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Long-range Transport of Particulate Matter in Nordic Countries

TemaNord 2005:569

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ISBN 92-893-1218-1

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Nordic Council of Ministers
Store Strandstræde 18
DK-1255 Copenhagen K
Phone (+45) 3396 0200
Fax (+45) 3396 0202

Nordic Council
Store Strandstræde 18
DK-1255 Copenhagen K
Phone (+45) 3396 0400
Fax (+45) 3311 1870

www.norden.org

Nordic Environmental Co-operation

The Nordic Environmental Action Plan 2005-2008 forms the framework for the Nordic countries' environmental co-operation both within the Nordic region and in relation to the adjacent areas, the Arctic, the EU and other international forums. The programme aims for results that will consolidate the position of the Nordic region as the leader in the environmental field. One of the overall goals is to create a healthier living environment for the Nordic people.

Nordic co-operation

Nordic co-operation, one of the oldest and most wide-ranging regional partnerships in the world, involves Denmark, Finland, Iceland, Norway, Sweden, the Faroe Islands, Greenland and Åland. Co-operation reinforces the sense of Nordic community while respecting national differences and similarities, makes it possible to uphold Nordic interests in the world at large and promotes positive relations between neighbouring peoples.

Co-operation was formalised in 1952 when *the Nordic Council* was set up as a forum for parliamentarians and governments. The Helsinki Treaty of 1962 has formed the framework for Nordic partnership ever since. The *Nordic Council of Ministers* was set up in 1971 as the formal forum for co-operation between the governments of the Nordic countries and the political leadership of the autonomous areas, i.e. the Faroe Islands, Greenland and Åland.

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Summary

This project has

1. Facilitated a direct and vital exchange of new high quality data and models between the Nordic research groups on modelling and measurements of atmospheric aerosols
2. Facilitated a direct flow of modelling and measurement results into the policy related EMEP model
3. Developed the PM-mass 3D EMEP model to included organic carbon (OC) and water
4. Concluded that the PM-mass 3D EMEP model has closed some of the gap between modelled and observed PM2.5/10 concentrations, but still uncertainty remains due to

    4.1. large uncertainties in the description of OC and
    4.2. observations of the water content in PM is missing and
    4.3. missing sources of dust

5. Induced the development of aerosol dynamic 3D EMEP model giving particle number and particle chemical composition in different size classes,
6. Concluded that the aerosol dynamic 3D EMEP model has failed to model the number of nucleation particles, but has some success in modelling the Aitken and accumulation mode particle. The major uncertainties are due to

    6.1. erroneous and lacking emission data base
    6.2. several of the process descriptions need of updating.

7. Developed a new modelling validation tool using a pseudo-Lagrangian approach when comparing data from observation sites along the transport path of air pollutants. This technique gives a statistical measure on how successful the process model actually describes the actual process in the natural environment. This techniques have been used to validate

    7.1. the newly developed process model for nucleation,
    7.2. the process model for coagulation, condensation and dry deposition

8. More process models as cloud processes under ways for validation, which will be concluded during 2005. Then a full dynamical model is at hand for updating the aerosol dynamic 3D EMEP model
9. Shown the primary importance of designated high quality monitoring data for the development of more accurate models.

To facilitate a possible use of the PM-mass as well as the aerosol dynamic 3D EMEP it is fundamental to proceed the research based interactive work of the Nordic groups. NMR has an important future task in initiating and integrating project resources

1. Background

In the late 90's there was a growing need for a better understanding of the mass concentrations of particulate matter in regional background air, and its spatial and temporal variation, as more and more evidence emerged on particles influence on health. This demand for particle mass concentrations has strongly increased as well as the need for a better description of the chemistry and other particle properties in order to better understand the observed health effects.

During the late 90's awareness was also rising on the importance of atmospheric particles for climate change. Anthropogenic emissions of particles and particle precursor gases have been put forward as potential climate influencing components of the same order as the green house gases, but cooling the climate. However the uncertainty was estimated as very large (IPCC). The interest in atmospheric particles for climatic reasons has also increased during the last years as indications have risen about their effects on clouds, concerning both life span and precipitation patterns. Emissions are strongly changing, with a significant decrease in the western world while a strong increase is observed in China and other rising economies especially in Asia.

This project was initially based on two project initiatives. Both had as general objective to develop and strengthen the EMEP model, but from different perspectives. One had with a rather short time perspective and focus to complement the PM-mass description with organic carbon compounds to achieve a more comprehensive component description and thus a better description of especially fine particulate mass. The other with a longer time perspective was more focused on implementing an aerosol dynamical description of particle formation, transformation and deposition. The descriptions to be implemented should be based on validated process models. This model development aimed at a more accurate model facilitating more accurate climate and health impact calculations.

The merged project kept both visions, but with emphasis on the particle dynamic model development. A specific chemistry based model for organic carbon compounds should be used for supporting the development of more comprehensive mass based model. The aerosol dynamic description should be developed both on process level and in an integrated Eularian transport model. These quite ambitious goals depends heavily on a close cooperation and exchange with larger project efforts as ASTA, EMEP, environmental monitoring programs in the different Nordic countries and several EU projects, in particular BIOFORE, CARBOSOL and QUEST. This NMR-project shall thus first of all be considered as support for merging several national initiatives on atmospheric particle research and monitoring into supporting a more policy oriented tool, i.e. the EMEP model.

2. Major Goals

The main goals for this project are:

a) The creation of a database with PM measurements in Nordic countries,
b) The development and implementation of a 3D aerosol transport model able to characterise both aerosol chemistry and aerosol number and size distributions, and
c) The validation of the results from the 3D aerosol transport model with observations and the characterisation of the model's performance with respect to other models.

3. Results

3.1 Model development and implementation

3.1.1 Introduction

The model development on including the particle dynamics in the 3D EMEP model was based on using the specific modeling technique, MONO32, recently developed by the Finnish group. This approach facilitated the implementation of the rather complicated particle processes into a large model with very limited remaining computing power. To ensure accuracy of the model, it was first tested in a box model arrangement. The model and the testing are described below. Further the present regional model with particle dynamics implemented is described. The present description in the model of the different processes briefly described.

As support in the development process a workshop on implementation of dynamic aerosol models for large scale applications called ”Dynamic Aerosol modelling: from box models to 3D transport models”, was held 30 January - 1 February, 2002, in Helsinki, Finland. The workshop was announced through and thus supported by EMEP. All modelling groups in Europe were represented. The discussions ended in a report to EMEP containing description on different modelling approaches and recommendations on what parameters needed to be measured and modelled. This report was as well incorporated in the Second Position Paper on Particulate Matter issued by the CAFÉ working group on PM.

3.1.2 Box model MONO32

Model description

An aerosol dynamics module MONO32 was developed for implementation in the EMEP Eulerian transport model in order to allow for aerosol formation, growth and interaction processes [Pirjola et al., 2003]. The model uses mono-disperse representation for particle size distribution with four size modes nucleation (diameters $d<20nm$), Aitken ($20nm<d<0.1\mu m$), accumulation ($0.1\mu m<d<2.5\mu m$), and coarse ($2.5\mu m<d<10\mu m$), however, the user can prescribe the number of size sections. A monodisperse model was chosen for our purpose since they have been shown to be rather efficient and despite their simplicity compare well with sectional and modal models [Pirjola et al., 1999: Korhonen et al., 2003].

MONO32 was developed on the basis of a Lagrangian type box model MULTIMONO (MULTI-component – MONO-disperse model), which includes gas-phase chemistry and aerosol dynamics allowing particles to be externally mixed, ie. particles with the same size can have different composition [*Pirjola and Kulmala,* 2000]. MONO32 is a simplified version of MULTIMONO, however, its computational efficiency is much better. In MONO32 all particles in a mode are characterised by the same size and the same composition. Particles can consist of soluble material such as sulphuric acid, ammonium sulphate, ammonium nitrate and sodium chloride, organic carbon which can be soluble, partly soluble or insoluble, and insoluble material like elemental carbon and mineral dust (Table 1). The water content of aerosols is calculated using empirical polynomials [*Tang and Munkelwitz*, 1994] for the mass fraction of solute as a function of water activity [*Binkowski*, 1995]. MONO32 has altogether 32 differential equations to predict the aerosol size and composition distributions; four prognostic equations to calculate the evolution of particle number concentration and seven equations for the total mass concentrations of different aerosol compounds in each of the size modes.

Table 1. Particle size modes and compounds in the MONO32 module. Water is added separately.

	H_2SO_4	Ammonium sulphate	Ammonium nitrate	Organic carbon	Elemental carbon	Sea salt	Mineral dust
Nucl. mode	X	X	X	X			
Aitken mode	X	X	X	X	X		
Accum. mode	X	X	X	X	X	X	X
coarse mode	X	X	X	X	X	X	X

MONO32 accounts for the main aerosol dynamics processes, i.e. nucleation, condensation and coagulation, and is to be coupled with gas and aqueous chemistry, emissions, as well as transport and dry and wet removal in the EMEP Eulerian transport model.

Two optional parameterisations for nucleation rate are implemented: a homogeneous binary H_2SO_4-H_2O nucleation by *Kulmala et al*., [1998] is updated by *Vehkamaki et al.,* [2002] and a parameterisation for ternary H_2SO_4-H_2O-NH_3 nucleation by *Korhonen et al*. [1999] is updated by *Napari et al.*, [2002]. Also added is the kinetically limited dimer nucleation mechanism [*Mc Murry*, 1983].

The growth of particles through multi-component condensation of H_2SO_4 and organic vapours is implemented according to *Fuchs and Sutugin* [1970]. It is worth mentioning that for organic condensation Kelvin effect is taken into account, however, there are large uncertainties since the condensable organic vapour is not yet identified, neither its thermodynamic properties such as its volatility or saturation vapour pressure, surface tension, molar mass, sticking coefficient and particle density. Some sensitivity tests can be found in *Pirjola et al.* [2002]. Organic va-

pour in the aerosol phase can be soluble or partly soluble (uptakes water) or totally insoluble. Water content of aerosols is calculated separately based on equilibrium thermodynamics.

Brownian coagulation coefficients between particles in two modes are calculated according to Fuchs [1964].

As particles grow by condensation and coagulation their average diameter can approach that of the larger mode. To transfer a fraction of the nucleation mode particles to the Aitken mode or a fraction of the Aitken mode particles to the accumulation mode, a scheme based on the ”mode merging by renaming” algorithm by Binkowski [1999] was implemented in MONO32.

