

## Final Report on the NMR-project

### “NORPAC - Validated models describing Nordic urban and regional concentration of particles and organic / elemental Carbon (OC/EC)”

December 2007

#### ***Project participants:***

<b>Group</b>	<b>Participants</b>
<b>DMU</b> , National Environmental Research Institute of Denmark	Matthias Ketzel, Ruwim Berkowicz, Finn Palmgren, Marianne Glasius, Per Løfstrøm
<b>FMI</b> , Finnish Meteorological Institute	Mia Pohjola , Mervi Haakana, Ari Karppinen, Jaakko Kukkonen
<b>GU</b> , Gothenburg University, Sweden	Mattias Hallquist, Sara Janhäll
<b>ITM</b> , Air-Pollution Laboratory, Stockholm University, Sweden	HC Hansson, Johan Ström, Christer Johansson, Peter Tunved
<b>KU</b> , University of Copenhagen, Denmark	Merete Bilde, Ole John Nielsen
<b>LU</b> , Lund University, Sweden	Erik Swietlicki, Andreas Massling
<b>MET.NO</b> , Norwegian Meteorological Institute	David Simpson, Svetlana Tsyro
<b>NILU</b> , Norwegian Institute for Air Research	Bruce Denby, Janne Berger, Dag Tønnesen, Chris Lunder, Leiv Håvard Slørdal
<b>SMHI</b> , Swedish Meteorological and Hydrological Institute	Gunnar Omstedt, Lars Gidhagen
<b>UH</b> , University of Helsinki, Finland	Markku Kulmala, Tareq Hussein, Liisa Pirjola (Stadia)

***Project leaders: M Ketzel / HC Hansson***

***Project duration: 2004-2006***

***Nordic Council of Ministers  
Working Group Marine & Air***

## Conclusions / Summary

### Introduction

Several cities in the Nordic countries have severe air quality problems. To carry out efficient traffic and air quality management, quantitative understanding of the processes controlling the emissions is necessary. Comparing data and models used in the Nordic countries is an important step to improve our understanding of these problems.

The project has proven to be a useful forum for the exchange of knowledge among Nordic particle researchers (e.g. aerosol modelling workshop, PM emission workshop, common database, instrumental development, intercomparison and exchange). Communication of data and knowledge has supported and increased the common Nordic competence on Atmospheric Aerosol processes from the urban to the regional scale.

### Technical facilities developed

A simple DMPS/SMPS (size distribution measurement) intercalibration unit has been constructed and tested. Results from laboratory tests show that intercalibrations are important. This unit has been operated by several NORPAC groups (LU, DMU, KU, NILU, GU, ITM) to test the various instruments for particle sizing and number counting efficiency. Such an intercalibration has never previously been performed within the Nordic countries.

Similarly simple and cheap soot photometers have been developed and tested. The project has and facilitated long-term soot measurements at 6 stations investigating the local contribution (kerbside), urban background and regional background. Soot measurements are ongoing since summer 2005 in Copenhagen and Stockholm. The instrument works in very different environments, from heavily polluted kerbside sites to clean remote sites as the Arctic or in Marine environments on the Danish research expedition Galathea. The construction is further made available to the whole group. Soot measurements are anticipated to reveal important insight in the different atmospheric processes effecting particle concentrations and particle chemistry similarly to the particle size distribution measurements performed at the Nordic sites.

A Nordic database on urban particle data has been set up for four cities (Stockholm, Copenhagen, Helsinki and Oslo), containing matching simultaneous regional background, urban background and kerbside measurements. Measurements of  $PM_{10}$ ,  $PM_{2.5}$ ,  $NO_x$  and particle number concentration (size range 3-900 nm) performed simultaneously at urban, near-city and rural locations can now be used e.g. to investigate the contributions from urban versus regional sources of particle number and mass concentration as well as estimation of traffic emission factors.

### Urban modelling

A specific Workshop was dedicated to *Modelling of PM emissions in different Nordic traffic environments*. Hereby emission factors for traffic-generated PM during the winter/spring PM maximum and as annual mean could be estimated separately for fine and coarse road dust and road salt, using a source-receptor model based on the elemental composition of the aerosol. The measured emission factors were used to validate the Swedish SMHI  $PM_{10}$  model. The different models used in Nordic countries consistently indicate that a large part (about 50% - 85% depending on the location) of the total  $PM_{10}$  emissions originates from non-exhaust emissions. This implies that measures relying on reducing the exhaust part of the vehicle emissions can have a very limited effect on ambient  $PM_{10}$  levels.

