many coastal areas (Meyer-Reil and Koster, 2000). It is well documented that the oxygen deficits are linked to the algae blooms (Ærtebjerg, G., Andersen, J.H. and Hansen, O.S., 2003). When the algae production in the sea is high, a result is that large amounts of dead algae deposit at the bottom. The oxygen in the bottom water of the sea is subsequently consumed in the degradation of the algae. It has been estimated that a 25% reduction in the overall input of N to the Danish estuaries would lead to a 50% reduction in the number of days with severe oxygen depletion (Møhlenberg, 1999). In this context, the high anthropogenic input of N is of special concern, and the algae growth in these coastal waters is generally limited by the supply of N (Kronvang et al., 1993; Paerl, 1995).

From the beginning of the 19th century and up to the 1980ties oxygen deficits and subsequent death of fish and benthic fauna have become frequent phenomena in European coastal waters (Meyer-Reil and Koster, 2000), and this tendency has continued up to now. In Danish coastal water, these phenomena are seen almost every year (Ærtebjerg, G., Andersen, J.H. and Hansen, O.S., 2003). On this background, the ACDEP model (see Section 5.2) is developed for assessment of N deposition to Danish marine waters. Later ACDEP is applied also for assessment of N deposition to the Baltic Sea and the North Sea within EU funded research programmes MEAD (Spokes et al., 2006) and ANICE (de Leeuw et al., 2001).

For terrestrial ecosystems, N is a nutrient that may be taken up directly through the leaves or indirectly through the root system as N-rich soil water (Sparks et al., 2001; Sparks et al., 2003). The N uptake may affect the competitive relationship between the organisms in such a way that more nutrient-demanding types of plants can enter the ecosystem. If a terrestrial ecosystem is saturated with N, it leads to a wash out of nitrate (NO_3^-) from the soil (Theobald et al., 2004), an effect that is well-known in agricultural areas (Kyllmar et al., 2005).

It has been shown that critical loads are exceeded for more than 70% of Danish terrestrial ecosystems (Bach et al., 2005). For the most sentitive Danish terrestrial ecosystems, calculations have shown that even the atmospheric background deposition of N exceeds critical loads (Hertel et al., 2003a). Calculations also show that Danish sources contribute 40 - 45% of the background deposition over land (Ellermann et al., 2006).

5.1 N deposition to Danish marine waters

The Sea90 calculations performed in 1994 show that the atmosphere in 1990 contribute 30 to 40% of the overall N load of the Danish Kattegat Strait (Asman et al., 1994a; Asman et al., 1995). The fraction related to atmospheric N deposition is here computed relative to the contribution from river run-off, and disregarding the contribution from deep water entrainment of nutrient rich bottom water as well as nutrient fluxes from adjacent marine waters. The deposition calculations are performed for a 30km x 30km grid covering the Danish as well as the Swedish part of the Kattegat Strait.

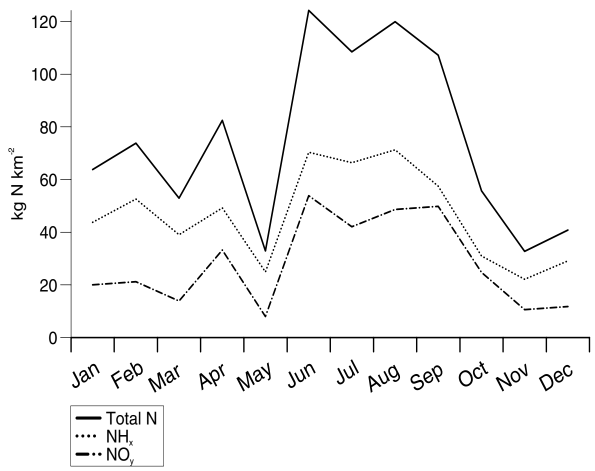


Figure 5.1 Calculated monthly N depositions to the Kattegat Strait for the year 1990. Calculations performed with the ACDEP model as a part of the Danish Marine Research Programme Sea90. Shown is the contribution from NH_x and NO_y as well as the total N deposition. The figures are given in kg N/km^2/month, but they may be converted to kg N/ha/month by dividing by 100. Source: (Asman et al., 1995).

The meteorological data applied for the ACDEP calculations above are data generated for use in the Lagrangian EMEP model (Eliassen et al., 1982), which up to the 1990ties is applied for the EMEP calculations in the mapping of air pollution in Europe. The meteorological data on 150km x 150km grid are kindly provided by the Norwegian Meteorological institute, and the applied trajectory model is furthermore also obtained from the Lagrangian EMEP model.

The calculated seasonal variation in N deposition to the Kattegat Strait in 1990 is shown in Figure 5.1. The analyses of the results show that a very significant part of the atmospheric N contribution is related to wet deposition. The main part of the wet deposited N arises from atmospheric N compounds in aerosol phase (NO_3^- and NH_4^+). Analyses furthermore show that the magnitude of the wet deposition to a large extent is governed by the precipitation amounts. The average N deposition in 1990 is calculated to be in the order of 9.6 kg N/ha/year for the Kattegat Strait. The NHx compounds contribute about 42% of the N deposition, and NOy compounds about 58%. Danish sources contribute 35% of the NHx and 7% of the NOy deposition (Asman et al., 1995).

5.2 The Background Air Quality Monitoring Programme

The available computer resources at NERI increase in the period shortly after the Sea90 programme is completed. Over the next years the ACDEP model is step by step integrated as a routine tool within the Danish Background Air Quality Monitoring Programme for mapping of atmospheric N deposition to Denmark. In the first step dry deposition of N compounds is computed by applying dry deposition velocities from the dry deposition module of ACDEP to measured ambient air concentrations at the measurement stations.

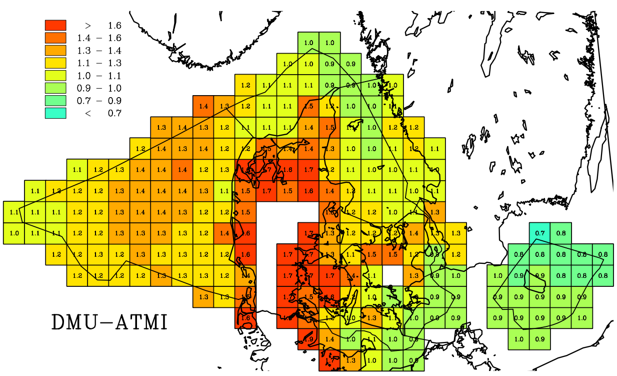


Figure 5.2 Totat N deposition (kg N/km^2/year) to Danish marine waters in 2003 calculated with the ACDEP model as a part of the Danish Background Air Quality Monitoring Programme. Source: (Ellermann et al., 2004)

The model is used for mapping atmospheric N deposition to Danish marine waters (Ellermann et al., 1996). The ACDEP calculations under Sea90 are restricted to the Kattegat Strait, but within the Background Air Quality Monitoring Programme calculations are over the time period 1994 to 2004 extended to include all Danish marine waters. From 1999 these calculations are further extended to include depositions to land surfaces. The calculations are performed by extending the 30km x 30km grid developed for the calculations of depositions to the Kattegat Strait in the Sea90 project. The specific depositions to each of the Danish marine waters are subsequently computed from redistributing the results from the 30km x 30km grid to the specific areas of the various Danish marine waters. For this purpose digital maps and GIS- based routines are applied to compute the fraction of a grid belonging to specific Marine waters for use in the redistribution. The calculation procedure is described in a technical report (Ellermann et al., 1996). The depositions to land surfaces are distributed on municipality and county level using a similar distribution procedure as for the marine waters.

Table 5.1 shows the reported atmospheric N depositions in the Background Monitoring Programme to the Danish land and water surfaces during the time period 1994 to 2005. The total atmospheric N deposition to Danish marine waters is computed by ACDEP to be in the range between 100.000 and 140.000 tonnes N. In 1997, low precipitation amounts lead to a significantly lower deposition. In 2004, ACDEP is substituted with DEHM. For this year, the results from both models are presented, and it is seen that the DEHM results are almost 30% lower than what is found with ACDEP. Calculations for the country in general show values that over land typically are 20 to 25% lower than what is found by ACDEP. The background for the change is e.g. that DEHM is considered to provide a better description of the atmospheric transport. The spatial resolution in the DEHM calculations is here coarser (50km x 50km) than in the ACDEP calculations (performed for the 30km x 30km receptor net). In the subsequent year DEHM calculations have been made with a second inner nest for Denmark and close surroundings of 16.67km x 16.67km resolution. In this DEHM calculation for 2005, the total N deposition is seen to decrease even further compared with the ACDEP calculation for 2004 (and previous years).

Table 5.1 Computed total annual N depositions to Danish land and sea surfaces reported within the Background Monitoring Programme 1994 to 2005. ACDEP calculations for 1994 to 2004, and DEHM-REGINA calculations for 2004 and 2005. The values are in 1000 tonnes N/year. NR: not reported. Sources: (Skov et al., 1995; Skov et al., 1996; Ellermann et al., 1997; Frohn et al., 1998; Skov et al., 1999; Ellermann et al., 2000; Ellermann et al., 2001; Ellermann et al., 2002; Ellermann et al., 2003; Ellermann et al., 2004; Ellermann et al., 2005; Ellermann et al., 2006; Ellermann et al., 2007).

	ACDEP											DEHM		
	1994	**1995**	**1996**	**1997**	**1998**	**1999**	**2000**	**2001**	**2002**	**2003**	**2004**	**2004**	**2005**	**2006**
Land	NR	NR	NR	NR	NR	90	92	87	80	85	77	68	64	72
Sea	128	102	101	80	100	120	140	118	107	124	138	107	76	97

1997 was a year with very little precipitation, whereas 2000 and 2003 are years with high precipitation amounts.

Table 5.2 Comparison of ACDEP calculations for the Baltic Sea with results reported in literature. Unit: kg N/km^2/year. Source: (Hertel et al., 2003b) (Paper IX).

Estimate	Year(s) of estimate	Dry deposition	Wet deposition	Total deposition	Source
Observed	1980	482	482	963	(Rodhe et al., 1980)
Observed	1981-82	382	900	1282	(Ferm, 1984)
Observed	1982-83		780		Referred in (Lindfors et al., 1991)
Modelled	1983		790		(Joffre, 1988)
Observed	1984-85		730		Referred in (Lindfors et al., 1991)
Observed	1983-85	80	872	952	(HELCOM, 1988)
Modelled	1985			619	(Iversen et al., 1989)
Observed	1980-86	149	850	999	(Lindfors et al., 1991)
Modelled	1988			687	(Heidam, 1993)
Modelled	1988			660	(Iversen et al., 1990)
Modelled	1989			601	(Iversen et al., 1990)
Modelled	1989			629	(Heidam, 1993)
Modelled	1990	271	688	959	(Asman et al., 1995) [a]
Modelled	1990			675	(Heidam, 1993)
Modelled	1994			530	(Marmefelt et al., 1999)
Modelled	1997			501	(Tarrason and Schaug, 1999)
Modelled	1999	150	534	684	(Hertel et al., 2003b) (Paper IX) [b]

[a] Obtained from the Danish EPA's Marine Research Programme Sea90, and the model was here only applied for the Kattegat Strait.

[b] The ACDEP calculations performed within the EU funded MEAD project.

Table 5.3 Comparison of the ACDEP calculations for the North Sea with results reported elsewhere in literature. Unit: kg N/km^2/year.

Type of estimate	Year(s)	Dry deposition	Wet deposition	Total deposition	Source
Observed	1988-89	548	443	991	(Rendell et al., 1993)
Modelled [a]	1990			455	(Asman and Berkowicz, 1994)
Modelled	1999	144	686	830	(Hertel et al., 2002) (Paper VIII)

[a] For the southern part of the North Sea – estimate for the entire North Sea 551 kg N/km^2/year.

Two changes are here introduced: The dry deposition module is substituted with a parameterisation based on the module developed for the latest version of the EMEP model. Furthermore, the meteorological data are now generated using the MM5 weather forecast model instead of the Eta model.

Figure 5.2 shows the annual atmospheric N depositions to the Danish marine waters in 2003 computed with the ACDEP model (Ellermann et al., 2004). Long-range transported N compounds from the northern part of the European continent result in a South-North gradient. However, the contribution from Danish sources makes this gradient less evident in the figure. Furthermore the areas over Jutland in the western part of the country have higher depositions than the eastern parts of the country. This is a result of local NH_3 emissions in the areas with intense agricultural activity, and a result of higher precipitation amounts in the eastern part of the country. These differences are in agreement with the observations from the measurements stations.