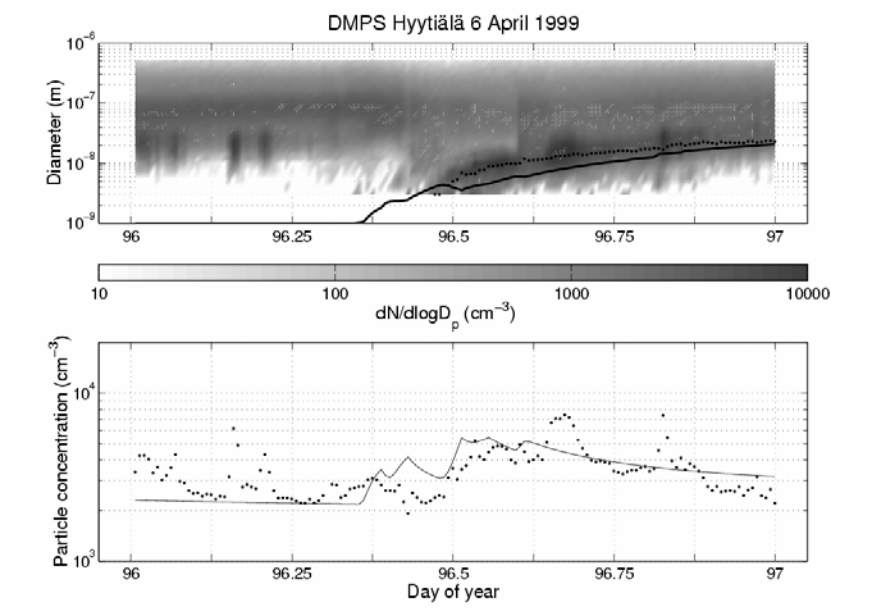
Two different time integration schemes were tested, a Fortran NAG-library routine [*FORTRAN*, 1999] and a two-step scheme [*Verwer and Simpson*, 1995]. The test results showed that both schemes had the same accuracy in integrating the aerosol dynamics equations, while MONO32 with the two-step integration scheme was considerably more efficient and therefore better facilitated for use in regional transport models.

Box model verification

MONO32 was compared with the sectional model AEROFOR2 [Pirjola and Kulmala, 2001] with 54 size sections [*Pirjola et al*., 2003]. The main simulations characterised rural conditions, however, they were also repeated under marine and urban conditions. The resulting total number concentration from MONO32 and AEROFOR2 after 24-hour simulation deviated less than 15–25%. The highest deviations in number concentrations appeared in the nucleation mode which did not have an effect on the mass concentration. If no nucleation occurred, the deviations in Aitken and accumulation mode number concentrations were much smaller, in almost all cases below 3%. The use of one mode in describing nucleation was shown to be enough even though formation of new particles during nucleation event moves the mean diameter of that mode smaller. The developed mode merging method to describe the transfer of mass and number concentration from mode to mode performed rather well in comparison with the sectional model predictions.

MONO32 was also verified against measurements available from the BIOFOR3 campaign, Hyytiälä, Finland [eg. Kulmala et al., 2001]. Two typical nucleation episodes were chosen for testing and comparison was mostly focused on evaluating the particle number and size evolution. By using the nucleation rate and the condensable vapour source rate calculated from the measurements [Kerminen and Kulmala, 2002] MONO32 was able to predict the total number concentration and the growth rate of the nucleation mode particles rather well in both cases (Figure 1).

Figure 1. The time development



Upper panel: The time development of the measured particle size distribution at Hyytiälä on 6 April 1999 (local winter time). X-axis refers to the day of the year, y-axis to the particle diameter and colour indicates 10 min average number concentration of particles in each size bin. Also shown is the dry nucleation mode diameter (solid) from the model as well as that from the observations (dots).
Lower panel: The time development of the observed number concentration (dots) of particles larger than 3 nm and the modelled total number concentration of particles larger than 1 nm (solid curve).

In Figure 1, MONO32 predicted an increase of 3 nm particles (N_3) somewhat earlier than observed, but the maximum N_3 concentration was the same order of magnitude than the measured one. The growth of particles larger than 3 nm was in good agreement with the measurements in the evening but just after noon the modelled diameter of the nucleation mode particles was underestimated by a factor of 0.5 in maximum. The dip in the modelled nucleation mode diameter was due to the high number of 1 nm size particles that decreased the size of the average nucleation mode. At the end of the simulation the resulting nucleation mode diameters were underestimated by 11–16 %. It was also shown that hygroscopic properties of the organic vapour noticeable affected the size distribution. New particles formed by nucleation increased largely the number concentration, while aerosol mass was produced through the condensation of gases onto the pre-existing particles (nucleation mode mass was always below 0.08 μg m^{-3}). It should be noted that in these simulations there were no sources of accumulation and coarse particle mass other than the condensation growth and coagulation of smaller particles.

Based on the box-model tests, the aerosol dynamics module MONO32, despite its simplicity, was found physically sound in describing particle formation by nucleation and further growth by condensation and coagulation. The combination of acceptable accuracy and computational efficiency makes it a promising candidate as the aerosol dynamics module for the EMEP long-range transport model.

Process model validation

Nucleation

Until recently, most regional and global models have focussed on mass concentration and mass size distribution, and since the mass of nucleation mode particles is negligible compared to the accumulation mode mass it has been well established to neglect the treatment of nucleation. However, when number distribution is of interest, such an assumption may not be justified. Modelling nucleation and subsequent growth is a challenging task since first, the nucleation mechanism is typically unknown, and second, each way of representing the size distribution (e.g discrete methods, fixed sectional methods, moving sectional methods and modal methods) has its profits and disadvantages.

Pirjola et al. [2004] have recently performed model calculations by a sectional aerosol dynamics model AEROFOR [Pirjola, 1999] to study under which atmospheric conditions nucleation is of importance, i.e. when the freshly formed particles are able to grow to the Aitken mode sizes and increase the concentration of particles larger than 20 nm at least with 10%. The kinds of regions of parameter space were identified and the contour plots were presented. Nucleation was an important process in the atmosphere excluding the cases when condensable vapour concentration was not high enough so that the nucleated particles had time to coagulate away before reaching the Aitken mode sizes. The results were strongly dependent on the total number of nucleated particles, the total condensable vapour concentration (H_2SO_4 plus organic vapour) as well as the number concentration and size distribution of pre-existing particles (condensation sink). In any case, this shows a strong support for including nucleation in regional/global models.

3.1.3 Regional model

Model description

The aerosol version of the Unified EMEP Eulerian model (UNI-AERO) distinguishes 7 gaseous components and 7 particulate matter components in 4 different size modes. The model version solves 38 prognostic equations using water as a diagnostic parameter. This implies that the EMEP aerosol model version is relatively cost-efficient in terms of CPU usage. The gaseous components used explicitly in the aerosol model version are primarily sulphur and nitrogen compounds: SO2, NO, NO2, HNO3, NH3, PAN, H2SO4. The seven chemical aerosol components are: sulphate (SO4), nitrate (NO3), ammonium (NH4), organic carbon (OC), elemental carbon (EC) mineral dust and sea salt (NaCl). Aerosols are considered to be internally mixed and are described in four different size classes where all aerosols are considered to have the same size (monodisperse). The four size modes are: the nucleation mode (dry diameters be-

low 0.02μm), Aitken mode (between 0.02 μm and 0.1 μm), accumulation mode (between 0.1 μm and 2.5 μm) and the coarse mode (between 2.5 μm and 10μm). However, the flexible design of UNI-AERO allows the inclusion of new size modes if needed. For example, to achieve a consistent verification of the model with measurements within this project, a test model version with 5 size modes have been used, which splits the accumulation mode to two modes with dry diameters between 0.1μm and 0.5μm and between 0.5μm and 2.5μm. Such a presentation of the size distribution is in fact believed to be more adequate for the analysis of aerosol sources and impacts.

The model version considers both emissions of precursor gases and of primary particles. It describes chemical reactions between gaseous components according to EMEP chemistry. There are three optional parameterisations of the gas/aerosol partitioning: a) the EMEP scheme, b) the Model for Aerosol Reacting System MARS that is also used in the EPA Models-3 Modelling System and in the MADE model developed in Ford Aachen, and c) the Equilibrium Simplified Aerosol Model EQSAM. Aerosol dynamic processes are coupled to the chemistry and parameterised following MONO32 except for nucleation processes that are parameterised according to the rates used in the MADMAcS model from the Institute of Tropospheric Research in Leipzig. Finally, removals by dry and wet deposition are considered to be size dependent processes. Dry deposition velocities are calculated for each size mode using the information on meteorology and land-use type. The parameterisation of wet scavenging is at present rather crude. Given the relevance of such processes in the transport and distribution of PM we envisage a verification and revision of the parameterisations to provide with more accurate descriptions of the dry and wet deposition processes.

In 2002, the UNI-AERO managed to be operational. The testing of the model is shown below.

3.2 Experimental activities

3.2.1 Network of sampling sites

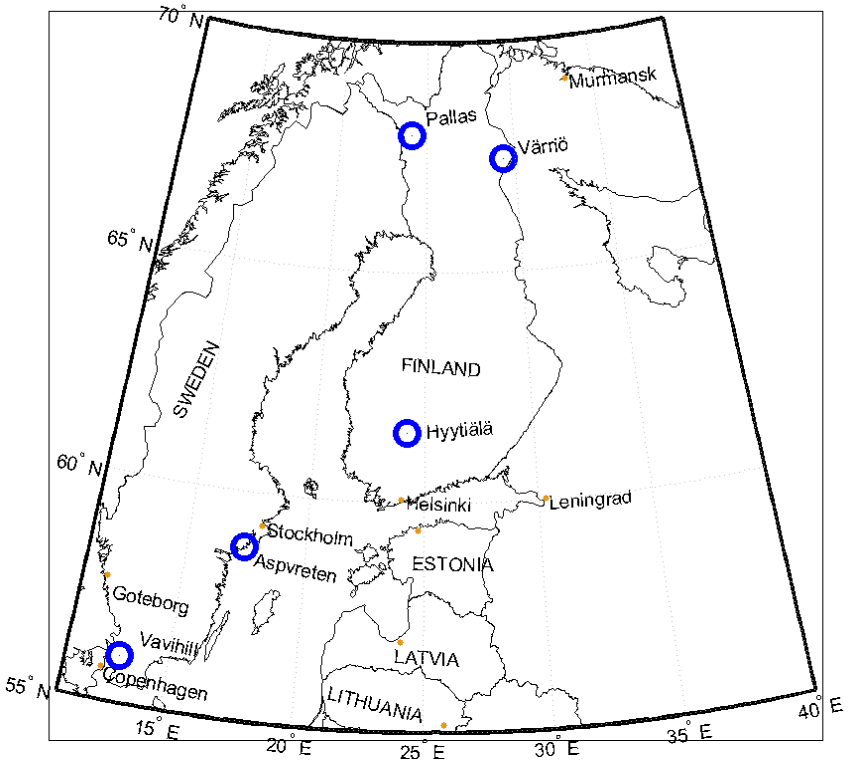
In parallel development of process description through theoretical and experimental work was performed mainly within other projects. BIOFORE and QUEST are example of projects focusing on in detail reveal the particle formation in and above the boreal forest. The boreal forest has during the project shown to be a major natural source of fine atmospheric particles. The process models and parameterizations emerging from these projects have further been tested by a pseudo Lagrangian evaluation technique developed partly within the project. This technique, described below, facilitates a more separate and detailed testing of the

processes giving a quantitative measure of the mean evolution in the atmosphere due to a specific process. Further a quantitative measure as well is achieved on the variation in the evolution due to that specific process.