A mobile measurement system has been developed and used in the Stockholm region by ITM to quantify the relative importance for the road particle emission and suspension of accumulated dust versus direct pavement wear, tire type, pavement type and vehicle speed. Highest particle mass concentrations were always observed behind the studded tire and lowest behind the summer tire. This was regardless of the road type and indicates that studded tires lead to higher emissions than friction and summer tires. Studded-to-summer tire ratios were 4.4–17.3 and studded/friction ratios were 2.0–6.4.

Progress has been made in estimating and comparing particle number emission factors between several places in the Nordic countries. Further, it has been shown how the emission factors for the number of particles emerging from engine exhaust particles depend on ambient temperature.

It was also shown that the largest number of particles generated from wear of the road-tire interface is found in the nano- and ultrafine particle size range ( $< 100$  nm) with number emission factors ranging

between 1-0.1 % of the tail-pipe exhaust emissions. The conclusion is that even cars with zero-emission engines will emit considerable quantities of nanoparticles.

An *Aerosol Modelling Workshop* was organized and a common understanding was reached concerning the importance of aerosol dynamical processes both for urban and regional scale and separated for particle mass and particle number concentrations.

In common for all scales it was agreed that emission and dilution are very crucial processes and have to be modelled with detail before aerosol dynamics can be considered. Major uncertainties still exist regarding the emission inventories for ultrafine particle number emissions despite the progress in getting consistent vehicle emission factors.

For urban scale air pollution modelling particle dynamics can be neglected concerning the total particle mass. Also for particle number concentration the dilution is totally dominating the concentrations, even though it can be observed that the particle number emissions can double when decreasing temperature from +20 to +5 °C due to particle formation in the immediate exhaust plume. Dry deposition might be strongly enhanced due the traffic motion and can reduce the total number with about 20% in a street canyon. The domination of the dilution over the aerosol dynamical processes is also implied by the very high correlation observed between particle number concentrations at all size classes and NO<sub>x</sub>, as the latter pollutant is inert in street scale. However, aerosol processes can be important in case of very high concentrations, and in the dispersion regime in the immediate vicinity of the tailpipe. Aerosol processes can also change the size distribution of particles in the ultrafine particle size range.

The empirical OSPM model was successfully applied to predict time series of total particle number concentration at street level in Stockholm and Copenhagen. A dependence of the particle emission factors on ambient temperature was observed and should be accounted for. Further, particle dynamics was indicated to be more relevant at urban background scale, as well inside road tunnels, where it should certainly be considered e.g. when emission factors are derived from road tunnel measurements.

A method for coupling aerosol process models and a roadside dispersion model has been developed for the simulation of aerosol processes for the dispersion of traffic exhaust particulate matter in local scale. The modelling systems contain the aerosol process models MONO32 (developed by UH and Stadia) and UHMA (by UH), both of these coupled with the roadside dispersion model CAR-FMI (by FMI). The predicted results have been evaluated against two datasets from field measurement campaigns.

A modelling system was developed for predicting the concentrations of PM<sub>2.5</sub> in an urban area, including also the long-range transported contributions (Karppinen et al., 2005 and Kukkonen et al., 2007). The predicted results were compared against measured data at two locations in central Helsinki in 2002. The predicted average PM<sub>2.5</sub> concentrations agreed fairly well with the measured values; e.g., the Index of Agreement (IA) values for daily averages were 0.82 and 0.85 at Vallila and Kallio, respectively. We also computed the spatial concentration distributions of PM<sub>2.5</sub> in the Helsinki Metropolitan Area. The predicted contribution from LRT to the street level PM<sub>2.5</sub> varied spatially from 0.4 in the most trafficked areas to nearly 1.0 in the outskirts of the area.

### **Regional modelling**

At regional scale aerosol dynamics (nucleation, coagulation, condensation) appear to make a relatively little effect on the calculated concentrations of **PM<sub>2.5</sub> and PM<sub>10</sub>**. Wet scavenging is the most important particle sink for fine particles, depending on that the fine hygroscopic particles form cloud droplets and thus deposit when raining. Dry deposition is the dominating removal process for coarse particles, but it is much slower. Cloud chemistry is a crucial aerosols source process, e.g. in the gas-particle conversion creating secondary inorganic aerosols.