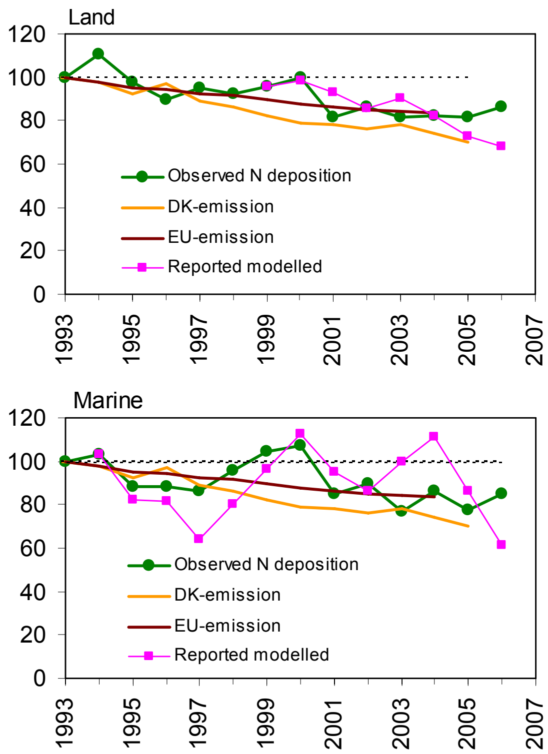


Figure 5.3 The relative trend in "observed" and modelled atmospheric N deposition. For the modelled values, the figures represent what has been reported in the annual reports under the monitoring programme. Shown are also Danish and European N emissions according the official inventories reported to EMEP. To the left deposition to land surfaces, and to the right depositions to Danish marine waters. ACDEP values 1993 to 2004, and DEHM values for 2005 and 2006. Source: see Table 5.1.

Figure 5.3 shows the trend in N deposition to Danish land and marine surfaces. The "observed" depositions represent values constructed from measured wet depositions and dry depositions obtained from measured ambient air concentrations multiplied by computed dry deposition velocities. Together with these values are shown also the computed N depositions as they have been reported in the annual reports within the background monitoring programme over the years. Calculations have for some of the years been updated in subsequent annual reports, but in the plots only the originally reported values are shown. For the land surfaces, it is seen that the reported modelled values reproduce well the "observed" values. However, the results are less good for the marine waters. The general trend in N depositions is seen to follow the development in emissions. The trend in Danish emissions in the beginning of the 1990ties decreased more rapidly than the emissions of the rest of Europe, but since then the trend has followed the same pattern.

One interesting way to look at the atmospheric N is make a budget for Denmark and compare the sum of the atmospheric N depositions to Danish marine and terrestrial areas with the total Danish atmospheric N emissions. As an example, the ACDEP calculations show that the total N deposition to Denmark in 2003 is on the order of 85,000 tonnes N to land surfaces and 124,000 tonnes N to marine waters (Table 5.1). This means that the total N deposition to the Danish areas make up 208.000 tonnes N, which may be compared with the Danish atmospheric emissions of reactive N compounds in 2003 of 144,000 tonnes N (83.000 tonnes NH_3-N and 61,000 tonnes NO_x-N) (Source: NERI www.dmu.dk-/emissioner).

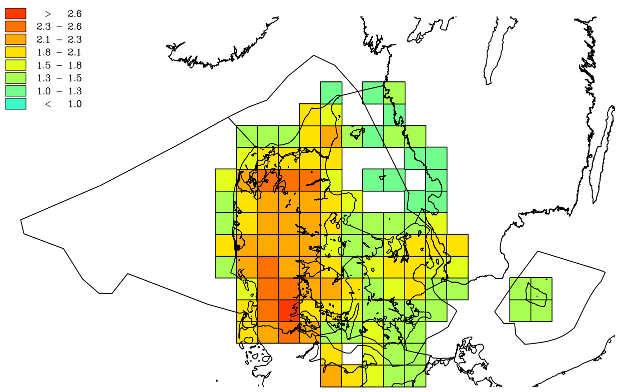


Figure 5.4 Atmospheric N deposition to Danish land areas in 2001. Calculated with the ACDEP model. Source: (Ellermann et al., 2002).

In total this means a net import of N compounds for Denmark in 2003 of about 60,000 tonnes N. The recent calculations with DEHM point at a somewhat lower total deposition to Denmark. The budget for 2005 thus point at a balance between atmospheric deposition of N compounds, and the N emissions from Danish sources (Ellermann et al., 2006).

Figure 5.4 shows the computed atmospheric N deposition to Danish land surfaces in 2001. Again the results reflect the Danish NH_3 emissions in the areas with intense agricultural activities. Danish sources are calculated to contribute 16 and 39% of the N deposition to marine waters and terrestrial surfaces, respectively. Our studies show that the contribution from Danish sources to deposition in Denmark is almost solely from ammonia emitted from agricultural activities (Hertel et al., 2003a). Nitrogen oxides have to be converted to nitric acid before rapid deposition takes place, and most of the nitrogen oxides are transported out of Denmark before this conversion has taken place. A minor contribution is from NO_2 being dry deposited to vegetation, but only few percent of the atmospheric N deposition are related to the Danish NO_x emissions.

5.3 Deposition to the North Sea

The ACDEP model is in 2002 applied for calculating N and S depositions to the North Sea (Hertel et al., 2002) (Paper VIII) (de Leeuw et al., 2003a) within the EU funded ANICE project (de Leeuw et al., 2001). The ANICE project aims at looking at the productivity as function of atmospheric N input (de Leeuw et al., 2003b), and includes a series of field studies conducted from a research platform just off-shore from the Dutch coast line. Furthermore gaseous and particulate N compounds are measured for one year onboard a ferry travelling between Germany, the Netherlands and United Kingdom (Tamm and Schulz, 2003). Finally N depositions to the entire North Sea area are mapped using the ACDEP model (Hertel et al., 2002) (Paper VIII), and more detailed local scale calculations are performed with the METRAS model for a limited sub-domain of the North Sea (Schlunzen and Meyer, 2007).

Results for the Danish marine water show that the N deposition calculations are highly sensitive to the spatial resolution in the emission data (Skjøth et al., 2002). A new emission inventory with high spatial resolution is therefore constructed in 2002 in co-operation with the University of Stuttgart. This inventory is based on the EMEP 50km x 50km inventory, but applied a true sub-grid of 16.67km x 16.67km using data from the EUROTRAC GENEMIS programme (9 grid cells per EMEP 50 km x 50 km grid cell). The derived 16.67km x 16.67km emission inventory is presented in (Hertel et al., 2002) (Paper VIII).

Table 5.4. Dry, wet and total deposition of nitrogen (tonnes N) and sulphur (S) to the North Sea in 1999. Source: (Hertel et al., 2002) (Paper VIII).

	Dry	Wet	Total
Nitrogen	108,000	513,000	622,000
Sulphur	190,000	286,000	476,000

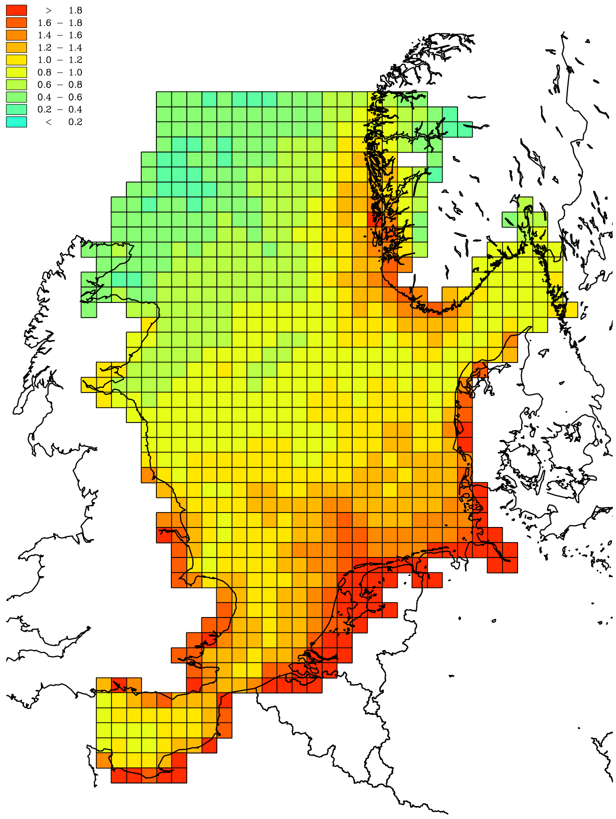


Figure 5.5 The total atmospheric nitrogen deposition to the North Sea in 1999. Depositions are given in Tonnes N per km^2. The results to a large degree reflect the distribution of the source areas around the North Sea, but also the distribution of precipitation plays an important role. For example, see the large depositions at the Norwegian coast, which reflect the very high precipitation amounts due to the mountains in this region. Source: (Hertel et al., 2002) (Paper VIII).

The ACDEP atmospheric N deposition calculations show a strong south – north gradient with N depositions of more than 20 kg/ha/year close to the Dutch coast and down to about 2 kg/ha/year in the most northern part of the North Sea (Figure 5.5). The average atmospheric N deposition to the North Sea in 1999 is calculated to be in the range of 9 kg N/ha/year. The total atmospheric N deposition to the North Sea is calculated to 622,000 tonnes N (Table 5.4).

The measurements from the research platform demonstrate that air-sea fluxes of NH_3 may be upwards and downwards (Quinn et al., 1988; Quinn et al., 1996; Lee et al., 1998; Barrett, 1998) depending on the NH_4^+ concentration and pH in the surface waters (Asman et al., 1994b). A parameterisation of the air-sea exchange of NH_3 developed by (Asman et al., 1994b) is implemented in the ACDEP model. This parameterisation is shown to improve slightly the model performance in comparison with measurements. Furthermore the results show that the computed fluxes to the coastal sea may be modified by up to about 20%, leading to a redistribution of the N depositions (Sørensen et al., 2003a). A new project has just been initiated in investigate the impact of the air-sea exchange of NH_3 for the atmospheric N deposition to Danish marine and terrestrial areas. In case this study shows a significant redistribution of atmospheric N deposition, this parameterisation will be implemented also in the version of the DEHM model that will be applied in the future calculations under the Danish background monitoring programme.

The onboard ferry measurements provide measurements of the long-term concentration gradients over the sea. Measurements are performed as monthly samples in different sections of the ferry route. Different sampling devices are started and stopped depending on the position of the ferry. The current position of the ferry is here determined using a GPS device. The results from the ACDEP calculations are reproducing reasonably well the observed gradients in the aerosol compounds (unpublished).

5.4 Deposition to the Baltic Sea

The ACDEP model is applied for scenario studies of atmospheric N deposition to the Baltic Sea (Hertel et al., 2003b) (Paper IX) within the EU funded MEAD project (Spokes et al., 2006). The aim of the MEAD project is to look at episodic events with high inputs of atmospheric N compounds and explore whether these episodes are able to trigger algae blooming in the marine waters. In a scenario part of the MEAD project, ACDEP is applied to evaluate to what extent the high N deposition events will be reduced as a result of emission reductions on both Danish and European scale.

A kind of spin-off from the MEAD project is the participation in the "NO COMMENTS" project. The aim of NO COMMENTS is to develop an operation system for forecasting algae blooms in the Baltic Sea. An automatic model procedure is established in a direct coupling of ACDEP to the air pollution forecasting system THOR at NERI (Hertel et al., 2003b) (Paper IX). Forecasted N depositions are transferred to the Danish Hydraulic Institute (DHI) and here they are used as input data for their marine ecology models.

The ACDEP model calculations of ambient air concentrations and wet depositions of N compounds are performed for a number of monitoring stations in the EMEP programme. The comparisons indicate that the model reproduce reasonably well the observed ambient air concentrations of NH_x (see the example in Figure 5.6), whereas total nitrate (HNO_3 + NO_3^-) is overestimated by up to 50% for some stations (not shown here).

Table 5.5 The impact of reducing NH_3 and NO_y emissions in Europe and Denmark on the atmospheric N deposition to the Kattegat Strait. Scenario studies performed under the MEAD project using the ACDEP model. Source: (Spokes et al., 2006).