The technique is based on continuous specific measurements with high time resolution on several sites at the same time. Air masses identified by meteorological analysis, e.g. air mass trajectories, connecting the sites can then be used to study the change in particle size and chemistry. Differentiating between different types of events, e.g. clear weather, clouds and precipitation, and initiating conditions, e.g. clean and polluted air, give possibilities to test different processes more or less separate.

The basis for the experimental studies used so far in this project is a number of stations in Sweden and Finland. The five stations Vavihill, Aspvreten, Hyytiälä, Värriö and Pallas cover a vast geographical area ranging from Söderåsen in Skåne (56ºN) to Finnish Lapland (68ºN). The stations in the existing Nordic measurement network are representative for the background boreal environment characteristic for Northern Europe. The European boreal region covers more than 2.900.000km^2. The type of ecosystem encountered in this region find counterparts in especially northwestern parts of USA and western Canada as well as parts of Asia. Thus, the European boreal region is representative for economically important areas, including both sparsely and highly populated regions. The geographical location of the stations is displayed in Fig. 2.

Figure 2. Location of the different station included in the measurement network



The Matorova station at Pallas (68.00ºN, 24.14ºE, 340m a.s.l.) is located in the sub-arctic pine forest in the Pallas – Ounastunturi National Park (e.g. Laakso et al., 2003). The particle measurements are performed by the Finnish Meteorological Institute (FMI). The station is part of the Global Atmospheric Watch programme (GAW). The station is situated some 150km east of Värriö.

The SMEAR I station (67.46ºN 29.35ºE, 400m a.s.l.) in Värriö is also classified as a background station and situated in the same vegetation as Hyytiälä (in this case a 40 year old Scots Pine forest). The station itself is located at a hill cap. The station is far from any pollution sources (Ahonen et al., 1997), although emissions on the Kola Peninsula give rather strong signals when winds are transporting air from this region (Kulmala et al., 2000). Montechegorsk is located some 150km east of the station and Nikel is located 190km to the north. Also, winds coming from the St. Petersburg area as well as Russia in general may bring elevated concentrations of acidifying gases as well as particulate pollution.

The background station Hyytiälä (61.5º1N, 24.17ºE, 180m a.s.l.) have been in focus in a number of large studies, especially concerning nucleation and new particle formation (e.g. Mäkelä et al., 1997; Mäkelä et al., 2000; Nilsson et al., 2001a, b; Kulmala et al., 1998; Kulmala et al., 2001, Kulmala et al., 2004). The station, SMEAR II, (Station for Measuring forest Ecosystem-Atmosphere Relations) is characterized as a boreal forest site, with surroundings dominated by a flora of Scots Pine of about 30 years age. The station is located fairly far from urban pollution sites (Tampere at a distance of ~50 km SW and Jyväskylä ~100 km NE). The station has facilitated particle size distribution measurements since 1996, which have resulted in an extensive database including continuous measurement for over eigth years.

The background station Aspvreten (58.80ºN, 17.40ºE, 25 m a.s.l.) is located in Sörmland approximately 70 km south-west of Stockholm and about 2 km from the coast of the Baltic Sea. The surroundings are characterized by mixed coniferous and deciduous forest. The area around the station is sparsely populated. The station is operated by the Institute for Applied Environmental Research, Air Pollution Laboratory, and is a part of the European Monitoring and Evaluation Programme network (EMEP).

Vavihill (56.01ºN, 13.09ºE, 172 m a.s.l.) is found at the top of Söderåsen, Skåne, Sweden. Deciduous trees mostly dominate the surroundings. The nearest population density is some 10km away. The area of Malmö and Copenhagen, with about 2 million inhabitants is situated about 60–70 km to the SSW. Size distribution measurements were initialized in early 2001. Lund University is responsible for the size distribution measurements performed at Vavihill.

3.2.2 Finnish activities

The main research subjects are aerosol dynamics (nucleation, condensation, coagulation, deposition), formation and growth of atmospheric aerosol particles and cloud droplets, atmospheric chemistry, urban aerosols, forest-atmosphere interactions (fluxes, photosynthesis, water transport) and aerosol-cloud-climate interactions. The basic theoretical resources consist of detailed computer codes describing basic phenomena such as multi-component nucleation and condensation, photosynthesis, and of extensive model for aerosol dynamics, atmospheric chemistry and cloud microphysics. The basic experimental resources consist of three field stations (SMEAR I and SMEAR II and Urban SMEAR) and a well equipped aerosol laboratory. In the field stations e.g., aerosol dynamics, atmospheric chemistry, micrometeorology, gas exchange between forest and atmosphere, soil chemistry and forest growth are measured continuously.

Studies on aerosol dynamic studies are done aiming to cover the whole 'research chain'; detailed molecular dynamic studies on e.g. nucleation, phenomenological models on nucleation and condensation that are based on detailed thermodynamics (Napari et al., 2002), parameterisations that are based on the detailed studies (Vehkamäki et al., 2002) and/or measurements (e.g. as in fig. 1) and aerosol dynamic models utilising the developed thermodynamics (Lehtinen and Kulmala, 2002; Korhonen et al., 2004). Much of the effort has been put into analysing experiments (such as shown in fig. 1) on particle formation bursts in detail in order to determine the particle formation and growth rates as well as background particle concentrations continuously at real field conditions (Kulmala et al., 2001 (BIOFOR-paper)). Such studies have allowed valuable deductions on both nucleation and initial steps of growth mechanisms (Kulmala et al., 2004). Based on the studies, the current hypothesis is that particle nucleation is controlled by sulphuric acid, progressing either by kinetic or ternary mechanism. However, sulphuric acid seems not to be able to explain all growth – it is likely that organic vapours have a significant contribution to the condensational growth rates. The question of having particles appearing in the measurable ranges is determined by the competition of growth and scavenging – if the background particle concentration is high enough, the growing freshly nucleated particles scavenge before growing above 3 nm (Pirjola et al, 2004).

Aerosol dynamics modelling has focused on box models, that have as detailed as possible aerosol physics included. A discrete method was developed for validation purposes, which avoids all numerical diffusion problems etc. (Lehtinen and Kulmala, 2002). The most general sectional model was named UHMA (University of Helsinki Multicomponent Aerosol model; Korhonen et al., 2004). It includes all recently developed nucleation schemes as well as thermodynamics needed to simulate atmospheric particle formation and growth (at least in Nordic conditions). The

aim is to incorporate UHMA into larger scale transport codes in the near future.

3.3 Database

3.3.1 General

The PM measurement database at NILU was updated and distributed for use in the model validation study. AIRBASE data on PM_{10} from a total of 55 rural and 188 urban stations from 16 European countries are available for 2000, of which we included data from two Finnish and three Swedish urban stations into our Nordic database. The PM_{10} data were supplemented by NILU data on chemical composition of aerosols sampled at Norwegian stations. In particular, daily values for separate ammonium and ammonia concentrations in aerosols measured in 2000 at Birkenes, Skreådalen, Tustervatn, Kårvatn, Osen, Karasjok, and on Svalbard (Zeppelin station) have been made available. For future validation studies, daily PM_{10} and $PM_{2.5}$ values measured at Birkenes in 2001 have been included in the distribution along with data on EC/OC speciation of the PM samples from Birkenes in both size fractions.

In addition a compilation of PM10 measurement from Finish stations from 1994-1998 (Pietarila, 2001) has been made available to this project.

To put our Nordic data base activity into a European and even a global context, an dialogue with the European PM database project CREATE and the global GAW data centre on PM measurements was done with the intention to harmonise PM data formats for both data submitters and data users.

The issue of how to establish data flow for the CREATE database was discussed at the CREATE-DAEDALUS kick-off meeting as well as at the GAW Aerosol SAG meeting in Lille, 24 - 26. March 2003. Both EMEP and GAW aimed at establishing monitoring of physical and chemical properties of the atmosphere and further to make the data available for various users. At that time, there were limited overlap for aerosol measurements between the two networks, and both used different formats for data submission (EMEP-CCC (Chemical Coordinating Centre) uses NASA Ames 1001, while GAW-WDCA uses the NARSTO Data Exchange Standard (DES)). However, with the expansion of the EMEP measurement programme to include additional measurements of aerosol properties, including those made at the European GAW sites, and the beginning of CREATE, which should be compatible with both EMEP-CCC and GAW-WDCA, there would have been considerable duplication of effort, if both databases had continued to operate independently.

The following solutions were identified, which minimised the duplication of data flow and effort for both the data providers and database, while allowing EMEP/CCC and GAW/WDCA to fulfil their objectives:

In future, all GAW aerosol monitoring sites in Europe will be encouraged to contribute to EMEP, by becoming level 2 or level 3 sites according to the Draft EMEP monitoring strategy (later the strategy has been adopted by the UNECE CLRTAP Executive Body), and all other EMEP level 2 and level 3 sites will be encouraged to join GAW, giving enough joint EMEP-GAW sites to meet the objectives of both networks within Europe. The infrastructure for EMEP data flow will be used for data submission from both these sites and the basic EMEP level 1 sites. This means that data only need to be submitted to the EMEP-CCC using NASA-Ames 1001.

EMEP-CCC decided to develop, in addition to routines already available at the centre, a routine for exporting data from the database into NARSTO DES. As there are inevitably differences in the meta-data definitions between NASA Ames 1001 and the NARSTO DES, this function was developed to give full compatibility with the NARSTO-DES, in close collaboration with GAW-WDCA. In addition to discussion during project meetings this collaboration included a day where the responsible person from GAW-WDCA visited EMEP-CCC. The source code of the exporting tool and rules for conversion of metadata were discussed in detail.

3.4 Model validation

3.4.1 Pseudo-Lagrangian approach

The existing measurement network database has been utilized in order to evaluate our current knowledge of aerosol dynamical processes adopting a pseudo-Lagrangian box model approach. By performing trajectory analysis several typical transport cases connecting southerly and northerly located stations were isolated. The goal was to test performance of a recently developed aerosol dynamic model (UHMA, Korhonen et al. (2004)) under clear sky conditions with special focus on nucleation, coagulation, deposition and condensation growth. The sparsely populated areas in-between the stations generally admit the assumption that anthropogenic emissions affect the aerosol at a minimum during transport. The only emissions considered during transport are temperature dependent terpene emissions from the forest. This in turn permits a direct approximation of contribution to aerosol laden from VOC emitted by the forest.

Focus was put on two different transport conditions; southerly transport bringing clean Arctic air southwards over Scandinavia and Northerly transport of more or less polluted air from sub-continental sources.