With respect to **particle number**, the comparison of model results with Nordic measurements shows that adequate model description of nucleation and further particle growth can be important for accurate calculation of Aitken particle number (which determines the total number). Dry deposition is one important removal process, while the largest is wet deposition, even more than when concerning mass. A major obstacle in the modelling is the total lack of emission data with sufficient and validated quality. To parts the emission data is totally missing.

The main emphasis of the regional modelling was to develop and evaluate modelling of specifically the organic fraction of the atmospheric aerosol as the organic fraction is a major fraction of the PM<sub>2.5/10</sub>, ranging 10 - 40% of the PM. The EMEP SOA (Secondary Organic Aerosol) model has been updated to

reflect recent knowledge on the composition of the atmospheric aerosol. The model has also been modified to provide calculations of tracers (e.g. alpha-pinene products). Preliminary comparison of modelled versus observed concentrations at the EMEP and CARBOSOL sites has been done and is submitted for publication (Simpson et al, 2007, submitted to JGR). A study using only size distribution measurement over a 6 year period from 2 Finnish regional background sites focusing only on air masses transported almost only over the boreal forest gives free standing estimates of the particle formation yield from terpenes (Tunved et al., 2006). The results show a good agreement with the modelling estimates over the boreal forest. Even though there is considerable lack of knowledge concerning the organic precursors and formation processes, which is reflected in the accuracy in the modelling description it seems like that the modelling is giving a fair description of the natural contribution. However continuous measurements of OC at selected sites over Europe is absolutely necessary to further develop and evaluate the model. Preferably the OC should also be measured such that the origin is revealed.

We have developed a simple statistical model for the estimation of the long-range transported (LRT) contribution to  $PM_{2.5}$  concentrations (Kukkonen et al., 2007). The modelling is based on linear regressions of the EMEP background ionic components with the monitored  $PM_{2.5}$  concentrations. The model was evaluated against data measured in the United Kingdom and in Finland. It was concluded that the model is a useful and simple tool for the assessment of LRT'ed  $PM_{2.5}$  that is applicable within a fairly good accuracy.

## Table of contents:

Conclusions / Summary	2
Table of contents:	5
<b>1 Introduction</b>	<b>6</b>
<b>2 Major Goals</b>	<b>7</b>
<b>3 Structure of the work and report and project milestones</b>	<b>7</b>
<b>4 Results</b>	<b>8</b>
4.1 DMPS inter-calibration	8
4.2 Soot photometers	12
4.3 Particle database NORDIC	16
4.4 Evaluation and Inter-comparison of Open Road Line Source Models currently being used in the Nordic Countries	19
4.5 Aerosol dynamics modelling exercise	23
4.6 Particle emission factors, including non-exhaust emissions	29
4.7 OC/EC parameterisation and Implementation in the EMEP model	33
<b>5 References</b>	<b>40</b>
Appendix 1: Report on related projects	43
Appendix 2: List of NORPAC-organised workshops and meetings	44

# 1 Introduction

In the late 90ties the health effects of particles in the ambient air became more and more evident. Research has since then established solid evidence on considerable effect on both mortality and morbidity. The increased risk in mortality is estimated to 4.5% and 6% for a  $10\mu\text{g}/\text{m}^3$  increase in  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ , respectively (WHO, 2004). This gives an overall effect of about 300000 yearly premature deaths in the EU. Even though Sweden is not generally showing high concentrations the effect is estimated to 5300 premature deaths annually (Forsberg et al, 2005). Consequently there is a strong need for assessment of the different sources and how they affect the local environment but also on a regional scale due to the long atmospheric life time of the particles and the production of secondary particles.

The Nordic countries are mainly on the down wind side of central Europe, which explain the high regional background particle concentrations also early manifested in acid rain. The high exposure due to long range transport has naturally driven the Nordic countries to take sometimes even a leading role in the assessment and abatement of air pollutants within different frameworks as EMEP (LRTAP) and CAFÉ (EU).

In the recent IPCC report (2007) it is shown that atmospheric anthropogenic particles probably have a substantial global effect on the climate. The particles mainly cool the climate as the sun light is directly scattered back to space and indirectly through affecting the reflectivity of the clouds and the amount of clouds. However soot particle absorb sunlight and thus warm the climate. As the concentration of anthropogenic particles are highest in and down wind the source areas the regional climate effects are expected to be larger in these areas. Conclusively abatement measures in Central Europe to improve the air quality will probably affect the climate in the Nordic countries. Actually the strong reduction in sulphur emissions might be one of the major reasons for the temperature increase in the Nordic countries during the last 15-20 years that is larger the global temperature increase.