Scenario	1999	2000	2001
No Danish emissions	-20%	-17%	-24%
-50% NH_3 emissions	-21%	-22%	-19%
-50% NO_x emissions	-24%	-25%	-21%
-50% SO_x emissions	-2%	-6%	-1%
-50% NH_3 & NO_x emissions	-43%	-41%	-40%
-50% NH_3, NO_x & SO_x emissions	-43%	-41%	-41%

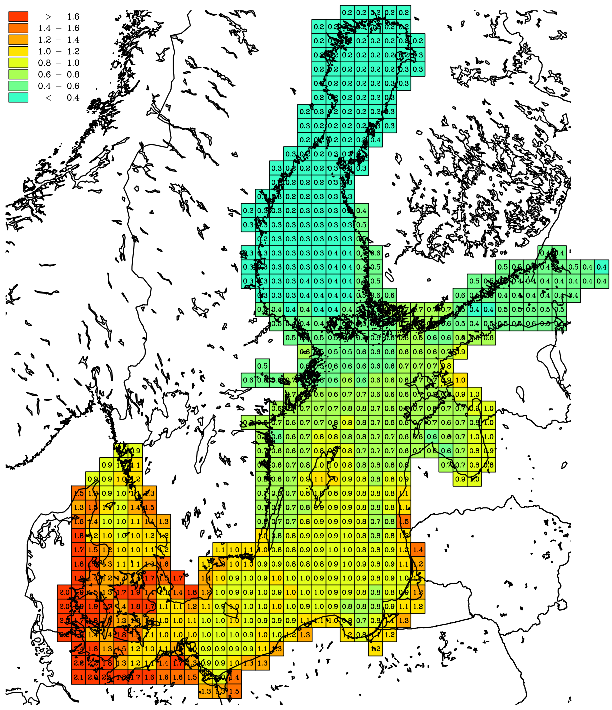


Figure 5.6 Computed N depositions to the Baltic Sea in 1990. Calculations performed with the ACDEP model for a 30km x 30km grid. Meteorological data obtained from the Eta model run within the Danish forecasting system THOR. Source: (Hertel et al., 2003b) (Paper IX).

The total atmospheric N deposition in 1999 to the Baltic Sea is calculated to 318.000 tonnes N, which is equivalent to an average deposition in the order of 7 kg N/ha/year. The calculations show a strong south-north gradient in the atmospheric N deposition. The atmospheric N deposition is found to be about 13 kg N/ha/year close to the coastline and decrease to about 3 kg N/ha/year to the far north of the Baltic Sea (Figure 5.6). In literature various average atmospheric deposition estimates for the Baltic Sea are found for the time period 1980 to 1999 (Table 5.2). Largest values are for the 1980ties with values in the range 7 to 13 kg N/ha/year, whereas values for the 1990ties are in the range 5 to 9 kg N/ha/year.

An important element of the MEAD project is to perform emission reduction scenario studies (Table 5.5). The scenarios are carried out by comparing ACDEP calculations for a basis scenario and for various reduction scenarios for European and Danish emissions of NH_3 and NO_x. The results indicate an almost linear relationship between the atmospheric N emissions and the atmospheric N deposition to the Kattegat Strait. A 50% reduction in either NH_3 or NO_y emissions in Europe is seen to lead to a reduction in the atmospheric N deposition to the Kattegat Strait of between 20 and 25%. When both NH_3 and NO_y emissions are reduced, the atmospheric N deposition is found to decrease by 40 to 45%. Almost no effect on atmospheric N deposition is seen of reducing the European sulphur emissions.

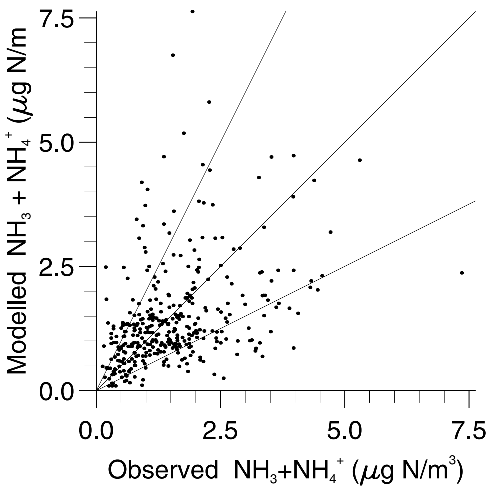


Figure 5.7 A comparison from the MEAD project of modelled and measured NHx at the monitoring site Preilia in Lithuania in 1999. Diurnal mean values are used in the comparison. Correlation coefficient 0.4, measured mean value 1.54 and modelled mean value 1.46 (360 observations). ACDEP calculations are performed using Eta meteorological data obtained from the THOR air pollution forecasting system at NERI. Source: (Skjøth et al., 2004) (Paper X).

As a part of the MEAD project, a mass balance for bio-available N is constructed for the inner Danish waters (Spokes et al., 2006). For this mass balance, transport by water flows in and out of the water domain is accounted for. Furthermore, only the bio-available fraction of N is considered. The results show that the atmospheric contribution is about 30% of the input of bio-available N compounds to the inner Danish waters (See Figure 5.8). The conclusion is that although atmospheric N compound do not seem to be able to trigger algae blooming, it is still a very important part of the overall N load of Danish marine waters. Thereby it is also important for the eutrophication of the coastal waters.

A retrospective study under the MEAD project of extreme atmospheric and marine nitrogen fluxes and chlorophyll-a level is carried out for the Kattegat Strait. Atmospheric nitrogen inputs are obtained from ACDEP calculations. The study show that the N pole released from deep water entrainment episodes greatly exceed the input from wet deposition episodes (Hasager et al., 2003). It is concluded that the atmospheric input even in the most extreme cases is insufficiently high to trigger algae blooming.

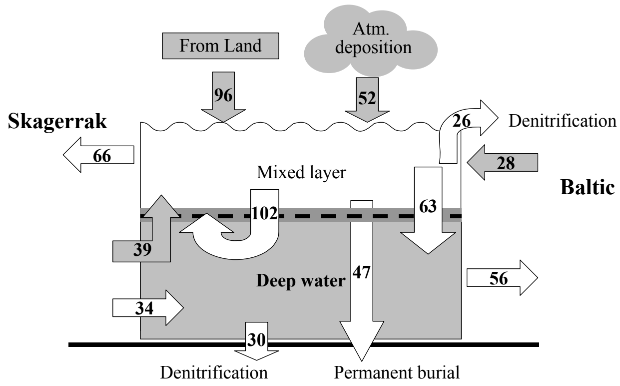


Figure 5.8 Budget of biologically active N compounds for the Kattegat Strait. The budget is an average for period 1999 to 2001. The atmospheric input is computed with the ACDEP model. Units for the shown figures are 1000 tonnes N per year. Source: (Spokes et al., 2006).

At the Department of Marine Ecology, NERI, a series of laboratory studies are carried out in 2004. In these experiments marine algae samples are fed by various levels of N in order to determine the necessary nutrient load to trigger blooming (Spokes et al., 2006). Our model studies show that wet deposition provides the largest atmospheric inputs both with regards to overall load (e.g. on annual basis) and with regards to largest deposition input. The ACDEP calculations indicate that the largest wet deposition episodes lead to inputs in the order of 1.0 to 1.5 kg N/ha/day (Hertel et al., 2003b). The highest loads are found for the Danish marine waters. Based on these deposition estimates, the conclusion is therefore that the atmospheric episodic N inputs are insufficient in magnitude to trigger algae blooming in the Baltic Sea (Spokes et al., 2006). However, the atmospheric N deposition contributes significantly to the overall load, and it is thus of high importance for the eutrophication problems observed in the Baltic Sea.

5.5 Conclusions

This chapter has treated the long-range transport component of atmospheric reactive N. A significant part of this work has been the introduction of air pollution models as routine tools within the Danish background monitoring programme, but also the application of models for N deposition calculations to the Baltic Sea and the North Sea. The chapter addresses the following environmental questions:

A.7. Does the atmospheric nitrogen deposition play a significant role in eutrophication – including algae blooming – of the marine waters?

The atmospheric load has been shown to contribute an amount of about 1/3 of the bio-available N input to the inner Danish waters. There is no doubt that atmospheric N deposition contributes significantly to the eutrophication of coastal marine waters. The relative importance of this contribution is currently increasing since the contribution from river run-off has been reduced as a result of waste water treatment and a reduced loss from agricultural fields.

The analyses in the MEAD project showed that the extreme episodes with the highest atmospheric N input arise from wet deposition episodes. Such episodes have been calculated to give an input in the order of 1.0 to 1.5 kg N/ha/day. Laboratory studies performed at Marine Ecology department of NERI showed that such loads are insufficient to trigger algae blooming episodes.

A.8. To what extend are terrestrial ecosystems threatened by atmospheric nitrogen deposition?

The ACDEP calculations performed in 2003 indicated that even the long-range transport component of the atmospheric N deposition exceed the critical load of the sensitive Danish terrestrial ecosystems. The deposition was calculated to be in the range of 15 to 20 kg N/ha/year. The recent analyses based on DEHM calculations in the background monitoring programme indicate that these estimates for some regions may have been somewhat too high. The DEHM provide values that over land are 20 to 25% lower than what was found with the ACDEP calculations. The DEHM calculations take into account the land use information and show values in the range 10 to 30 kg N/ha/year (Ellermann et al., 2006). However, even these estimates indicate that the atmospheric N loading is exceeding the critical loads at many places in the country. The emission reductions that will take place over the next 5 to 10 years are likely to reduce the atmospheric N deposition significantly.

A.9. How important are the Danish atmospheric N emissions for the atmospheric nitrogen deposition, and is the long-range transport contribution alone sufficiently high to exceed critical loads of Danish nature?

The ACDEP calculations performed in 2003 indicate a contribution from Danish sources of 16 and 39% of the N deposition to Danish water and land surfaces, respectively. However, these figures cover a wide range that depends strongly on the distance to especially Danish agricultural sources of ammonia. For fjords, creeks and bays the contribution from Danish sources can be up to 40%, and for the Kattegat Strait between Denmark and Sweden about 30% arises from Danish sources. For land surfaces in the western part of the country which has intense agricultural activity, the contribution is up to 50% of the N load – again deposition of locally emitted ammonia is the main reason for this high contribution from Danish sources. The ACDEP calculations for 2001 indicate that even without any local sources, critical loads will still be exceeded for Danish terrestrial ecosystems (Hertel et al., 2003a). However, the more recent DEHM calculations point at 20 to 25% lower atmospheric N depositions that are close to the critical loads for many of the sensitive terrestrial ecosystems. These results call for carrying out detailed analyses of in which parts of the country the atmospheric N deposition exceed critical loads of Danish nature. Such analyses would make it possible to identify the need for specific actions directed towards protection of specific nature areas in Denmark.

A.10. How large are the atmospheric nitrogen depositions to the Baltic Sea and the North Sea, respectively?

The ACDEP calculations showed that the total atmospheric N deposition in 1999 to the Baltic Sea is in the order of 318.000 tonnes N, which is equivalent to an average deposition in the order of 7 kg N/ha/year. Estimates from the early 1980ties have pointed at a deposition in the range of 9 to 12 kg N/ha/year. The EMEP model and the Swedish Match model have estimated N depositions just above 5 kg N/ha/year, which is relatively close to the ACDEP calculations.

The ACDEP calculations for the North sea area estimate an atmospheric N load on the order of 622,000 tonnes N in 1999. This is equivalent with about 9 kg N/ha/year. For the North Sea area even larger gradients in atmospheric N deposition are obtained compared with the Baltic Sea. Near the coast of the Netherlands the depositions are in the range of 20 kg N/ha/year whereas in the most northern part of the North Sea depositions have decrease to about 2 kg N/ha/year.

In addition to the environmental questions, the chapter addresses a number of more technical questions.

B.9. How well does the parameterised model describe the transport and deposition of atmospheric nitrogen compounds?

The results from the ACDEP model have been shown to reflect reasonably well the observed ambient air concentrations and depositions (Hertel et al., 1995; Skjøth et al., 2002; Hertel et al., 2002; Hertel et al., 2003b). However, the annual mean nitrate concentrations are in some cases overestimated by up to 50% (Frohn et al., 2007). Part of this overestimation is believed to be due to the lack of a seasonal variation in surface resistances in the dry deposition module. This lead to an underestimation of ozone concentrations, since dry deposition of ozone is overestimated during the winter season. An underestimation of ozone again leads to an underestimation of the conversion of nitrogen dioxide to nitric acid (and further to nitrate). Thereby the relation between local deposition and long-range transport is affected.