3.4.2. Model description

Aerosol particle dynamics was evaluated with University of Helsinki Multicomponent Aerosol model (UHMA) described in detail by Korhonen et al. (2004). The model treats the major aerosol microphysical processes contributing to aerosol evolution under clear sky conditions. In the model, the aerosol is assumed to consist of an internal mixture of sulphuric acid and a low-volatile organic species, except water vapour and ammonia. Sulphuric acid is formed by oxidation of SO_2 by hydroxyl radical (OH). The low volatile organic species is assumed to derive from the degradation of terpenes emitted from especially pine and spruce.
The terpene emissions in turn are parameterised from latitude dependent biomass density and temperature according to Laurila and Lindfors (1999).

The reactions governing the production of low volatile organics include, besides terpenes, the major oxidants OH, nitrate radical (NO_3) and ozone (O_3). Concentration of OH and nitrate radical is calculated assuming steady state, in accordance with Janson et al. (2001). Ozone is taken from observations. As constrains for the calculations of steady state concentrations, we used observed NO_x, SO_2 and O_3 concentrations as well as approximated concentrations for species HCHO, CH_4 and CO. The concentrations of the latter were taken from some old measurements at Hyytiälä.

The vertical mixing is treated conceptually assuming a two layer structure of the lowermost troposphere, in the following denoted mixing layer and residual layer. The mixing layer height is given by the trajectory model (HYSPLIT4, Draxler et al., 1997) and the residual layer vertical extent is constrained by an upper limit of 2000m and the mixing layer height. Surface emissions of terpenes only takes place in the mixing layer and the exchange between the two layers is governed by diurnal variation in mixing layer height.

Nucleation

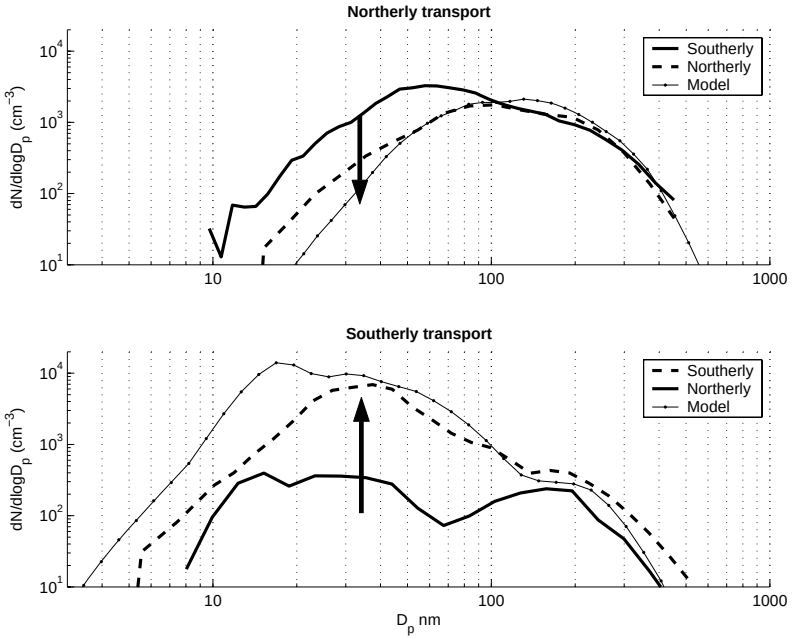
Nucleation was simulated according to the kinetically limited nucleation mechanism, which essentially gives the maximum nucleation rate under prevailing conditions. While the details of the atmospheric nucleation mechanism in the boundary layer are not known, Laakso et al. (2004) have shown that observed particle formation events in Hyytiälä are more likely limited by gas-phase kinetics than by thermodynamics

In the modelled cases, the low-volatile organic compound vapours condensing onto aerosol particles followed the nano-Köhler mechanism, suggested by Kulmala et al. (2004), and were thus capable of contributing to the growth of newly formed particles after a threshold size of a few nanometres was reached. The vapour pressure of the condensing organic species was set to $3*10^6$ cm^{-3}. This value is in accordance with estimates by Kulmala et al. (1998). Although such low volatile organic compounds

have not been identified in the atmosphere, analysis of new particle formation events in boreal forest have shown that sulphuric acid can explain only a small fraction of the observed growth of nucleation mode particles (Boy et al, 2003).

During northerly transport it was shown that although frequently occurring, nucleation did not contribute to an increase of number concentration of particles >10nm during transport, while during southerly transport, nucleation significantly contributed to a number increase in especially Aitken size range. This is in complete agreement with observations indicating aerosol number depletion during northerly transport and number increase during southerly transport. The main reason for this discrepancy is the pre-existing aerosol concentration. High pre-existing aerosol concentration generally quenches the nucleation during northerly transport while initial aerosol concentrations associated with southerly transport generally admits a significant nucleation taking place. In the case of southerly transport vertical mixing plays a crucial role. Both model and observation holds likely that series of nucleation events are taking place during southerly transport. In the absence of mixing, the model in many cases suggests that the first nucleation is sufficient to considerably quench subsequent nucleation events, which in turn yields unrealistic results. Although the model captures the general features during southerly transport, number concentration still remains overestimated by a factor of 2.7.

Figure 3. Northerly and southerly transport



Simulated and observed changes of aerosol size distribution properties during southerly and northerly transport under clear sky conditions. Aspvreten size distribution data represent the initial conditions in the case of northerly transport and Värriö the final aerosol properties. Transport time during northerly transport was on average 85 h. During southerly transport, Värriö and Pallas represent the initial aerosol properties and observations at Hyytiälä represent final size distribution. Transport time was on average 66 h.

Coagulation/condensation/deposition

In the base cases we assume some unidentified product from terpene oxidation to participate in the growth of aerosol particles. Typically encountered sulphur dioxide mixing ratios were found not to be sufficient to explain the observed growth of aerosols. Additional condensable vapours are required. Formation of low volatile products from oxidation of terpenes was argued sufficient to reproduce the observed growth rates. The molar yield required to sustain the observed aerosol growth rates was found to be in the order of 10–15%. This yield is sufficient to account for the observed mass increase during northerly and southerly transport. It is furthermore indicated that terpenes act as a source of aerosol mass during northerly transport and a source of aerosol number *and* mass during southerly transport.

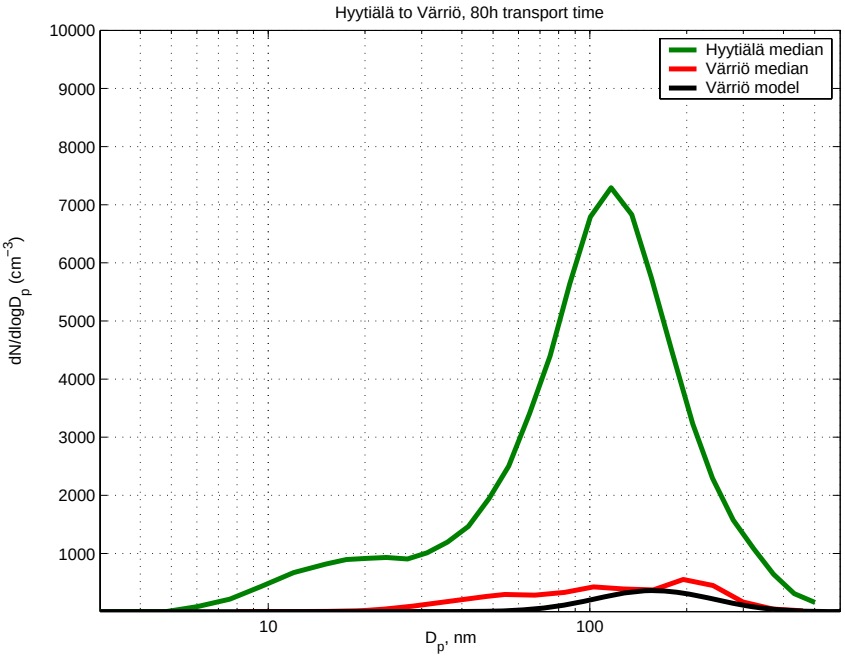
Brownian coagulation is clearly an important factor in determining the magnitude of number decrease in the Aitken size range during southerly transport and further more significantly quench the magnitude of nucleation, especially under more polluted transport occasions.

The dry deposition rate of particles was calculated according to Rannik et al. (2003) over land and according to Slinn et al. (1978) over sea. Dry deposition is argued to be of lesser importance for the accumulation mode particles, both over forested areas as well as over sea. For smaller particles (Aitken mode) dry deposition may contribute to a slight number depletion.

The role of clouds and wet deposition

Although not addressed in the model approach described above, cloud processing of aerosol particles and subsequent wet deposition remain to be the true challenge for future modelling exercises. It was shown using a non-Lagrangian approach (Tunved et al., 2004) that clouds and wet deposition likely are the major contributors to the appearance of the aerosol size distribution associated with a majority of observations. Furthermore, pseudo-Lagrangian investigations over Scandinavia clearly indicate the importance of in-cloud chemistry and wet deposition.

Figure 4. Modelled and observed aerosol number size distribution at Värriö after 60h of transport from Hyytiälä during cloudy conditions.



At present, attempts are being made to specifically address the role of clouds by implementing a cloud scheme in the above described model design. The cloud scheme includes simplified cloud physics assuming constant updraft and adiabatic water content. The cloud horizontal- and vertical distribution is parameterised from relative humidity profiles supplied by the HYSPLIT4 model. A bulk water chemistry scheme is applied allowing for oxidation of SO_2 by hydrogen peroxide and ozone in the cloud. Precipitation rate is given by the trajectory model and washout is determined from precipitation amount and cloud liquid water content (LWC). Below cloud scavenging is calculated following a parameterisation by Laakso et al. (2003).

The results as exemplified by Fig. 4 so far seem promising although additional work is required to improve the representation of clouds.

3.4.3 Summary

The above described results from modelling under clear sky conditions suggest that by adopting current knowledge concerning general aerosol dynamical processes we are able to describe the aerosol size distribution reasonably well in selected cases. The most important results from the above described studies are outlined below:

- Changes of aerosol properties during northerly transport under clear sky conditions were characterised by number reduction determined by coagulation and mass accumulation by condensation growth. Changes

in aerosol properties during southerly transport were dominated by increase in aerosol number concentration due to series of nucleation bursts. The role of vertical mixing during southerly transport was demonstrated.
The extent of nucleation is dependent on especially pre-existing aerosol surface and this reflects the balance between the generation and removal of nucleating and condensable vapours.