There is consensus among the atmospheric scientists that air quality and climate change have common issues in understanding sources, transport, formation, deposition and abatement strategies for particulate air pollutants.

The present project is intended to increase knowledge on particulate air pollutants, their sources, particle formation in the atmosphere and regional impact of emissions. Another intention with the project is to involve all major active groups within this research area in the Nordic countries to both make use of the common knowledgebase as well as to disseminate specific knowledge within one group to all participants. This process of course also carries new knowledge faster to the policymakers within the Nordic countries facilitating operation from a common knowledge base.

The project was set up in a two way fashion, starting with focusing on the urban measurements and modelling and then turning more into regional scale. The intention was besides increasing the understanding for the urban environment and its effects but also to bring better emission data to the regional modelling and thus understanding for how the urban emissions affect the concentrations, physical and chemical properties of the regional background aerosol.

## 2 Major Goals

Main goal of the project is to enhance Nordic research on measurements and modelling of ultrafine particles (UFP) and particles mass (PM) through extended co-operation between Nordic research groups working on local/urban as well as regional scales. The first phase (2004-2005) of the project covered the following activities:

1) Compilation, evaluation and synthesis of existing particle data and ongoing measurements in the Nordic urban and suburban areas, 2) Intercomparison of SMPS instruments and 3) Comparison and improvement of Nordic dispersion models for particles, including re-suspension of road dust.

The second phase (2005-2006) of the project focused on the further development the EMEP 3D Eulerian model taking the NMR project “Long-range transport of particulate matter in Nordic countries” as a starting point. This new project aims at implementing a better description, i.e. parameterisations of Elemental Carbon (EC) and Organic Carbon (OC), in the EMEP Unified Eulerian model, improving its capability to address sources, concentrations in the atmosphere, transformation and deposition.

The project uses existing co-operation and will establish new collaboration with other relevant Nordic and European projects.

## 3 Structure of the work and report and project milestones

In order to accomplish the aims of the project, the project was structured in to the following activities or working packages (The leading group is mentioned in the brackets together with the participating groups):

1. DMPS inter-calibration (LU, DMU, KU, ITM, NILU, GU)
2. Soot photometer (ITM, DMU)
3. Particle database NORDIC (SMHI, DMU, FMI, NILU)
4. Local modelling exercise (NILU, DMU, FMI)
5. Aerosol dynamics modelling exercise (DMU, FMI, UH/Stadia)
6. Emission factors UFP, PM, incl. re-suspension (SMHI, DMU, ITM, FMI, NILU, GU)
7. OC/EC parameterisation and implementation in EMEP model (METNO, ITM, UH, FMI)

The work in each of the subprojects is described in a section within the next chapter, following the structure: objective / summary / results / conclusions and recommendations / references.

Since the work reported here is partly performed and funded within other national and international projects the Appendix 1 gives a table with related projects. The Appendix 2 contains a list of meetings and workshops that were organised within the duration of the project.

A web page (<http://NORPAC.dmu.dk>) has been established and serves as information portal for the project members and interested experts in the field. The web page contains:

- an overview and the status of the different working activities,
- announcements, minutes and presentations from project meetings and work shops
- list and full text of publications of the network
- links to related projects and documents

Both the web page and the Nordic Database will be available also after the project is finished.

## 4 Results

This chapter gives short reports on objective and results for each activity within the NORPAC project.

### 4.1 DMPS inter-calibration

#### **Objectives:**

The aim was to develop a cheap simple system facilitating control of the quality of the DMPS/SMPS measurements used in continuously operation at Nordic field sites.

#### **Summary**

To maintain high quality size distribution measurements, DMPS/SMPS instrumentation needs to be checked, which is usually done by collecting several systems in a common laboratory intercomparison, which is costly, time consuming and causes data loss of sometimes several weeks. Within this project, a simple device / calibration system was developed that could be moved between the DMPS/SMPS systems and thus achieves an intercomparison. In the first step emphasis was put to ensure correct particle sizing. Results from tests show that intercalibration is important to maintain a high quality on the measurements. The developed unit has been used by several NORPAC groups (LU, DMU, KU, NILU, GU, ITM) to test the various instruments for particle sizing and number counting efficiency. The results obtained with the chosen intercalibration method show that the particle sizing of the Nordic DMPS/SMPS systems is reasonably accurate, but there is certainly room for improvements.