Comparisons with the DEHM model showed results that generally were in line with the ACDEP calculations. Later the dry deposition module of DEHM has been modified and the calculated depositions are in some cases more than 30% lower than what was preciously found. It is generally considered that DEHM reflects the atmospheric transport better than ACDEP, and this has been an important parameter in the decision to change model in the background monitoring programme. The implementation of a new dry deposition module in DEHM has given some additional changes to the results and in some cases reduced the estimated deposition even further. This change is found to be in good agreement with the measurements.

5.6 Fingerprints on science and environmental management

Through 10 years ACDEP was the applied tool under the Danish background monitoring programme for mapping atmospheric N deposition to Danish marine and terrestrial ecosystems. The ACDEP calculations were used in analysing the compliance with the goals in the Danish Aquatic Action plan (Andersen et al., 2006).

Within a couple of EU funded research projects, ACDEP has been applied for calculating atmospheric N deposition to the North Sea and the Baltic Sea. The results have been used to compare the atmospheric contribution with contributions from river run-off and water transport etc. These studies have contributed to a better understanding of the algae blooming and the role played by atmospheric N deposition.

The moderate calculation time make ACDEP suitable for carrying our scenario studies as it e.g. was the case in the EU funded MEAD project. This advantage has been reduced with the access to still more power full computers that makes the Eulerian models more competitive from this point of view.

6 Local scale nitrogen deposition

Local scale atmospheric N depositions are mainly of importance for the terrestrial ecosystems. The atmospheric loads of lakes and streams are generally dominated by the contributions from surface runoff, and this is also the case for the marine waters very close to the coastline (Jickells, 1998). However, for many of the terrestrial ecosystems, the atmospheric deposition is the only important input of N (Theobald et al., 2004). Close to intense agricultural activities, the atmospheric N deposition related to local NH_3 emissions may even exceed the background contribution (van der Salm et al., 1996). However, the relationship between the contribution from background and the local contribution depends of course on the emission but also on the distance to the local sources, the meteorological conditions and the local surface characteristics (Hertel et al., 2006b).

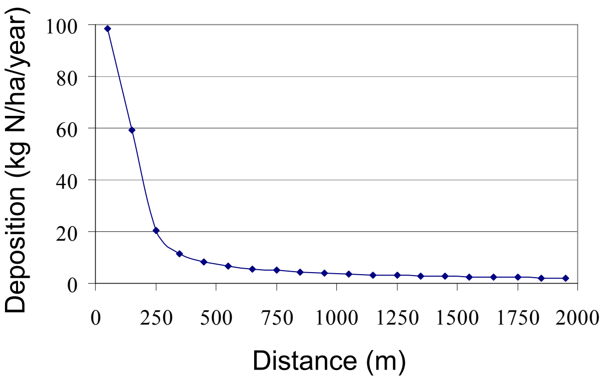


Figure 6.1 Ammonia deposition (kg N/ha/year) from a livestock farm with 100 livestock units[6] of cattle (68 mature cows and 69 calves), which yields an ammonia emission of 1114kg N (895 kg N from barn and 219 kg N from storage). Depositions are shown as function of the distance from the source. Calculations have been performed using NERI's local scale plume model OML-DEP under the Danis Ammonia Modelling System (DAMOS). Source: (Hertel et al., 2006a).

The annual NH_3 emission from barns and storages may be in the range of 20 - 40 kg N and up to even several thousands of kg N. Therefore, the average annual N deposition associated with a single source within a 2km radius is espected to contribute the in the range from 0.1 kg N/ha/year and up to even 60 to 100 kg N/ha/year very close to the source (see Figure 6.1). The deposition decrease with a steep gradient within the first 200 to 300 m from the source. In addition, the atmospheric depositions will depend on the frequency of various wind directions – in this context it is worth to note that the prevailing wind direction in Denmark is from south-west. Similarly to the contribution from emissions arising from barns and storages, there is a contribution from the emissions from fields mainly related to manure application and to a smaller extend also arising from evaporation of NH_3 from crops. The magnitude of the evaporation of NH_3 from crops is uncertain, and this has been debated in recent years. Concerning manure, it has been assumed that the farmer apply the maximum allowed amount to the field. The contribution from application of manure on the fields has been estimated to give a contribution to the atmospheric N depositions in the range of 2 to 4 kg N/ha/year as an average in a 2km zone around the fields. Like in the case of the barns and storages, there is a realtively steep gradient in atmospheric deposition going from the edge of the field in the down wind direction (Hertel et al., 2004).

Emissions of NH_3 from Danish livestock farms are strongly regulated compared with most other countries. Manure applications to the fields are restricted to take place during the growth seasons of crops and within certain limits for the total N load per hectare on annual basis. Farmers need to document access to fields for application of the manure. Finally the farmers need to apply to the local authorities when they intend to increase or otherwise change the animal production. This regulation has been shown to be reflected in current ambient air NH_3 concentrations in Denmark (Skjøth et al., 2008). Until January 1st 2007 these applications have been treated by the counties using an official Guideline for Environmental Impact Assessment (EIA) of NH_3 loads of the local nature (Bak, 2003). After this date the responsibility is moved to the municipalities and the guideline has been substituted by an internet based scheme with a more simplified procedure (see Section 6.3).

6.1 The Background Air Quality Monitoring Programme

Local scale calculations of atmospheric N deposition are included in the Background Air Quality Monitoring Programme in 2005 (Ellermann et al., 2005).

[6] One livestock unit is defined as the animals producing 100kg N in storage when using the stable with the lowest N loss (although for cows the average type stable has been used as reference). To give some examples: 1 livestock unit may e.g. be 1 dairy cow, 35 slaughter pigs, 167 chickens for egg products etc.

Table 6.1 Empirical critical loads for N deposition (kg N/ha/year) to natural and semi-natural groups of ecosystems. Only nature types relevant for Denmark have been selected (my selection). Source: (UNECE, 2004).

Ecosystem	Critical load (kg N/ha/year)	Indicators of exceedance
Coniferous forests	10 - 15	Increased nitrate leaching
Deciduous forests	10 - 15	Increased nitrate leaching
Wet heaths (upland moorland)	10 - 25	Transition heather to grassland
Dry heaths	10 - 20	Transition to grass; decline in lichens
Inland dune pioneer grasslands	10 - 20	Decrease in lichens, increase biomass
Inland dune siliceous grasslands	10 - 20	Decrease in lichens, increase biomass, increase succession
Molinia caerulea meadows	15 - 25	Increase in tall graminoids; decrease fiversity; decrease of bryophytes
Heath (juncus) meadows and humid (Nardus strica) swards	10 - 20	Increase in tall graminoids; decrease diversity; decrease of bryophytes
Raised and blanket bogs	5 - 10	Change in species composition; N saturation of Sphagnum
Poor fens	10 - 20	Increase sedges and vascular plants
Rich fens	15 - 25	Increased tall graminoids, decreased diversity, decrease of characteristic mosses
Shifting coastal dunes	10 – 20	Biomass increase, increase N leaching
Coastal stable dune grassland	10 – 20	Increase tall grasses, decrease prostrate plants, increased N leaching
Coastal dune heath	10 – 20	Increase plant production; increase N leaching, accelerated succession
Moist to wet dune slacks	10 – 25	Increased biomass tall graminoids
Pioneer and low-mid salt marches	30 - 40	Increased late-succession species, increase productivity

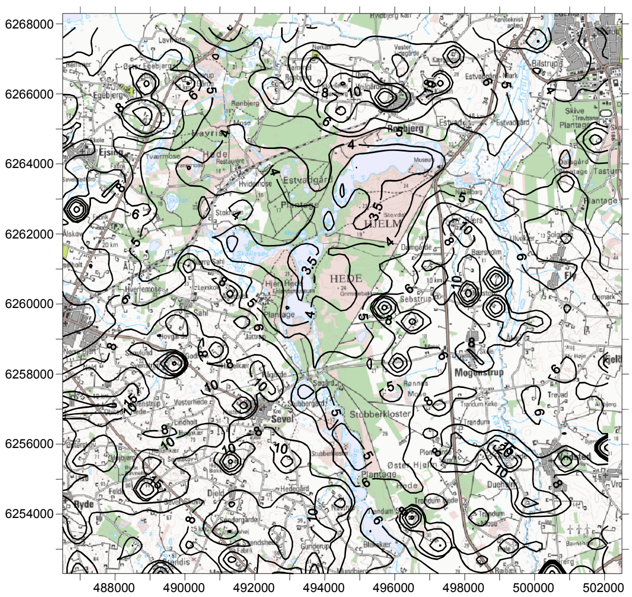


Figure 6.2 Atmospheric N loads (kg N/ha) from local NH_3 emissions to Hjelm Heath in Jutland in the western part of Denmark. Coordinates along the axis represent UTM-32N. Calculated with the OML-DEP model within the Danish Background Monitoring Programme (BOP). The background N deposition is about 11 kg N/ha for this area. Source: (Ellermann et al., 2006) but also depicted in (Hertel et al., 2007a) (Paper XII).

The calculations are now a routine element in the monitoring programme (Hertel et al., 2007a) (Paper XII), and the calculations are performed using the OML-DEP model under the DAMOS system (see section 4.3). Measurements of horizontal NH_3 concentration gradients are in the monitoring programme performed for shifting nature areas (new areas are selected every year). The calculations and measurements for the selected sampling sites within the areas are afterwards compared in order to evaluate the model performance. Finally are the DAMOS/OML-DEP calculations used for a mapping the local N depositions to the nature areas and determine the contribution from local and regional sources.

Figure 6.2 shows OML-DEP calculations performed in connection with the Danish background monitoring programme (BOP) (Ellermann et al., 2005) for a heath area in the western part of the country. It is evident that depositions in this area are arising from local livestock farming and may contribute 3 to 10 kg N/ha on annual basis. The impact on specific nature areas will be governed totally by the specific situation of livestock farms in the vicinity of the area. Compared with the DEHM calculations, The application of OML-DEP on to DEHM is seen to improve radically the agreement between model calculations and observations from the measurement stations in the monitoring programme (Ellermann et al., 2006) and also depicted in (Hertel et al., 2007a) (Paper XII). This is discussed in section 4.3, and illustrated in the comparisons shown in Figure 4.5.

6.2 The Buffer zone project

The Buffer zone project is initiated by the Danish Forest and Nature Agency in 2003 (Jensen et al., 2004a). The aim of this project is to evaluate the potential benefits of establishing buffer zones around Danish nature areas where NH_3 emissions from agricultural activities are kept to a minimum. The study include among other parts a survey on best available technologies (BAT), a review concerning NH_3 emissions, a review concerning knowledge about local scale dispersion and deposition (Hertel et al., 2004), and some economical calculations.

The DAMOS system is applied for calculating the potential reduction in atmospheric N deposition when manure application is avoided in buffer zones of 200 to 500m around sensitive ecosystems. The calculations are performed for different sizes of both nature areas as well as for buffer zones (Table 6.2). In Denmark, the average NH_3 emission from manure application to the field is estimated to be in the range of 10 kg N/ha/year in 2004. In the calculations it is assumed that the farmers apply the maximum allowed amount of manure to the field. The model calculations show that atmospheric N depositions to local nature areas would typically be reduced by 1 to 2 kg N/ha/year by establishing 200m buffer zones (Hertel et al., 2004).

Table 6.2 Reduction in NH_3 deposition (kg N/ha/year) when emissions related to application of manure is avoided in a buffer zone around the nature area. The emission from the nature area is set to 10 kg N/ha/year which is considered as an average value for Denmark in 2004, assuming that the maximum allowed manure is applied to all the fields. Source: (Hertel et al., 2004).

Area	300m buffer zone			500m buffer zone		
Diameter	max	mean	min	max	mean	Min
200	1.67	1.24	1.02	1.71	1.50	1.30
400	1.62	0.97	0.72	1.66	1.20	0.96
1000	1.43	0.68	0.43	1.62	0.87	0.61

A GIS based analysis of affected farms is determined from digital information about the placement of the Danish nature areas, and the location of the Danish farms. Three different scenarios are analysed in the project (see Table 6.3):

- Only EU habitat areas
- All Danish Bogs and oligotrophic lakes, heath land larger than 10ha, and dry grasslands larger than 2.5ha also included
- All designated nature conservation locations in Denmark are included

In the scenarios, it is taken into account that the shape of the buffer zone has an impact on the efficiency of the zone with respect to protecting the nature area. The reason for this is that the most frequent wind directions will also be the directions from which the nature area is most affected by upwind sources. Using the same area of the zone, but weighting the width of the zone by the wind direction frequency will therefore give a better protection of the nature area than a standard 250m buffer zone, even when the a fixed total area of the zone is applied.