- Typically encountered sulphur dioxide mixing ratios were found not to be sufficient to explain the observed growth of aerosols. Additional condensable vapours are required. Formation of low volatile products from oxidation of terpenes was argued sufficient to reproduce the observed growth rates. Based on these results, the boreal forest is argued to be a source of mass during northerly transport and source of both number and mass during southerly transport.
- Besides the modelled clear sky transport cases, processing by non-precipitating clouds and wet deposition were found to strongly influence the evolution of the size distribution. The investigations performed so far suggest aerosol-water interplay to contribute to the aerosol number size distribution properties associated with a majority of observations. The dependence of size distribution properties on clouds and precipitation highlights the need of future efforts to accurately describe aerosol-water-vapour interplay on regional scales. Some first modelling attempts regarding the role of clouds have been performed and initially proven successful. Still, more work has to be performed in order to better parameterise cloud vertical and horizontal properties.

3.4.4 Performance of the EMEP mass based model

Performance of the EMEP aerosol model is regularly evaluated against available observations with respect to particle mass concentrations, chemical composition and number concentrations.

Particle mass (PM_{10} and $PM_{2.5}$)

The basic method for the PM calculations in the EMEP model is adding major components in the aerosols. The major fractions recognized in the model are sulphates, nitrates and OC/EC. Model calculated concentrations of PM_{10} and $PM_{2.5}$ have been compared with measurements from EMEP monitoring network for years 1999-2001. Also available to MET.NO data from EIONET monitoring network (Airbase database) and from some national monitoring stations and research campaigns have been used in the model evaluation. It should be pointed out that the spatial coverage of PM measurements is still rather sparse, and PM data currently available is overrepresented by data measured in central Europe and Spain. Therefore the conclusions on the model performance may be not representative to all geographical areas.

Annual mean PM

For 1999–2001, the model has been found to systematically underestimate measured $PM_{2.5}$ and PM_{10} by 40–60% on average. Several plausible reasons for that have been identified:

Uncertainty in currently available emissions of primary PM10 and PM2.5 and their geographical distribution

The input information on PM_{10} and $PM_{2.5}$ emissions used in the model runs was based on the TNO CEPMEIP emission inventory and the total national PM emissions for those countries which submitted those data to the LRTAP Secretariate (it was first in 2002 that countries started submission of PM emissions) (Vestreng, 2003). PM_{10} and $PM_{2.5}$ emission were distributed by TNO between 10 SNAP-I source sectors and disaggregated in the 50x50 km^2 EMEP grid. No appropriate information on the chemical speciation and size distribution of PM emissions has yet been available (Appendix A in Simpson, 2003).

Model deficiencies

One of the reasons for model underestimation of PM mass is that not all aerosol sources and processes are yet implemented in the model. Developing and testing of the sound parameterisation of secondary organic aerosol (SOA) formation is in progress, but it has not been included in the EMEP aerosol model. The particular large model underestimation of PM_{10} in Spain is believed to be due to not accounting in the model for wind blown dust and also Saharan dust. As mineral dust contributes mainly to coarse particle mass, calculated $PM_{2.5}$ concentrations are closer to Spanish observations. Among other aerosol sources unaccounted for in the model are road re-suspended dust and primary biogenic organic aerosols (see discussion of PM chemical composition below).

Measurement artefacts

Both positive and negative measurement artefacts can affect the observation data. The most common artefacts are associated with e.g. evaporation of semi-volatile aerosol compounds (ammonium nitrate, organic aerosols), condensation on the filter of organic and inorganic (nitric acid) vapours, or particles adsorption of water (see discussion on particle-bound water below).

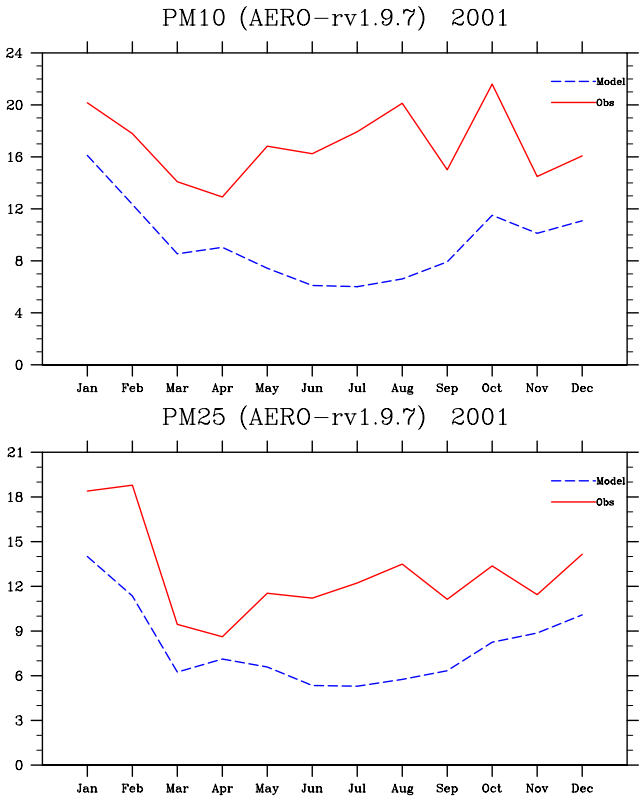
The scatter-plots of calculated versus measured PM concentrations show that the model gives rather a realistic description of the *regional $PM_{2.5}$ and PM_{10} gradients,* with the spatial correlation coefficients between 0.52–0.68. The lower correlation for PM_{10} in 2001 (correlation coefficient 0.22) is due to model greater underestimation of PM_{10} at Spanish sites, reported firstly for that year. On the other hand, the scatter-plots reveal that the model calculates too small gradients (too flat distribution) of PM concentrations, especially for PM_{10}. This can partly result

from the incorrect description the geographical distribution of primary PM emissions. This can also be due to the effect of local PM sources on measured concentrations or/and aerosol components being unaccounted for in the model (wind blow and re-suspended dust, SOA, primary biogenic aerosols).

PM seasonal variation

The model underestimation of observed $PM_{2.5}$ and PM_{10} is in general smaller in winter as the model tends to overestimate NO_3^- and NH_4^+ concentrations in cold seasons (Figure 6(a)). On the other hand, the greater model underestimation of PM in warm seasons is probably due to model not accounting for biogenic primary and secondary organic aerosols and from wind blown dust, which then has larger contributions to PM mass. As a result, the model tends to over-predict the seasonal variation of PM_{10}, whereas monthly variation of $PM_{2.5}$ is captured by the model somewhat better.

Figure 5. Monthly series of model calculated and observed PM_{10} and $PM_{2.5}$ concentrations averaged over all EMEP sites with measurements in 2001 (26 sites for PM_{10} and 17 sites for $PM_{2.5}$).



Verification of daily PM

The bias and temporal correlation coefficients between calculated and measured at EMEP sites daily PM_{10} and $PM_{2.5}$ are summarised in terms of country averages in Table 2.

The model reasonably represents the daily variation of PM_{10} and $PM_{2.5}$ with correlation coefficients largely between 0.45–0.65. Lower correlation is found at Spanish sites (wind blown and Saharan dust events) and the elevated (above 1000 m) stations. The frequency distribution plots have shown that the model under-predicts the occurrence of days with PM concentrations higher than 10-12 μg/m^3, whereas it over-predicts the number of days with lower concentrations.

Table 2. Country averaged bias (%) and temporal correlation coefficients at EMEP sites for modelled versus measured PM_{10} and $PM_{2.5}$ (N is the number of sites)

		PM_{10}	2000		PM_{10}	2001		$PM_{2.5}$	2001
Country	N	Bias	Corr.	N	Bias	Corr.	N	Bias	Corr.
Austria				3	-35	0.42	1	-49	0.56
Germany	8	-26	0.49	9	-14	0.50	3	8	0.44
Italy				1	-61	0.42	1	-55	0.41
Norway				1	-44	0.50	1	-27	0.54
Spain				9	-67	0.27	9	-51	0.34
Switzerland	4	-38	0.45	4	-42	0.42	2	-26	0.39

Fine and coarse PM

Based on the strong evidences that fine particles are more hazardous than the coarse ones the latest WHO review recommended to use $PM_{2.5}$ as the indicator for health effects induced by particulate pollution. Therefore, the work on improvement of model calculation of $PM_{2.5}$ concentrations has recently been given the first priority. Since fine PM has a predominant long-range component the model calculates $PM_{2.5}$ concentrations on average better than PM_{10} (the existing underestimation is expected to be improved when SOA is implemented in the aerosol model). Notoriously, measured $PM_{2.5}$ includes (are "contaminated by") a fraction of coarse particles and this is likely to worsen the model performance with respect to $PM_{2.5}$ concentrations.

On the other hand, WHO Report says that coarse PM has not been found harmless for human health either. Therefore coarse PM concentration levels should also be calculated with the aerosol model. Comparison with observations shows that the model underestimation of measured PM_{10} is greater than $PM_{2.5}$. This means that the aerosol model underestimates coarse PM mass. This can be either due to the underestimation of/not accounting for coarse PM sources or due to too effective removal of coarse PM in the model. For example, comparison of calculated daily coarse PM with measurements at Birkenes, Norway has shown that the model greatly underestimates coarse PM concentrations and there is a rather poor correlation between calculated and measured coarse PM (at Birkenes coarse particles are contributed by coarse organic aerosols probably of biogenic origin, e.g. pollen, and sea salt aerosols). The adequate description in the model all those rather uncertain components is essential for improvement of model calculations of coarse PM and PM_{10}.

PM chemical composition

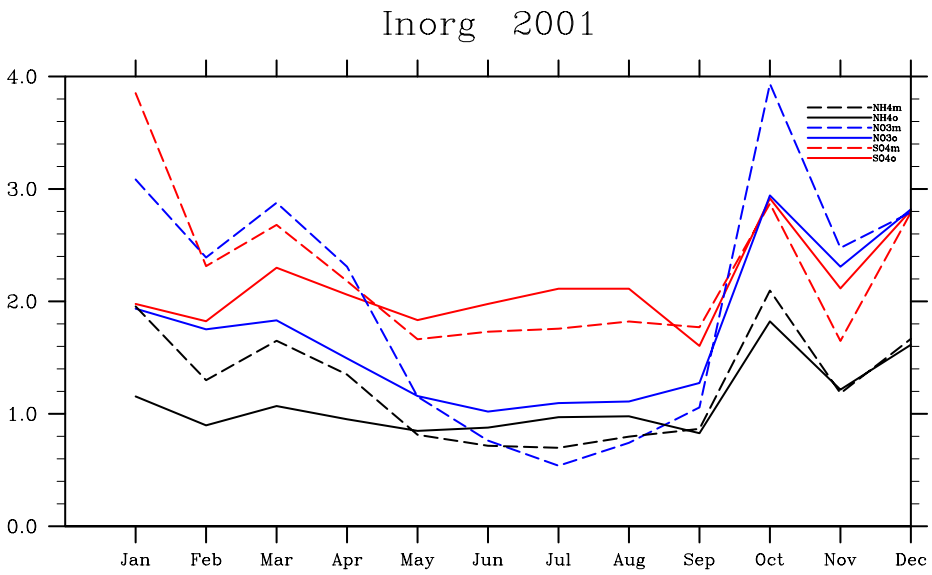
PM is not a single pollutant, but a complex mixture of many different pollutants. Therefore, model accurate calculating PM concentrations relies on the proper description of all its significant constituents. Besides, to understand the discrepancies between modelled and observed values of $PM_{2.5}$ and PM_{10} all the individual aerosol components need to be verified against observations.