#### 4.1.1 Introduction

At present, the best method to perform precise particle number size distribution measurements in the submicrometer size range is based on the application of Differential or Scanning Mobility Particle Sizers (DMPS/SMPS). These include a Differential Mobility Analyser (DMA) to select a narrow particle size range, in which the particles are counted by a Condensation Particle Counter (CPC), (Birmili et al., 1999). Ideally, the Nordic DMPS/SMPS systems should all be calibrated with respect to both particle sizing and counting efficiency for all sizes measured. Such thorough calibrations can however only be performed when the systems are gathered at a calibration facility, such as the World Calibration Centre of Physical Aerosol Particle Measurements (WCCAP) at the Leibniz-Institute for Tropospheric Research in Leipzig, Germany. A few of the Nordic systems did take part in two such extensive calibration workshops during the NORPAC project time period (autumn 2004 and November 2006), but these workshops were supported through the EU FP6 projects ACCENT and EUSAAR.

No previous intercalibrations have been performed for all the Nordic systems prior to NORPAC due to costs and interruption of the data acquisition connected with such laboratory intercalibration. To facilitate a less costly and faster intercalibration a simple and easily transportable calibration unit, that could be operated by the users themselves and at the sampling sites, was developed and tested.



### 4.1.2 Methods

The developed unit consists of an aerosol nebulizer and a mixing chamber and was constructed at the Lund University. The participating Lund SMPS system has then been used to test this unit. Figure 1 shows a simple sketch of the portable intercomparison unit.

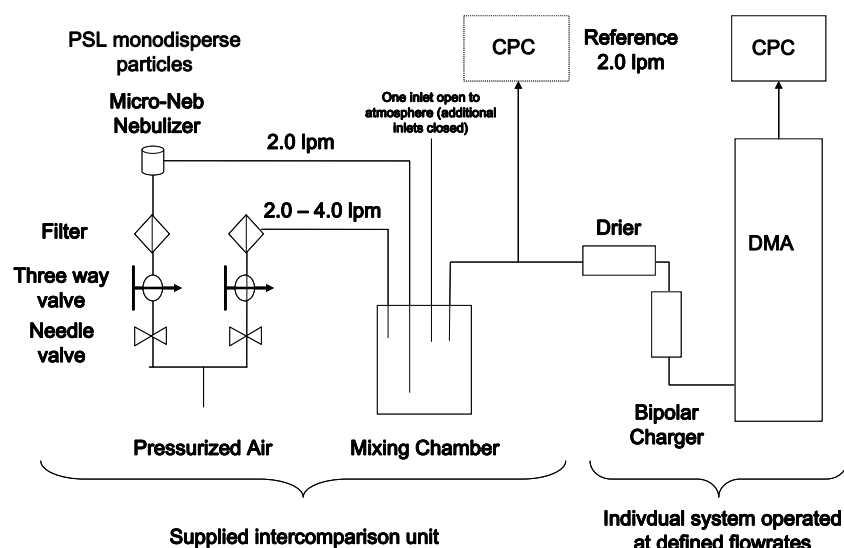


Figure 1: Schematic sketch of the intercomparison unit for calibration in sizing of particle number size spectrometers.

The unit was sent to different locations circulating since June 2005 within the participating NORPAC groups (LU three systems, DMU, KU, NILU, GU, ITM). The calibration was always done at the sampling site of the system. Individual properties of the compared devices ranged from different DMAs and CPCs to different sample and sheath flowrates, systems running in a closed loop or non closed loop set-up and systems stepping (DMPS) or scanning (SMPS) through the size range. For each experiment, polystyrene spheres of known size in the submicrometer size range were nebulized and emitted into the mixing chamber, where the generated monodisperse particles were diluted with dry and particle-free air. Thus different systems can be compared how well they size particles having three different sizes, namely 101, 277, and 420 nm using these quasi-monodisperse PSL (polystyrene latex) spheres. Results on this project have been reported at different conferences and meetings (Massling et al., 2005a, 2005b, 2006) and a technical note on the intercalibration is in preparation for publication in a peer-reviewed journal (Massling et al. 2007).