Table 6.3 Area and number of farms affected of buffer zone. Source: (Schou et al., 2006).

	Scenario		
	1	2	3
Nature area affected	105,000ha	148,000ha	253,000ha
Affected farms[1]	9,507	16,857	39,218
Farm areas in zone	62,000ha	111,000ha	315,000ha

[1]In 2002 the total number of farms in Denmark is 71,913.

The NH_3 reductions are calculated using three types of abatement strategies: using acidification treatment (for farms >110 livestock units), manure injected into the soil (all farms) and for discontinuation of the livestock production (for farms <110 livestock units). The socio-economic costs of these emission reductions are calculated for the three different scenarios. The results showed a total cost of 10, 11 and 12 DKR/kg N/year (Schou et al., 2006), which may be compared with the measures applied in the Danish Ammonia Action Plan. With a total annual reduction in emissions of 8.2 million kg N, the average yearly cost of the Danish Ammonia Action is expected to be 9 DKR/kg N/year (FOI, 2001). Thus, it seems that the buffer zone regulation is competitive with the initiatives of the action plan. In addition the buffer zones aim at reducing the N deposition at specific locations, whereas the action plan aims at reductions on national level.

The economical calculations show that buffer zones are cost-efficient for reducing the atmospheric N deposition to local terrestrial ecosystems (Schou et al., 2006). However, until now buffer zone have not been implemented as a tool in Danish environmental management of sensitive terrestrial ecosystems, and there does not seem to be political will for such actions in a nearby future.

6.3 Validation of DAMOS/OML-DEP

Model calculations performed with the OML-DEP indicate that 20 to 25% of the emitted NH_3 deposit within the first 2km from the source (Hertel et al., 2004). The calculations with DAMOS and OML-DEP have been verified in different ways. One way has been to compare computed NH_3 concentration gradients down wind from livestock farms with similar measured gradients reported in literature. Such comparisons are performed within the buffer zone project (see section 6.2), and the results are briefly outlined in the following.

Calculations have been performed for a livestock farm with 250 livestock units (emission from stable 3.600 kg N/year and storage tank 1.300 kg N/year). The results show a contribution to the concentration in the close vicinity (within the first 50m) of the source of 35 to 95 µg N/m³ (depending on the wind direction), and 0.5 to 1.7 µg N/m³ in a distance of 270m from the source. These results are in good agreement with the experimental studies of (Fowler et al., 1998). Fowler et al. measured NH_3 concentrations around a Scottish poultry farm with an annual emission of 4.800 kg N/year. Down-wind from the farm in the most frequent wind direction, an annual mean NH_3 concentration of 63 µg N/m³ was measured at a distance of 15m from the source, whereas the concentration had decreased to about 2 µg N/m³ at a distance of 270m from the source. The OML-DEP calculations show up to 50 to 180 kg N/ha/year in the immediate surrounding of the source (the range indicate the differences between various wind directions), 13 to 80 kg N/ha/year in a distance of 50m, and 5 to 20 kg N/ha/year in a distance of 100m. In a distance of 200m form the farm, the load had decreased to 2 to 7 kg N/ha/year, and at a distance of 300m from the source the load was 1 to 3 kg N/ha/year. The OML-DEP calculation (as shown in Figure 6.1) is in good agreement with the observed gradient from the Scottish poultry farm.

Comparisons carried out as a part of the research programme under the Danish Aquatic Action plan have furthermore shown very good agreements between OML-DEP calculations and NH_3 measurements using passive samplers in two studies on a Danish poultry and a Danish pig farm, respectively (Løfstrøm and Andersen, 2007). In selected campaign periods, hourly mean NH_3 concentration gradients are measured down wind from the farms. These measurements are designed to characterise the ammonia plume down wind. In addition, long-term measurements are performed for one year using a sampling time of one to three weeks. The studies show in general a good agreement between measurements and model calculations (see Figure 6.3). However, the studies also showed that it is crucial that the emissions are determined with high detail.

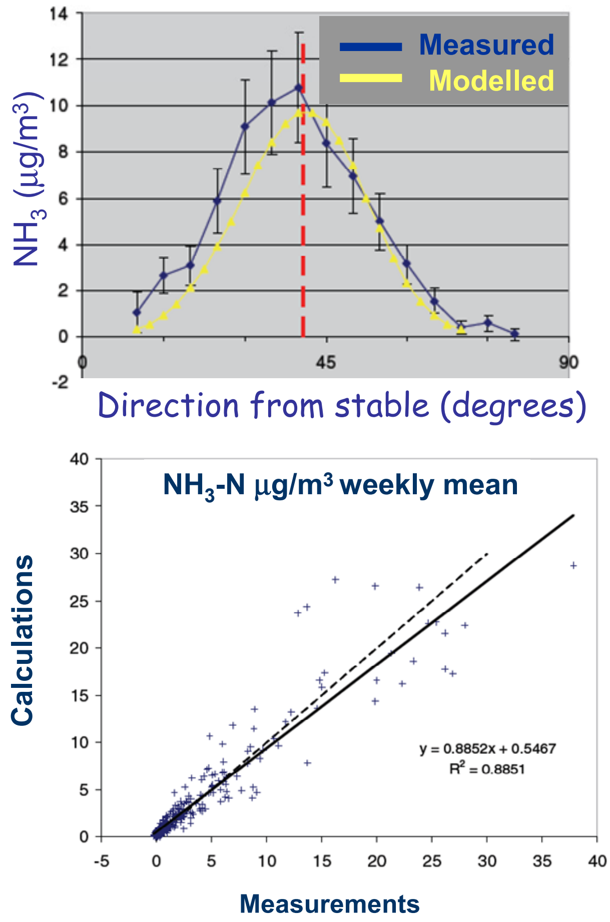


Figure 6.3 Two examples from the project under the Danish Aquatic Plan. The examples show comparisons of DAMOS/OML-DEP calculations with measurements. Upper plot shows the one-hourly measured and modelled NH_3 concentrations in a half-circle 200m down wind from the poultry farm. The stippled red line indicates the wind direction. For the measurements also uncertainty intervals are indicated in the figure. The lower plot shows comparisons for weekly mean NH_3 concentrations for all sampling points around the pig farm. These calculations are performed for an estimate emission from barn and using an assumed emission from a manure storage tank. Source: (Løfstrøm and Andersen, 2007).

The results indicate that long-term averages are well reproduced by the model. However, when the uncertainty in emissions and in measurements is taken into account, it cannot be fully concluded whether the OML-DEP provide systematic errors on the long-term averages. Short term measurements show that the horizontal dispersion in the model is too small for the conditions present during the experiments (daytime and in a distance of 200m from the source), whereas the model overestimates the peak concentrations.

Table 6.2 Examples of atmospheric N deposition mapping to 26 selected sensitive terrestrial ecosystems in Eastern Jutland, Denmark. Calculations performed using the DAMOS system. Sources: (Frohn et al., 2008)

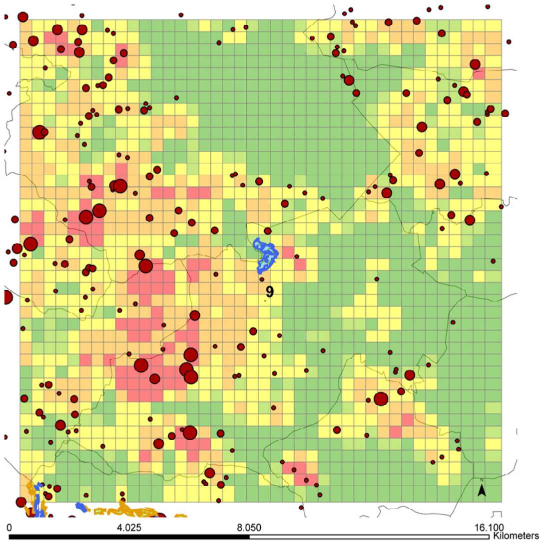
Site	**Nature type**	**Calculated load (kg N/ha/year)**	**NH_3 deposition (kg N/ha/year)**	**Critical load for the type of nature in the area (kg N/ha/year)**
Skals Ådal	Rich Fens	14	4 (2 local sources)	5 - 10
Tuemosen	Raised Bog	11	2 (1 local sources)	5 - 10
Overdrev ved Fussing sø	Meadow	12	3 (2 local sources)	10 - 20
Nipgård Sø	Lake	10	3 (2 local sources)	5 - 10
Sømose i Løvenholm Skov	Raised Bog	10	1 (0.5 local sources)	5 – 10
Færgemosen	Bog	11	2 (~2 local sources)	10 – 15/10 – 20
Rødesø	Lake	8	1(~0.5 local sources)	5 - 10
Hængesæk ved Påruplund	Meadow	10	1(~0.5 local sources)	5 - 10
Bavnhede	Heath	13	3 (2 local sources)	10 - 20
Grane Langsø	Lake	9	2(~0.5 local sources)	5 - 10
Ved Røverstuen	Bog	12	3 (~2 local sources)	5 - 10
Spidsbjerg/Madbjerg	Heath	12	2 (~1 local sources)	10 - 20
Ejer Skov	Deciduous forest	25	6 (~4 local sources)	10 - 20
Uldrup Bakker	Deciduous forest	22	6 (~3 local sources)	10 - 20
Søby Fredskov	Deciduous forest	25	8 (~6 local sources)	10 - 20
Jenskær	Deciduous forest	18	3 (1 local sources)	10 - 20
Gudenåens kilder	Bog	12	3 (2 local sources)	5 - 10
Lyseskov	Deciduous forest	21	3 (2 local sources)	10 - 20
Enslev Bjerge	Bog	13	3 (2 local sources)	5 - 10
Dyrby Krat	Deciduous forest	25	8 (~6 local sources)	10 - 15
Stabelhøje	Rich Fens	13	3 (~2 local sources)	10 - 20
Stenholt Mose	Bog	11	2 (~1 local sources)	5 - 10
Bjerre Skov	Deciduous forest	25	10 (~8 local sources)	10 - 20
Ringelmose Skov	Deciduous forest	23	5 (~3 local sources)	10 - 20
Tåstrup Mose	Bog	12	3 (~2 local sources)	10 - 15
Bygholm Ådal	Rich Fens	15	5 (~4 local sources)	10 - 15

6.4 The Frederiksborg county project

In 2006 the Frederiksborg county project is carried out (Geels et al., 2006b). The mapping of atmospheric N depositions performed in the Background Air Quality Monitoring Programme (BOP) has shown that Frederiksborg County is an area with relatively low atmospheric N depositions. This opens for the possibility that regulation of local NH_3 emissions may reduce the loads of some of the ecosystems in the county down below critical loads. The basic idea is therefore to map the local and the regional atmospheric N depositions to selected sensitive terrestrial ecosystems in the county, and compare these loads to the critical loads of the specific ecosystems. The calculations are performed using the DAMOS system for 15 ecosystems that have been selected by the county as highly valuable nature areas.

An example of a mapped nature area – the Børstingerød moor – is shown in Figure 6.4. This area is dominated by the nature-types water-meadow and marsh. According to a working group under UN-ECE, such nature types have critical loads on the order of 15 to 25 kg N/ha/year (see Table 6.1). According to the DAMOS calculations, the total atmospheric N deposition for 2004 is in the order of 11 to 13 kg N/ha/year. The calculations this indicate that the critical loads for this nature system are not exceeded in 2004.

The mapping performed with the DAMOS system for 2004 shows that the total atmospheric load of the nature areas in the county is in the range between 11 and 20 kg N/ha/year. The international and regional contribution from non-local sources is calculated to be in the order of 11 to 12 kg N/ha/year, whereas the local contribution is found to be between 1 and 8 kg N/ha/year. The lower boundary for the critical loads is exceeded for 11 out of the 15 nature areas. The results from this work calls for a strategy aimed at preserving the biodiversity in these areas in the future, since several of the nature areas may be protected with actions that in some case concern just moderate reductions in the atmospheric N loads.