At the present, the evaluation of model ability to calculate appropriately PM chemical composition is impeded by the lack of appropriate measurement suits. A large number of EMEP sites measures SO_4^{2-} and considerably fewer sites measure NO_3^- and NH_4^+ aerosols. Comparison of model results with all available EMEP data show fairly good performance of calculated SO_4^{2-}, NO_3^- and NH_4^+. For annual mean concentrations, the model overestimates those components within 15% and the correlation coefficient are between 0.7 and 0.9. On monthly basis, the model tends to overestimate inorganic aerosols in some months of the cold period (Figure 2 (a)). The outcome of the European model intercomparison qualifies EMEP model performance for SO_4^{2-}, NO_3^- and NH_4^+ to be state-of-the-art (EMEP TFMM).

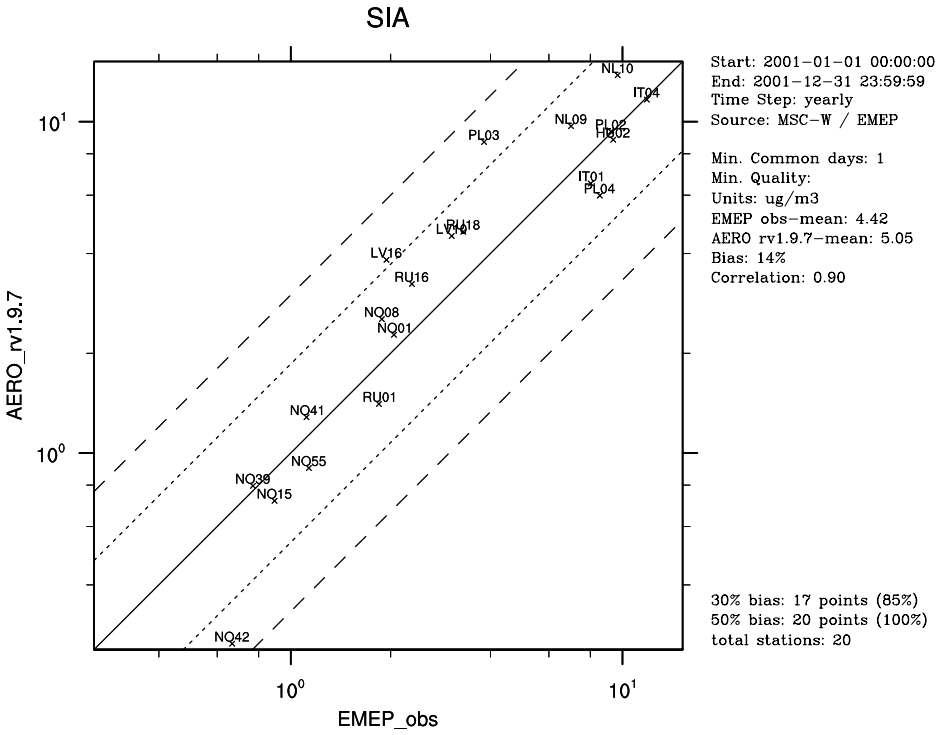
A smaller number of EMEP sites (20 sites in 2001) measured SO_4^{2-}, NO_3^- and NH_4^+ concurrently. Model calculated SIA (the sum of Secondary Inorganic Aerosols) compares fairly well with measurements at those sites (Figure 2). Among those stations, only Birkenes (NO01) and Ispra (IT04) monitor also PM_{10} and $PM_{2.5}$ concentrations. There is found a very good agreement between model calculated and measured SIA at these sites. This does not help to explain the model underestimation of PM, especially at IT04, which should be then due to other PM constituents.

Figure 6. Model performance for SIA: a) monthly series of model calculated (dashed lines) and measured (solid lines) concentrations of SO_4^{2-}(red), NO_3^- (blue) and NH_4^+ (black) at all EMEP sites in 2001; b) scatter-plot for model vs. measurements at 20 EMEP sites (annual mean in 2001).

a)



b)



Unfortunately, among all of the EMEP stations, only at Birkenes data on PM_{10} chemical speciation was available for 2001, which included daily

measurements of SIA, Na, Cl, Ca, K and weekly measurements of OC and EC. Beside that, chemical speciation of $PM_{2.5}$ was available from the AUPHEP research campaign (Puxbaum *et al*. 2003) at two Austrian sites. For those three, one Norwegian and two Austrian sites, quite reasonable agreements were found between modelled and measured inorganic components (SO4, NO3, NH4, Na, Cl), with correlation of 0.56-0.66. The largest problem was found for carbonaceous particles. The model considerably underestimated measured OC and EC concentrations, though the correlation (except for OC in Birkenes) is comparable with that for inorganic particles.

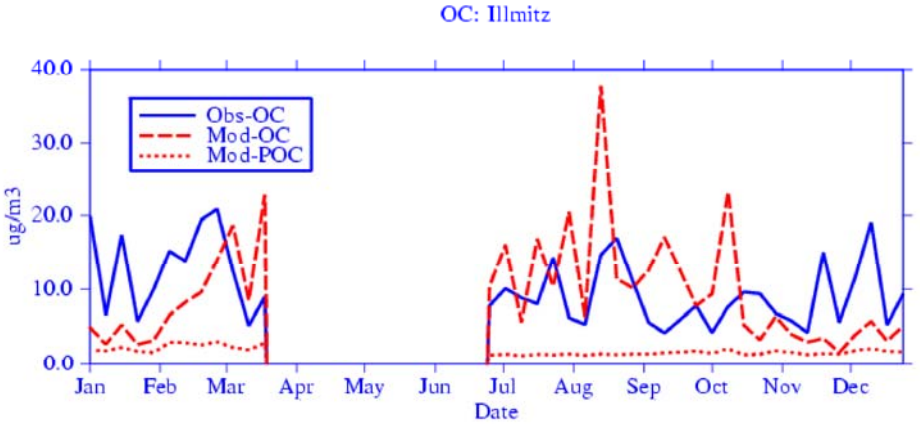
Secondary Organic Aerosols

The model used for SOA within EMEP is an extended version of the EMEP MSC-W Unified 3-D model (Simpson et al., 2003a). The SOA model extends the chemical mechanism of the EMEP model with the inclusion of (a) POC: Primary emissions (anthropogenic); (b) ASOA: Anthropogenic SOA (from aromatics); (c) BSOA: Biogenic SOA (from terpenes); (d) BGND: Background OC (mix of POC/BSOA). The methodology derives from that used in Andersson-Sköld and Simpson (2001), but with more explicit anthropogenic compounds. Some more details can be found in Simpson and Makar (2004).

Comparison with measurements

Figure 7 shows a comparison of the modelled OC against measurements made at the Austrian site Illmitz as part of the OC/EC campaign (Kahnert, 2003). Two modelled values are shown. The ‘mod-POC’ gives the contribution of modelled primary emissions of OC. The ‘Mod-OC’ gives the total model estimate of OC levels, including SOA. In general the levels of modelled and observed OC are quite similar at this site, especially considering the large uncertainties discussed above. However, one discrepancy is obvious and appears also in all other comparisons we have performed: the observations show large OC values in winter and do not show the clear seasonal cycle that the model results would suggest. The underprediction of winter-time OC may have a number of explanations (e.g. underestimation of POC emissions, insufficient ASOA production) and is currently under investigation.

Figure 7. Modelled versus observed OC concentrations at Illmitiz (Austria). Data are from 2002 for the July-Dec values and 2003 for the Jan-March values. See text for explanation of lines.



Discussion and Conclusions

We have presented a very brief overview of activity with an extended version of the EMEP model, designed for studies of SOA formation over Europe. This work and related studies conducted over several years suggest that (a) a model with no SOA and current emissions strongly underpredicts OC across Europe; (b) adding a 'standard' SOA module gives much more OC in summer, sometimes too much; (c) the SOA-model predicts strong summer maxima in OC which are not reported; (d) it seems likely that the missing OC in winter-time results from both SOA and missing POC. It should be noted again that SOA theories are undergoing rapid change. Increasing evidence for polymerisation and other reactions within aerosol (e.g. Hoffer et al., 2004, Gelencser et al., 2003, Kalberer et al., 2004) would suggest that even more SOA should be formed than given in the standard models.

It is currently impossible therefore to assign much certainty to the results of any SOA model. However, with the increasing number of measurements from, for example, the NILU EC/OC campaign, the EU CARBOSOL project (http://www.vein.hu/CARBOSOL), or from national projects, there is some hope of evaluating the model against observations in a semi-empirical way. In future studies we will more closely compare the EMEP model results with these measurements and work towards a reconciliation of the modelled sources and the observations.

Particle bound water in PM mass

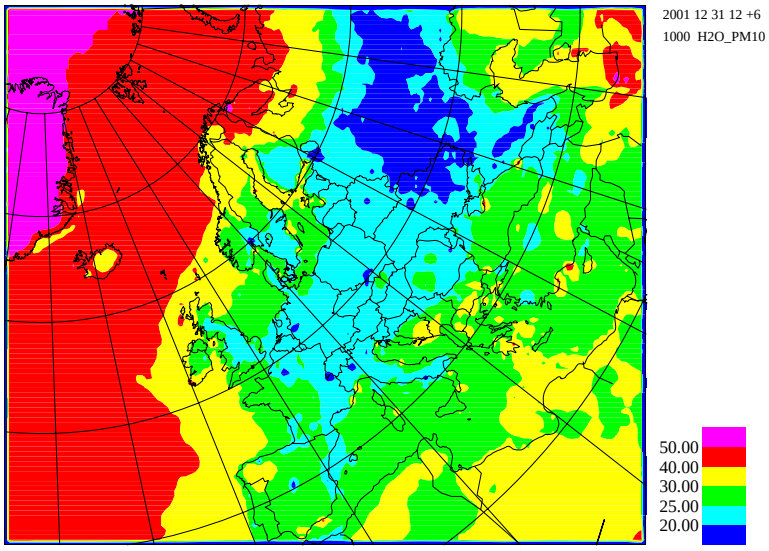
Virtually all aerosol mass balance analyses experiments fail to achieve full mass closure. The unaccounted fraction can be as large as 30-40% of PM mass and can be partly attributed to particle bound water. Gravimetric methods recommended by EU Directive and EMEP Monitoring Manual for determining PM mass require equilibrating of the dust loaded filters for 48 hours at 20°C temperature and 50% relative humidity before

they are weighed. However, equilibration does not remove all particle-bound water. Therefore, PM_{10} and $PM_{2.5}$ concentrations measured with filter-based gravimetric methods are liable to include water (10–30% by mass) and thus do not necessarily represent dry PM mass.

The EMEP aerosol model has been used to calculate aerosol water content at 50% relative humidity. The model calculates the amount of water using semi-empirical water activity coefficients of the chemical composition as calculated by the model. According to the model estimate, aerosol water contributes with 20–35% to the annual mean concentrations of $PM_{2.5}$ and PM_{10} over Europe, with largest water content associated with sea salt aerosols in coastal areas (Figure 8).

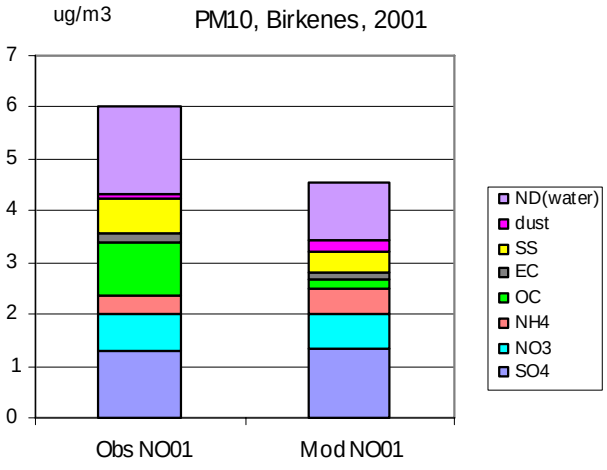
Figure 8. Model calculated fraction (%) of aerosol water in PM_{10} at temperature 20C and relative humidity 50% (a) and yearly mean in 2001 PM_{10} chemical composition at Birkenes, modelled and measured (b).

a)



The purple colour designates the undetermined fraction in measured PM mass (ND) and the particle-bound water in the model results.

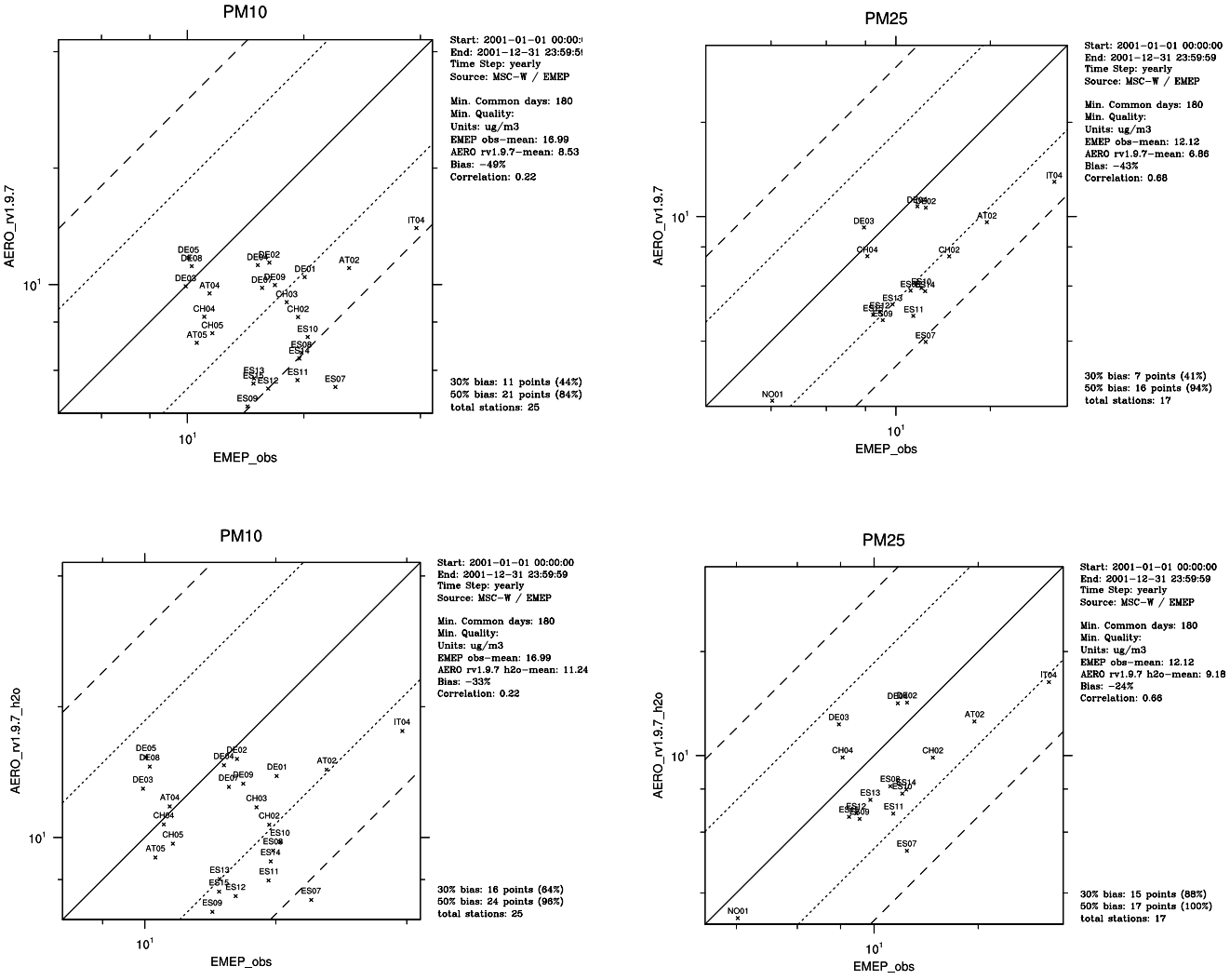
b)



Furthermore, accounting for particle-bound water in calculated PM mass has reduced the model underestimation of measured $PM_{2.5}$ and PM_{10} (Figure 9). It also slightly improved at most of the sites the temporal correlation between calculated and measured daily $PM_{2.5}$ and PM_{10}.

Thus, the accounting for particle water in PM concentrations results in better agreement between the model results and observations. However, more measurements are needed on PM chemical speciation, including aerosol water, in order to validate the model results. Correctness of model calculation of particle water relies, on the one hand, on its accurate calculation of particle chemical composition and, on the other hand, on the adequate parameterisation for calculating aerosol water content itself. Therefore, both model calculated PM composition and aerosol water content need to be properly verified against observations.

Figure 9. Scatter-plots for PM_{10} and $PM_{2.5}$ model calculated vs. measured at EMEP sites in 2001: dry PM mass (upper panel) and PM mass including particle water (lower panel).



Summarising, the EMEP aerosol model in its current form under-predicts PM_{10} and $PM_{2.5}$ mass. The improvement of the model performance is expected to be achieved by *inter alia* including all relevant processes contributing to total PM mass.

Summary on performance of the regional PM-mass model

All the results above suggest that the largest uncertainty in calculation of PM-mass concentrations is associated with PM emissions and not fully accounting for all aerosol sources and processes (SOA formation, primary biogenic OC, wind blown mineral dust and re-suspended particles). However the work SOA formation is in progress and the water content calculations are included even though validation is missing. At the present stage, the EMEP model results should not be used in policy applications depending on the analysis of absolute values of PM mass, but they are reasonable to study the effect of identified emission changes.

3.4.5 Performance of the EMEP aerosol dynamical model

Regional modelling of particle number concentration and size distribution involves far more uncertainties than calculation of particle mass and composition. The uncertainties are presently associated with the lack of information on the size disaggregation of anthropogenic PM emissions and adequate model description of aerosol dynamics processes and accounting for all particle sources. Furthermore, validation of model results on particle numbers are impeded by rather limited measurement data available.

Measurements of particle numbers from ASTA project were made available to MET.NO. The dataset included the hourly number concentrations of particles up to 500 nm measured at four Nordic stations (Hyytiälä, Varriö, Pallas and Aspvreten) in the period from 1 June to 31 December 2000. The particle number concentrations were disaggregated in four size modes, which roughly corresponded to the model's nucleation, Aitken and accumulation "small" modes (a special model version with 5 size modes developed particularly for this exercise included accumulation small (diameters between 0.1 and 0.5 μm) and large (diameters between 0.5 and 2.5 μm) modes).

The main results from the comparison of model calculated particle number concentrations with measurements are presented here.

Nucleation particles

The model fails to describe properly the formation of new particle by nucleation. At southern pair of sites Hyytiälä and Aspvreten), the model does not capture the nucleation events occurrence and greatly underestimate the number of nucleated particles. On the other hand, the model

tends to over-predict the frequency of nucleation bursts and the number of nucleated particles at the northern pair of sites, Varriö and Pallas.

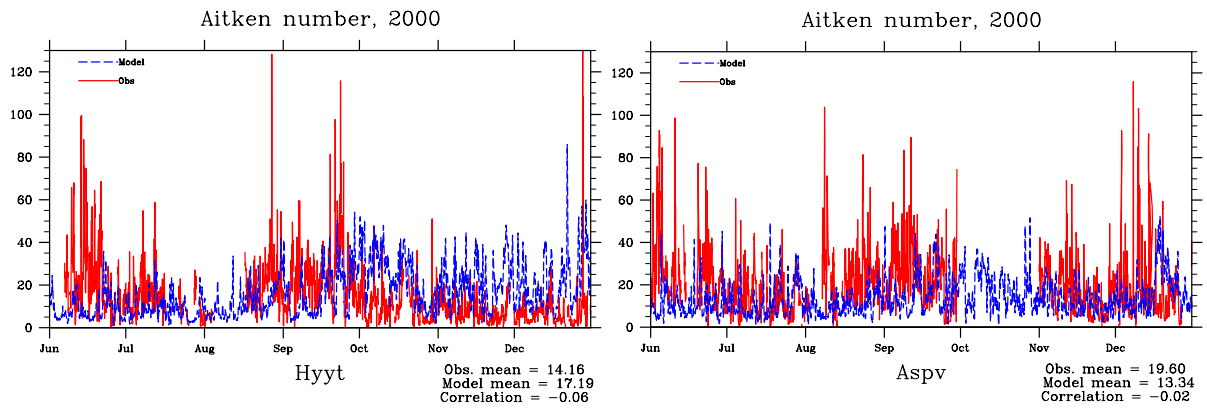
Aitken particles

Given the lack of information and therefore rather crude assumptions on PM emission size distribution used in the EMEP model, the model was not expected to accurately predict the Aitken particle number. Assumptions on the size disaggregation of $PM_{2.5}$ emissions and on the size of emitted particles adopted for these model runs were based on results of the tests on PM emissions size distribution. Then, model calculated levels of Aitken number concentrations are on average reasonably close to observations. However, the seasonal variation of the number of Aitken particles predicted by the model does not agree with measurements: modelled Aitken number increases in late autumn and winter, following the seasonal variation of emissions in the model, whereas measurements show somewhat larger Aitken number in summer (see the discussion on monthly variation below).