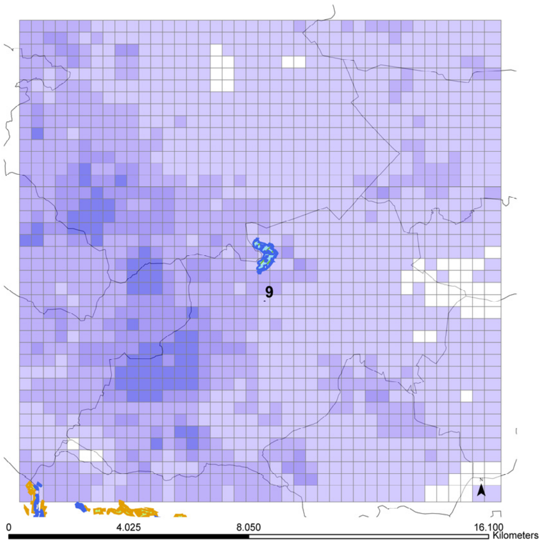


Figure 6.4 of atmospheric N deposition to Børstingerød moor for the year 2004. Upper plot: the ammonia emission inventory for the area surrounding the moor. Lower plot: the computed total atmospheric N deposition to the area computed with the DAMOS system. Source: (Geels et al., 2006b).

6.5 Surveys for the Environment Centres in Jutland and on Sealand

The interest from the policy makers in having detailed mapping of local N deposition to sensitive terrestrial ecosystems is quite substantial. In 2008 two different surveys were carried out by NERI for the newly established Environment Centres in Jutland (Århus) and on Sealand (Roskilde), respectively. The aim is here to compare the atmospheric loads obtained from DAMOS calculations with the critical load values for the type of nature. These two surveys give quite different results as they concerned terrestrial ecosystems in a high and low NH_3 emission area, respectively. Again DAMOS/OML-DEP is applied for the calculations of local N deposition to the nature areas. In the high NH_3 emission area in Jutland, 26 nature areas are investigated. Out of these 16 are found to have atmospheric N loads exceeding the upper limit value for the critical load, 9 ecosystems are exceeding the lower limit value for the critical load and only one area does not have atmospheric N loads that are exceeding the critical load. For the low NH_3 emission area, 17 nature areas are investigated. Only two exceedances of the upper limit value and four exceedances of the lower limit value for the critical load are found in this case. For the remaining 11 nature areas the critical loads are not exceeded.

6.6 Regulation of ammonia from Danish livestock farms

The Danish structural reform has lead to a series of substantial changes of the local authorities in Denmark. The structural reform is in action since January 1st 2007. With the structural reform the Danish counties are closed down. Thereby the responsibility for regulating Danish livestock farms has moved from the counties to the municipalities. At the same time a number of municipalities merged to form larger municipalities. With the new responsibility for the regulation of livestock farming, the Danish municipalities thereby have to make decision concerning applications for modifying and increasing livestock production.

By January 15st 2007 the new official procedure is implemented for impact assessment of NH_3 emission from Danish livestock production. This procedure is a highly simplified method compared with the previous method. The new procedure considers only the additional N deposition to nearby nature areas, which is directly associated with the implemented change in food production. The decision is made depending on whether this additional N deposition exceeds a limit which is defined as a function of the presence of other livestock farms also affecting the nature area in question. The Forest and Nature Agency has thus determined that the additional N deposition may not exceed:

- 0.3 kg N/ha/year in the case of more than two livestock farms affecting the nature area
- 0.5 kg N/ha/year in the case of just two livestock farms affecting the nature area
- 0.7 kg N/ha/year in the case of only one livestock farm affecting the nature area

This procedure makes the assessment easy to perform, but it also introduces some unknowns concerning the level of protection provided for the nature area. It would be safer for nature with a system that is still based on an assessment of the total N load to the nature area and a subsequent comparison with critical loads determined for the specific nature area.

In 2005 the Danish Forest and Nature Agency initiate a project to revise the procedures for the environmental impact assessment of NH_3 loads of the local nature in connection with approval of changes in animal production (Geels et al., 2006a). In this project we suggest an easy to apply three step procedure (Geels et al., 2006a; Hertel et al., 2006a) based on DAMOS/OML-DEP calculations. I will here shortly outline the suggested procedure.

The suggested Guideline for NH_3 from animal production includes three steps with increasing complexity (Figure 6.5):

- Step A: Simple screening for quick assessment of potential environmental impact as a result of airborne ammonia emitted from smaller livestock farms. This method is to be applied for smaller livestock farms (<75 livestock units) and is solely intended for a first crude screening of farms with insignificant impact on the local nature. The screening is not carried out for cases when manure is brought to a biogas plant.
- Step B: Standard method for assessment of environmental impact based on nomograms and tables. This method is intended to be used for all cases that cannot be closed after step A, except for situations when the applicant or others ask for more detailed treatment after step C.
- Step C: Detailed model calculations for mapping of N loads and similarly detailed critical load estimates for the local nature in the nearby region of the livestock farm. This method is intended to be used only when the applicant or others may wish so on basis of predefined cases where such a possibility should be open.

The simple screening is intended to be used for quickly excluding farms for which no significant impact on nature is foreseen. The second step consists of nomograms and tables for determining the deposition at various distances from the livestock farm. These nomograms and tables are intended to be established from analyses of DAMOS/OML-DEP calculations.

The intension is here to use these nomograms (based on curves in line with the one shown in Figure 6.1) and tables to update the spreadsheet developed previously by the Danish counties for performing calculations according to the Guideline of 2003 (Bak, 2003). The third step is a full calculation performed with the DAMOS system.

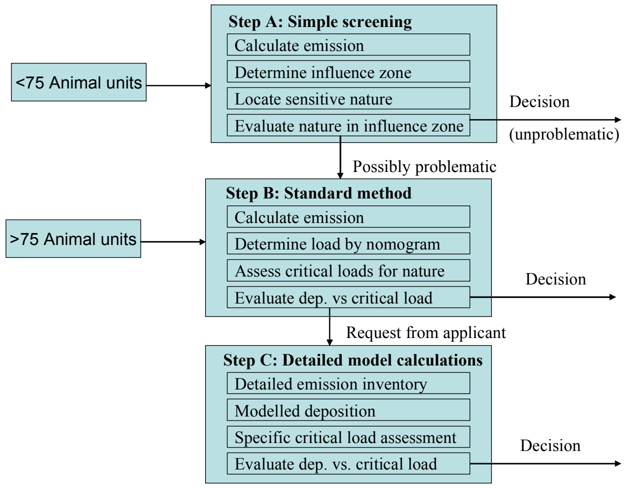


Figure 6.5 Sketch to illustrate the overall calculation procedure in the suggested new Guideline for assessment of environmental impact of ammonia emissions from livestock production in Denmark (Geels et al., 2006a).

In connection with the development of the suggestion for a calculation procedure, a sensitivity analysis of the OML-DEP calculations is performed. The aim is to determine which of the governing parameters that are the most critical and therefore need to be accounted for in the calculation procedure under Step B.

Not surprisingly, the surface resistance turns out to be a critical parameter. The meteorological conditions are naturally also crucial, especially the frequency distribution functions for wind speed and wind direction are central in this context. The final curves for the calculation procedure under Step B are therefore generated using calculations for a 5 years time period in order to account for the most common meteorological conditions.

It is my personal view that the above suggested procedure would provide a higher level of protection for Danish nature areas compared to the system implemented January 15th 2007.

6.7 Conclusions

The present chapter addresses the issue of determining the local atmospheric N deposition to Danish terrestrial ecosystems. The chapter addresses the following environmental questions:

A.13. Is it possible in a Danish region with moderate loads through regulation of local sources to reduce the atmospheric nitrogen loads below critical loads?

A survey was carried out for Frederiksborg County in northern part of Sealand. Calculations are performed with the DAMOS system for 13 ecosystems selected by the county as highly valuable nature areas. The results show that the lower boundary for the critical loads is exceeded for 11 out of the 15 nature areas. The results also indicate that with moderate regulation of the local loads, some of the nature areas would be below the lower boundary for the critical load. The results from this work calls for a strategy aimed at preserving the biodiversity in these areas in the future, since the atmospheric N deposition is close to the critical loads, and a specific regulation aimed at these areas may bring the atmospheric N depositions down below the critical loads. With the general decrease in atmospheric N loads, this will apply to other regions in Denmark.

In 2008 two new surveys were carried out for the new Environment Centres in Jutland (Århus) and on Sealand (Roskilde). The aim was to map N deposition to selected nature areas and compare these with critical load limit values. These surveys gave quite different results as they concerned terrestrial ecosystems in a high and low NH_3 emission area, respectively. Again DAMOS/OML-DEP was applied for the calculations. In the high NH_3 emission area, 26 nature areas were investigated. Out of these 16 had atmospheric N loads exceeding the upper limit value for the critical load, 9 exceeded the lower limit value for the critical load and only one area did not have loads exceeding the critical load. For the low NH_3 emission area 17 nature areas were investigated. Only two exceedances of the upper limit value and four exceedances of the lower limit value for the critical load were found. For the remaining 11 nature areas the critical loads were not exceeded.

A.14. Are buffer zones with restricted ammonia emissions around local nature areas an efficient way to regulate the atmospheric nitrogen load?

The buffer zone study show that atmospheric N depositions of to Danish nature areas would typically be reduced by 1 to 2 kg N/ha/year by establishing 200m buffer zones around sensitive Danish nature areas. The economical calculations revealed that establishing buffer zones is a cost-efficient way to reduce the atmospheric N deposition to local terrestrial ecosystems in comparison with the actitivities in the Danish action plans.

Currently there does not seem to be the polical will to apply this type of regulation, although it seems to be a cost efficient way of obtaining local load reductions of sensitive ecosystems.

The chapter in addition has addressed one technical question.

B.10. What is needed in order to develop an easy to apply assessment system for use in regulation of ammonia emissions from livestock farms?

An easy to apply assessment system was outlined and suggested for implementation. The three steps in the assessment system included a simple screening, a nomogram method based on standard calculations performed with OML-DEP, and a full DAMOS calculation. The procedure that at the end was implemented contained only step two and without considering the total load of the ecosystems and how they related to critical loads. There is a need for scenario studies in order to investigate to what extend this procedure protect the most sensitive ecosystems in the country.

6.8 Fingerprints on science and environmental management

The DAMOS system is now an integrated part of the Danish Background monitoring programme (BOP). The combination of the long-range transport model and the local scale model represent state-of-the-art (Hertel et al., 2006b), and has been shown to improve the agreement with observed NH_3 concentrations substantially. The DAMOS/OML-DEP calculations has been used as the basis for the revised procedure for assessment of N deposition from Danish livestock farms in connection with the handling of the farmers applications for modifying/increasing livestock production. The DAMOS has been used for impact assessment of implementing buffer zones around sensitive Danish nature areas. DAMOS has also been applied for mapping atmospheric N deposition to nature areas in region with moderate loads. This has brought new insight into possibilities for protecting sensitive nature areas.

7 Discussion and Conclusions

7.1 Measurements and models

Integrated Monitoring and Assessment (IMA) of air pollution is today well established at NERI-ATMI. The applied procedures and the implemented methodologies have been the result of work than has been carried out over the past 20 years. In the implementation of IMA at NERI-ATMI, *the measurements* are used for determining:

- Actual ambient air concentrations and/or depositions of pollutants at the monitoring sites.
- Seasonal variation in pollution load.
- Long-term trends in ambient air concentrations and/or depositions of pollutants.
- Source apportionments.
- Validation and parameterisation of air quality models.

The air-quality measurements are irreplaceable for studying the actual levels and trends in pollution load. This type of information cannot be obtained from model calculations since the calculations rely on the validity of the input data and the applied parameterisations. The validity of actual input data as well as the validity of the applied parameterisations may change over time. In the IMA at NERI-ATMI, *the model calculations* are used for obtaining:

- Improved geographical distribution in the mapping of the pollution load.
- Distribution between contributions from local and regional sources.
- Distribution between contributions from different source sectors.
- Scenarios and prognoses, the impact of new or modified sources, and the impact various reduction strategies etc.

The results from the model calculations are generally used for providing information about concentrations and/or depositions at locations where measurements are not performed, as well as information about the contributions from different sources and source regions. Finally they are used for scenario studies e.g. to evaluate the impact of implemented and/or planned emission reduction strategies etc. The implementation of air quality models in the monitoring activities is in compliance with EU directives on air quality monitoring and assessment. The EU directives are thus open for the application of models as supplement in assessment of the air quality, e.g. where measurements are not available. The EMEP programme (European Monitoring and Evaluation Programme) has furthermore performed a slightly different form of IMA by linking closely the measuring and modelling activities within the programme. In the case of the EMEP programme this is obtained through cooperation between modellers mainly based in Norway and various European groups performing the measurements. However, in the case of NERI-ATMI, the people performing measurements and model calculations are based at the same institute. The development and implementation of IMA at NERI-ATMI have added significant value to the air pollution studies and e.g. made it possible to perform:

- Mapping of air pollution levels in urban streets where measurements are not performed.
- Impact analysis of environmental zones around central parts of the urban areas.
- Analysis of the impact of particle filters on nitrogen dioxide pollution.
- Assessment of human exposure to air pollution for large epidemiological cohorts.
- Linking of air pollution exposure data to data for various health outcomes.
- Annual mapping of nitrogen and sulphur depositions to Danish marine and terrestrial areas.
- Assessment of nitrogen deposition from single farms.
- Source apportionment for the urban as well as the regional pollution loads.