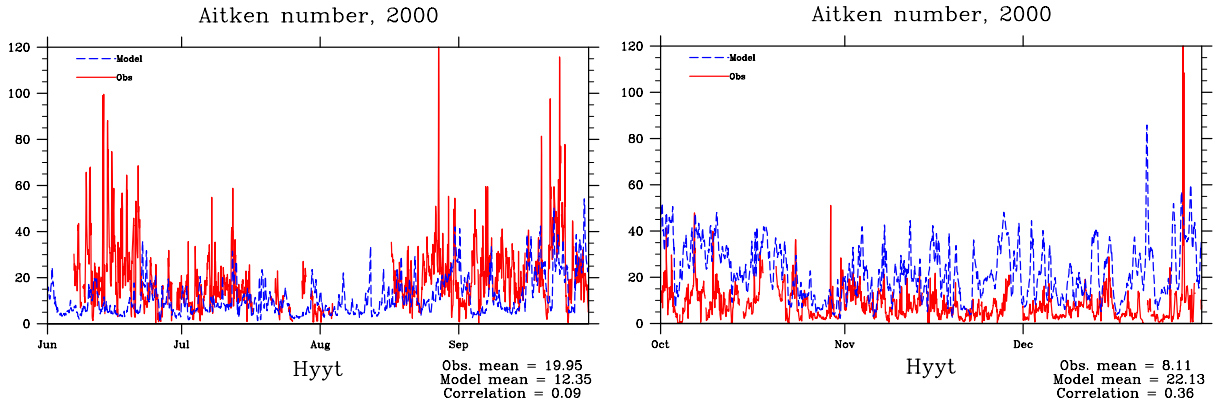
When the whole period is considered, there is practically no correlation between calculated and measured number concentrations of Aitken particles, both hourly and daily averaged. The failure of the model to predict the time variation of Aitken particle number is probably due to its inadequate description of or/and not accounting for the main processes and sources, presumably of a local character, determining particle numbers. For example, at Hyytiälä much better correlation of calculated Aitken numbers with observations is found in the period from November to December, when nucleation events were not observed. While from June to September, when nucleation bursts probably contributed to the Aitken particle number, the correlation is very poor (see section ”model description”). In fact, for all the sites except Aspvreten, better correlation between calculations and observations is found in October through December months, but that cannot be explained by the effect of nucleation bursts alone.

Figure 10. Hourly number concentrations of Aitken particles, calculated (blue) and measured (red), (a) for the period of 1 June–31 December 2000 at Hyytiälä and Aspvreten and (b) at Hyytiälä in warm and cold months.

a)



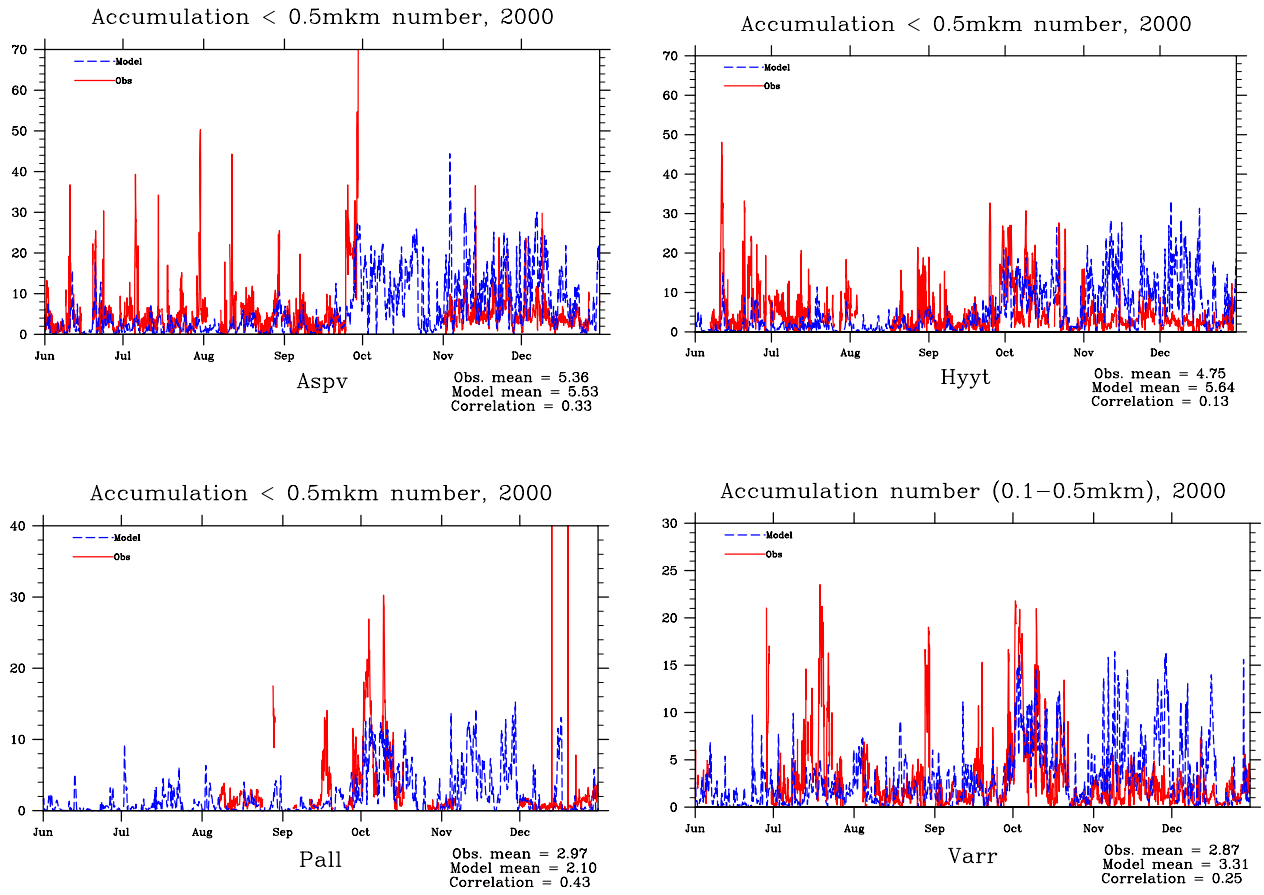
b)



Accumulation (<0.5 μm) particles

As anticipated, the model performs for small accumulation particles better than for Aitken particles. This is probably because the number concentrations of accumulation particles are to a larger degree determined by the emissions and transport and less affected by the sub-grid scale processes, and thus are less stochastic. Averaged over the period 1.6–31.12.2000, model calculated accumulation particle numbers are rather close to observations. Similarly to Aitken particles, the model calculates greater number concentrations of accumulation particles in winter than in summer, whereas the observations show the opposite (see the discussion below). The correlation coefficients are between 0.13 and 0.43 for hourly and between 0.12 and 0.55 for daily accumulation number concentrations.

Figure 11. Hourly number concentrations of accumulation mode particles, calculated (blue) and measured (red), (a) for the period of 1 June – 31 December 2000 at Aspvreten, Hyytiälä, Pallas and Värriö.



Monthly variation

At all the stations, the model predicts lower number concentrations of particles (and PM10 and PM2.5 at Aspvreten) in June-September and higher values in October-December, while observations show insignificant seasonal variation or lower concentrations in autumn-winter. As it was pointed out above, one of the plausible reasons for model inaccurate prediction of the monthly variation of particle numbers is representation of the seasonal variation of emissions in the model. Another reason may be related to the uncertainty in emitted particle sizes, as the same particle diameter was assumed for all types of PM emission sources (e.g. power generation, industrial and domestic combustion, traffic etc.) and constant through the year. On the other hand, modelled and derived from measurements total aerosol volume (calculated using dry particle diameters and density of 1000 kg/m^3) agree rather well. This implies too small particle size in winter calculated with the model. However, time-series for modelled and observed diameter of accumulation particles do not confirm that.

Summary on the regional aerosol dynamical model

The regional aerosol dynamic EMEP model has shown considerable deviation from measured values concerning mainly the number of nucleation particles. Aitken mode show as well significant deviations at certain periods and sites most likely due to the inadequate description of nucleation. Accumulation mode particles show usually fair agreement between observations and modelled data. The deviations are most likely depending on several factors as inaccurate emission databases, lacking accuracy in the model and too few measurement sites giving relevant data. The emissions data base and the measurement network have to and will most likely be developed on a European basis but will take several years. However with the results from the pseudo Lagrangian validation described above the EMEP model can be updated and be used to study the effect of identified emission changes.

4. Present status and future needs

4.1 Model / model validation

The 3D EMEP PM mass model gives a fair agreement for inorganic compounds, while there is considerable difficulties still with the OC estimates. However further refinement and validation on the model description of ammonium and nitrate concentrations. The largest uncertainty in calculation of PM-mass concentrations is associated with PM emissions and not fully accounting for all aerosol sources and processes (SOA formation, primary biogenic OC, wind blown mineral dust and re-suspended particles). However the work on SOA formation is in progress and the water content calculations are included even though validation is missing. At the present stage, the EMEP model results should not be used in policy applications depending on the analysis of absolute values of PM mass, but they are reasonable to study the effect of identified emission changes.

There is an urgent need for reaching a satisfactorily agreement for the PM-mass model including a correct description of the major chemical components. This can only be reached by further efforts on the theoretical and experimental research on organic components. Further is monitoring according to EMEP strategy level 2 needed to facilitate validation of the model efforts.

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A fully working aerosol dynamic EMEP model is based on the actual physical and chemical processes acting on air pollutants and natural components in the atmosphere. Further emissions from the natural sources such as the sea and forests will be more accurately included. Thus will such a model be useful in reaching a fully operable PM-mass model with acceptable accuracy. Further, in order to address how the climate change

depends on changes in the air pollution, it is necessary to fully develop the aerosol dynamic EMEP model to be used as a base for radiation calculations. Evidence is rising that the health effects found related to particle mass concentrations are more directly related to specific PM-compounds as e.g. soot. To fully describe the dispersion of soot and how the soot particles evolve in the atmosphere an aerosol dynamic model is needed.

Future work on completing the aerosol dynamic model includes further testing of the different processes, development on parameterisation especially for the EMEP model, implementation and testing versus other models as well as comparison with monitoring data.

4.2 Database

Data bases on detailed particle measurements are presidential to evaluate detailed process descriptions as well as parameterizations and finally for comparison with different levels of model output. The new EMEP monitoring strategy level 1 and 2 are to large part essential to verify the PM-mass model while level 3 activities as size distribution measurements are necessary for evaluating the aerosol dynamic model.

Projects like CREATE have today facilitated the collection of necessary monitoring data in the official data bases of EMEP and WMO-GAW. The most urgent today is actually to perform the necessary monitoring according to the EMEP strategy, while else is all data base infrastructure quite useless.

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