The IMA has been gradually developed over a long period of time and must be considered as a substantial success at NERI-ATMI and it is continuously being refined and updated. The advantages include an improved quality of the air pollution monitoring and assessment studies, and a better understanding of the governing processes for the pollution loads in Denmark. The use of IMA means also a more optimal use of the resources, since more information is derived and the interpretation of the measurements is improved.

In order to be applied in IMA there are certain demands for the documentation and validation of the model calculations. These demands are in many ways similarly to what concerns sampling, handling and analysis of measurements, and they include:

- Documentation.
- Validation.
- Reproducibility.
- Quality assurance.

The applied models and model parameterisations at NERI-ATMI are documented mainly in journal articles and conference proceedings, but also to some extent in technical reports. The validations of the models concern testing the:

- Internal logic of the model.
- The applied numerical methods.
- Comparisons with observations.

It is crucial that the various procedures and parameterisations have been put together in a logical and operational way. The applied numerical methods have to be tested and validated to ensure that the numerical errors is kept sufficiently small and thereby are not affecting the results in a substantial way. Finally the model calculations have to be compared with measurement to ensure that the applied parameterisations are working in a satisfactory way and that the integrated model reproduces well the observed levels and trends. In the cases when the model results cannot reproduce the observed trends, one has to be very careful when the model is applied for prognostic purposes.

It is crucial that the performed model calculations may be reproduced. It is therefore important to document the version of the model and the set of input data that has been applied for the specific calculations.

The handling of obtained results may in some cases lead to errors, and it is therefore crucial that the results are quality checked. In the monitoring programmes this may done by comparing the computed levels and loads to measurements or to calculations performed for previous years. Another way is to perform various types of mass balance checks.

7.2 Institutional requirements

In this thesis I believe to have documented that IMA is a very strong tool in air pollution management. The provided examples have demonstrated that model calculations are indispensable for use in interpretation of, and as an extension to measurements in field experiments and routine monitoring programmes. The shown examples demonstrate clearly that model calculations may in general supplement and extent the information that can be derived from measurements in field experiments and routine monitoring programmes. The work also heavily underlines the need for a very close cooperation between the people that conduct the measurement activities and the modellers. This cooperation is important already in the design of field experiments and monitoring programmes in order to ensure that the necessary input is available for the subsequent application of models. The two Danish Air Quality Monitoring programmes for the urban and the rural environments are strong examples of Integrated Monitoring where model calculations are performed on routine basis to supplement measurements.

NERI-ATMI has in 2007 implemented a new structure that aims to enhance the integration further between modellers and measurement experts. This new development was made possible by the work demonstrated in this thesis, among others.

8 Perspectives

The unique strength in the atmospheric research at NERI over the past 20 years has been the strong interaction between measurements and modelling activities. One of the goals of developing the coupled air pollution forecasting system THOR was to sell data produced by the system as well as to export the entire model system to other countries. Selling the coupled model system has turned out to be highly difficult. The THOR system is based on air quality models at a very high level, but the user interface and the graphical presentations could be more advanced. In one occasion an institute with considerably less advanced models but a very professional interface was selected instead of the THOR system. Furthermore it is evident that the various institutes working in air pollution research and air pollution management prefer to develop air quality models of their own or at least to have access to source code for understanding in depth the process description and possibly further developing or modifying these models.

It is now generally accepted that the climate of the Earth is changing, and that these changes may be even more pronounced in the coming years. The changes in the climate will affect the transport as well as the chemical conversion of atmospheric air pollutants. The fossil fuels represent a limited resource, and various alternatives have been suggested. Hydrogen has been suggested as an alternative, but also bio-fuels are strongly debated. Changes in fuel types will affect among others the pollutant emissions and thereby also affect the air pollution levels. Changes in the precipitation amounts will affect the deposition of atmospheric N compounds. Increased humidity will affect the atmospheric radical chemistry and thereby the atmospheric fate of pollutants.

The impact of air pollution on health and environment will be affected by the changes in climate as well as in energy sources. NERI's suite of air quality models provide the basis for scenario studies of the impact of these changes on the two main topics of this thesis - human exposure to air pollution and atmospheric N loads of terrestrial and marine ecosystems. This is of course only possible in case there is access to climatologic data from a General Circulation Model (GCM), which has been obtained in previous cooperation with the Danish Meteorological Institute.

Integrated air quality monitoring and assessment is well developed at NERI. Many of the field experiments are designed for being used in the validation of the models or for improving process understanding. The monitoring programmes have been optimised by including model calculations as an integrated element in the programmes. Still there are various ways that the integration of measurement activities and model calculations may be extended and improved in the future.

Model calculations may be used as a guideline in the quality control of measurements. Air pollution forecasts are already made on routine basis within the THOR system at NERI. Automatic graphical procedures could be established for displaying measurements and model calculations together for the monitoring sites. Such a procedure would make it easy to identify periods with large discrepancies. These periods might either be the result of errors in the measurements or periods where the model for some reason fails to describe the governing atmospheric processes.

Air mass trajectories may be strong tools in the interpretation of measurements. Automatic procedures could be established to produce plots with air mass trajectories to the monitoring sites on routine basis. These trajectory plots might then be displayed in part of the display when time series of measurements and model calculations are viewed in connection with the quality control.

8.1 Traffic pollution

The OSPM is integrated as a routine tool in the Urban Air Quality Monitoring Programme. The OSPM is here used on routine basis for mapping traffic pollution in streets where measurements are not carried out. However, the model is also applied in various emission reduction studies and forecast of the development in traffic pollution. The OSPM is a strong tool in modelling traffic pollution in urban streets. The validation studies show that the model works well for street canyons, but until now very few validation studies have been carried out for other street types. There is thus a great need for experimental data designed for testing the OSPM model performance for other street types.

The model performs well when tested for particle number concentrations as it has been done for streets in Copenhagen. However, only a small part of the emission of particle mass is related to vehicle exhaust. More important is the resuspension of dust, wear of breaks, wheels and road material. These processes are difficult to parameterise. The OSPM is currently only able to reproduce parts of the particle mass and more studies in this area are still needed.

8.2 Human exposure

There are still many unknowns concerning health effects of air pollution. A number of studies have shown an association between distance to trafficked road and various health outcomes. However, indoor pollutant levels seem not to be related to the same outcomes. Outdoor air pollution may therefore be more important than indoor air pollution; or short term air pollution exposure may be more important than long term exposure. Here we are still facing a number of unresolved questions regarding exposure and health outcome.

The AirGIS system is shown to be a strong modelling tool in exposure assessment of traffic air pollution. However, there are still a number of features that would be very useful to have included in the system. This counts for diffusive point sources that in some case may have significant impact on the local air pollution levels. Another issue relates to the air pollution from wood stoves. Particle pollution levels in area with intensive use of wood stoves, the particle pollution levels may be similar to what is observed in the most trafficked streets in the larger Danish cities (Glasius et al., 2006; Glasius et al., 2008).

In the Netherlands significant effort has been put into mapping the time-activity pattern of the population. No similar studies have been carried out in Denmark, and in fact the demographic data concerning the Danish population is rather scarce with respect to time-activity pattern. Such data may be generated for smaller cohorts in epidemiological studies using diaries and e.g. GPS mapping of routes. However, for mappings of the general air pollution exposure of the Danish population, it would be highly beneficial with a large scale study of the Danish population e.g. carried out in cooperation with socio-researchers.

Allergy is a growing problem in the industrialised countries including Denmark. Some studies indicate that pollen exposed to air pollution may be more potent allergens compared with "clean" pollen. In order to study such phenomena, it would be obvious to use AirGIS in combination with the dynamical pollen model that is currently under development at NERI. Currently a joint PhD position between University of Worcester and NERI is vacant for studying these phenomena.

8.3 N deposition from long-range-transport

The gap between long-range transport modelling and local scale modelling of air pollution is diminishing these years. Previously, background pollution levels where modelled at a spatial resolution of 150km x 150km. Then the models moved to a 50km x 50km resolution, and now the DEHM model has a 16.67km x 16.67km resolution for the inner nest (2-way nesting is performed). Computer time is still an obstacle in this context, since various scenarios need to be run at the same resolution. There is no doubt that the next step is to go down to about 5km x 5km. An alternative to full 2-way nesting is to combine the DEHM model runs with another model with an even higher resolution for the Danish area. This could be a Lagrangian type model like ACDEP, or it could be Eulerian. A PhD position to study this is currently vacant at NERI within the Research Centre CEEH.

In long-range transport-chemistry modelling, the so-called aerosol mass gap is currently one of the largest challenges. Current models only reproduce about 60% to 70% of the ambient air particle mass, and the remaining 30% to 40% is not fully understood and accounted for in the transport-chemistry models. This discrepancy is often termed the aerosol mass gap. It is believed that some of this mass consists of water and volatile organic compounds and some of this is likely to be related to particles generated from biomass burning. There is a strong need for studies that explore this area, since a proper modelling of $PM_{2.5}$ and PM_{10} is totally depending on a better understanding of these processes. Another question is to what extent mass is the right way to study particles in relation to health effects. It is likely that the health effects depend on the chemical composition of the particles. Many ongoing studies focus on chemical characterisation of particles in ambient air. In a few years from now our knowledge in this area is likely to have expanded significantly.

Measurements of concentration gradients over sea are generally difficult to establish. The onboard ferry long-term measurements conducted under the ANICE project is a unique example of how such gradients may be obtained. There is a lot of potential in conducting more experiments of this kind and thereby improve current estimates of N depositions to marine waters.

8.4 Local N deposition

Development of coupled model systems like the DAMOS system is a tremendous step forward in the mapping of local atmospheric N deposition to especially terrestrial ecosystems. It has been a general tendency that more nature areas with exceeded critical loads have been reported for increasing the geographic resolution in the models. Currently we are moving towards access to even more detailed input data for these model calculations. The geographic and especially the temporal resolution in emission data will be improved considerably in the coming years. In this context there is still a great need for collecting detailed information about agricultural praxis in the various countries in order to extend the detailed ammonia emission model to the rest of Europe – since the model currently only apply for Denmark and nearby surrounding areas. Various activities are ongoing to make this extension of the emission module to cover the entire European area.

Experimental data has shown that fluxes to nature may be bi-directional. The transport of e.g. NH_3 may be in as well as out of the plant depending on the concentration of NH_4^+ in the plant and the NH_3 in the ambient air. Specific dry deposition sub-models for surface resistance that include a description of the bi-directional flux. Until now these sub-models have mainly be applied in model studies that are highly adapted to the experiment. There is thus still a great need for further developing these sub-models, generate the necessary input data for e.g. European scale and test them intensively in operational transport-chemistry models. For the later there is furthermore a great need for more experimental data in order to further improve our understanding of these processes and for use in model validation studies.

9 Glossary

As a help to the reader this glossary has been split into four sections: 1) Methods, models & systems, 2) Centre, programmes & projects, 3) Organisations & institutions, and 4) Other abbreviations.

9.1 Methods, models and systems

1DIM	One-dimensional chemistry-transport model - developed at NERI for describing chemical transformation of biogenic sulphur compound emitted from the sea. It was developed at NERI-ATMI/University of Bergen in the early 1990ties.
ACDEP	Atmospheric Chemistry and Deposition model – Lagrangian chemistry-transport model developed at NERI-ATMI in the mid 1990ties for use in the Danish Marine Research Programme Sea90. For many years it was applied in the Danish Background Monitoring Programme under NOVA/NOVANA.
AEOLIUS	British monograph method for estimating street pollution levels. This monograph method has been developed on basis of OSPM results.
AirGIS	A GIS based system for generating input data to the UBM and OSPM models e.g. in connection with exposure assessments for epidemiological studies. It was developed at NERI-ATMI in the late 1990ties.
CAR	The Dutch CAR is an empirical model for calculating air pollution levels in multiple types of urban streets. The CAR model has been developed from statistical analysis of measurements in Dutch urban streets.
CBM	Carbon-Bond Mechanism is a lumped chemical mechanism where hydrocarbons are represented partly by groups in order to reduce the chemical scheme.
COPERT	COmputer Program to calculate Emissions from Road Traffic – the most recent version is COPERT IV. The model is developed by European researchers. COPERT is build into WINOSPM.
DAMOS	Danish Ammonia Modelling System consists of OML-DEP for local scale dispersion and deposition and DEHM (originally ACDEP) for the long-range transport component. It was developed at NERI-ATMI for assessment of NH_4 deposition to sensitive terrestrial ecosystems.
DEM	Danish Eulerian Model was developed at NERI-ATMI in the 1980ties.
DEHM	Danish Eulerian Hemispheric Model – 2-way nested grid Eulerian chemistry-transport model describing the entire Northern hemispheric; developed at NERI-ATMI and now applied in variety of activities in the department.
DEHM-REGINA	Version of the DEHM that contains 2-ways nesting.
EBI	Eulerian Backward Iterative method is a numerical method developed at NERI in the early 1990ties for solving chemistry in atmospheric transport-chemistry models.
EVA	Environmental Valuation system developed at NERI in cooperation between ATMI and SYS for valuation of the monetary costs for the society associated with air pollution.
HIWAY-2	US Air Pollution Model for Road Traffic on Highways.
LUR	Land-use Regression is an empirically derived modelling technique using GIS and spatial data.
MM5	MM5 is a flexible high resolution American weather forecasting model. It is applied in the THOR system for providing meteorological data to the air quality models.
NBB	Swedish abbreviation for the Nordic calculation method for vehicular traffic (In Swedish: Den Nordiske Berekningsmethod for Bilavgasser). It was developed on basis of the US STREET model. In the early 1990ties it was modified on basis of OSPM results in a cooperation between a number of Nordic Research institutes including NERI-ATMI.
OML	Danish abbreviation for Operational Meteorological Air Quality Models (In Danish: Operationelle Meteorologiske Luftkvalitetsmodeller). A suite of Gaussian plumes models developed at NERI-ATMI over the years since the late 1970ties.

OML-Multi	Multi source version of OML which handles both point sources and area sources.
OML-DEP	More recent version of the OML-Multi developed for describing deposition of air pollutants on local scale.
OSPM	Operational Street Pollution Model was developed at NERI-ATMI in the late 1980ties for calculating pollution levels in urban streets. It is now applied in a variety of countries mainly in the windows based version (see WINOSPM)
SIMAIR	Model system for mapping air pollution from road traffic, and developed by SMHI for use by the Swedish municipalities. The model system contains OSPM.
THOR	the air pollution forecasting and scenario system developed at NERI-ATMI which is consisting of a suite of air pollution models on various scales from hemispheric scale down to single streets in urban areas. Currently it includes the MM5 weather forecasting model, DEHM, UBM and OSPM.
UBM	Urban Background Model is a highly parameterised model developed at NERI-ATMI in the early 1990ties and still applied for describing air pollution levels in urban background.
VLUFT	Norwegian abbreviation for Road Air (In Norwegian: Veiluft) which is a model system based on HIWAY-2 and NBB. The model system contains OSPM for calculating pollution levels in urban streets.
WINOSPM	Windows based and most recent version of OSPM that may be downloaded from NERIs homepage in a trial copy. This version is now applied in a variety of mainly European countries but also other parts of the world.

9.2 Centres, programs and projects

AIRPOLIFE	AIR POllution in a LIFEtime perspective a Research Centre of Excellence funded by the Danish Research Council 2004-2008. NERI-ATMI was one of the central partners.
ANICE	Atmospheric Nitrogen Input to the Coastal Ecosystem – EU funded marine research programme 1997-2000. NERI-ATMI was a central partner.
ASEPS	The Air-Sea Exchange Process Studies Funded by US ONR 1993-1996. and lead by NERI-ATMI.
ATMI	Danish abbreviation for the Department of Atmospheric Environment (In Danish: Afdeling for Atmosfærisk Miljø) at NERI.
Auto Oil II	It is an assessment programme funded by the European Commission to assess future trends in emissions and air quality. OSPM was applied in some of the assessments.
BOP	A Danish abbreviation for The Danish Background Monitoring Programme (In Danish Baggrunds-overvågningprogrammet).
CEMIK	Danish abbreviation for Centre for Environment Associated Cancer (In Danish: Center for Miljørelateret Kræft) 2000-2004. NERI-ATMI was a central partner.
CML	Danish abbreviation for Centre for Environment and the Respiratory System (In Danish: Center for Miljø og Luftveje) funded by the SMP programme 1998-2002. NERI-ATMI was a central partner.
Dobris	A series of assessments of the European Environment carried out by the EEA.
EXPLUS	Danish abbreviation for Exposure to particles and airway reactions in small children with Atopic risk (In Danish: Eksponering for Partikler og Luftvejsreaktioner hos Småbørn med Atopisk Risiko). NERI-ATM;I was a central partner.
HOTGAP	Health Outcome and Traffic Generated Air Pollution – project in the TRIP centre under the SMP programme. NERI-ATMI was a central partner.
LMP	Danish abbreviation for the country wide Air Pollution Monitoring Programme for Urban Areas (In Danish: Landsmåleprogrammet). Planned and operated by NERI-ATMI.
MEAD	Marine Effects of Atmospheric Deposition - EU funded research project 1999-2004. NERI-ATMI was a central partner.

NO-COMMENTS Research project funded by the Nordic Council of Ministers and with participants from Norway, Sweden, Finland and Denmark. The project aimed at coupling atmospheric and marine models in an integrated online system. NERI-ATMI was a central partner in the atmospheric part of the project.

NOVA Danish abbreviation for the National Monitoring Programme for the Aquatic Environment (In Danish: Nationale Overvågningsprogram for vandmiljøet) previously VMOP and now NOVANA. NERI-ATMI operates the atmospheric part of the programme.

NOVANA Danish abbreviation for the National Monitoring Programme for Water and Nature (In Danish: Nationale Overvågningsprogram for Vand og Natur) previously VMOP and NOVA.

RAV Danish abbreviation for Risk of Asthma among adults (In Danish: Risiko for Astma bland Voksne). NERI-ATMI was a central partner.

Sea90 Danish Marine Research Programme funded by the Danish EPA 1989-1994. NERI-ATMI was a central partner in the atmospheric part of the programme.

SMP Danish abbreviation for the Danish National Environmental Research Programme (In Danish: Det Strategiske Miljøforskningsprogram) 1992 - 2004. Danish inter-ministerial environmental research programme with funding from the Danish Law of Finances. NERI-ATMI was a central partner.

TOV Danish abbreviation for the Traffic Monitoring Programme (In Danish: Trafikovervågningsprogrammet) was funded by the Danish Ministry of Transport in the 1990ties. NERI-ATMI was a central partner.

TRIP Centre for Transport Research on environment and health impact and Policy funded by the SMP programme 2000 - 2004. NERI-ATMI was a central partner.

VMOP Danish abbreviation for the Marine Monitoring Programme (In Danish: Vandmiljøplanens Overvågningsprogram) later NOVA & now NOVANA.

VMP Danish abbreviation for the Danish National Aquatic Action Plan (I, II, III and IV) (In Danish: Vandmiljøplanen).

9.3 Organisations & institutions

DHI Danish Hydrological Institute.

EEA European Environment Agency under the European Commission which is residing in Copenhagen.

MAR Department of Marine Ecology at NERI.

NCEP National Centres for Environmental Prediction in the US. NERI obtain course resolution meteorological data for use in the THOR system from NCEP.

NERI National Environmental Research Institute under Aarhus University.

NILU Norwegian abbreviation for the Norwegian Institute for Air Research (In Norwegian: Norsk institutt for luftforskning).

NOAH Danish abbreviation for natural science Wednesday evenings formed at Copenhagen University in 1969 is an NGO working for preserving the environment (In Danish: Naturvidenskablige OnsdagsAftener).

RIVM The Netherlands National Institute for Public Health and the Environment (In Dutch: Rijksinstituut voor Volksgezondheid en Milieu), is a Dutch research institute that is an independent agency of the Dutch Ministry of Health, Welfare and Sport.

SMHI Swedish abbreviation for the Swedish Institute for Meteorology and Hydrology (In Swedish: Sveriges meteorologiska och hydrologiska institute).

SYS Department of Policy Analysis at NERI (In Danish: Afdeling for Systemanalyse).

WHO World Health Organization is a UN body to take charge of the international health.

WWF World Wide Foundation is an NGO working to preserve nature and environment.

9.4 Other abbreviations

ADT Annual Diurnal Traffic is a common measure of road traffic.

CRT	Continuous Regenerating Trap filters are used for reducing particle emissions from heavy duty vehicles.
IMA	Integrated Monitoring and Assessment.
GIS	Geograhic Information System is computer software developed for handling and analysing data with spatial reference.
GPS	Geographical Positioning System based on satellite signals.
Radiello	A highly efficient device developed in Italy for passive sampling of various specific air pollutants. Available for CO, NO_2, BTX etc.

9.5 Air pollutants

BTX	Abbreviation for Benzene, Toluene and Xylenes.
C_6H_6	Benzene is a gaseous pollutant i.e. emitted from gasoline driven vehicles. Content in gasoline has been decreased due to carcinogenic effects of benzene.
CO	Carbon Monoxide is a gaseous pollutant emitted from combustion processes incl. road traffic.
H_2SO_4	Sulphuric Acid which is a particle phase atmospheric pollutant form from the atmospheric oxidation of SO_2.
HNO_3	Nitric acid is a gaseous pollutant from the chemical conversion of NO_2 in the atmosphere. HNO_3 has a short atmospheric lifetime since it reacts with NH_3 or sticks to any surface on ground or ambient aerosol.
NH_3	Ammonia is a gaseous pollutant mainly emitted from agricultural activities related to animal household.
NH_4^+	Ammonium is a particulate pollutant formed from ammonia is atmospheric reactions with acid gases and particles.
NH_4NO_3	Ammonium nitrate is formed in the atmosphere in the reaction between NH_3 and HNO_3.
NH_x	Is the sum of NH_3 and NH_4^+.
NO	Nitrogen Oxide is a gaseous pollutant emitted from combustion processes incl. road traffic.
NO_2	Nitrogen Dioxide is a gaseous pollutant emitted from combustion processes incl. road traffic.
NO_3^-	Nitrate is a particulate pollutant from uptake of HNO_3 in existing particles or the atmospheric reaction between HNO_3 and NH_3.
NO_x	Nitrogen oxides is the sum of NO and NO_2.
NO_y	The sum of NO_x + all atmospheric reaction products both gaseous and particulate.
O_3	Tropospheric ozone is a secondary gaseous pollutant with negative impact on nature and human health. In the stratosphere it acts a protection against incoming ultraviolet radiation.
PM	Particulate Matter is a mass based measure of particles.
SO_2	Sulphur Dioxide is a gaseous pollutant emitted from combustion of fossil fuels.
SO_4^{2-}	Sulphate is the particulate oxidation product of SO_2 in the atmosphere.
Total nitrate	The sum of gaseous HNO_3 and particulate NO_3^-. Measured on routine basis by the filterpack method at NERI with the NOVANA programme.
VOC	Volatile Organic Compounds.

10 References

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National Environmental Research Institute
Frederiksborgvej 399
PO Box 358
DK-4000 Roskilde
Denmark
Tel: +45 4630 1200
Fax: +45 4630 1114

Management
Department of Arctic Environment
Department of Atmospheric Environment (main address)
Department of Environmental Chemistry and Microbiology
Department of Marine Ecology (main address)
Department of Policy Analysis

National Environmental Research Institute
Vejlsøvej 25
PO Box 314
DK-8600 Silkeborg
Denmark
Tel: +45 8920 1400
Fax: +45 8920 1414

Department of Freshwater Ecology
Department of Marine Ecology
Department of Terrestrial Ecology

National Environmental Research Institute
Grenåvej 14, Kalø
DK-8410 Rønde
Denmark
Tel: +45 8920 1700
Fax: +45 8920 1514

Department of Policy Analysis
Department of Wildlife Ecology and Biodiversity

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INTEGRATED MONITORING AND ASSESSMENT OF AIR POLLUTION

Improved quality, better understanding of processes and optimisation of allocated resources, these are the main advantages of applying Integrated Monitoring and Assessment (IMA) in air quality management. The IMA is defined as the combined use of measurements and model calculations. The use of IMA is demonstrated with examples with different aims: to obtain data for air pollution in urban streets, to assess human exposure to traffic air pollution, and to assess atmospheric deposition of nitrogen compounds to marine and terrestrial ecosystems.

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