### 4.1.3 Results

The particle size distributions yielding raw concentrations were evaluated by fitting log-normal functions to the monomodal size distributions comparing the midpoint of the individually observed PSL latex peak as the important parameter. An example plot of the obtained results by comparing eight systems is given below.

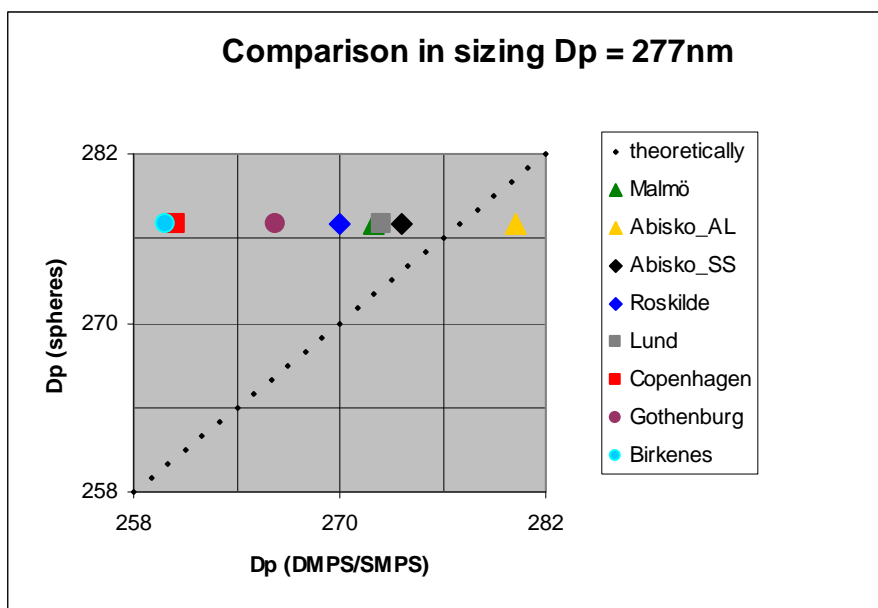


Figure 2: Comparison in sizing of eight DMPS / SMPS systems using PSL latex spheres at a diameter of  $D_p = 277\text{ nm}$ .

As one result, we found a systematic underestimation for particles of smaller sizes ( $D_p = 101, 277\text{ nm}$ ) covering the accumulation mode range of typical atmospheric particle number size distributions. This underestimation amounted to an average relative deviation of about 4.6 % for 101 nm particles and 2.8 % for 277 nm particles. As can be seen easily in Figure 2, except one system, all systems underestimated the real size of 277 nm PSL spheres.

As a second result, a systematic overestimation for particles of larger sizes ( $D_p = 420\text{ nm}$ ) covering the accumulation and coarse mode range of typical particle number size distributions was observed. This overestimation amounted to an average relative deviation of about 2.4 % and was also consistent for all investigated systems.

As a third result, we found, that the variation in sizing within the eight tested systems was relatively small. The sizing of individual systems did not deviate more than  $\pm 4\%$  in relative deviation from the mean value calculated from the results of all systems.

Furthermore, it has been observed, that the relative deviation of the systems among each other had a systematic order. This means that systems showing smaller values in particle sizing compared to the other systems underestimated the particle size for each of the investigated sizes, and vice versa.

#### 4.1.4 Conclusions and recommendations

The results obtained with the chosen intercalibration method show that the particle sizing of the Nordic DMPS/SMSP systems is reasonably accurate, but there is certainly room for improvements. The fact that the intercalibration found a systematic underestimation for particles of smaller sizes ( $D_p = 101, 277\text{ nm}$ ), while a systematic overestimation was observed for the larger particle size ( $D_p = 420\text{ nm}$ ) is not easily explained from DMA theory.

Preliminary test by the group at Gothenburg University to use an electrospray unit to generate the PSL particles showed promising results, in the sense that the background below the PSL peaks was considerably reduced. This alternative aerosol generation method is worth pursuing further in the future.

Building on the valuable findings of the NORPAC intercalibration study, we are able to make several recommendations regarding future intercalibrations between the Nordic DMPS/SMPS systems:

- Intercalibration between the Nordic sites is needed and should be continued.

- An upgraded round robin calibration unit should ideally be circulated endlessly around the Nordic sites to ensure continuous DMPS/SMPS data quality.
- The use of an electro spray unit to facilitate the aerosol generation (in particular of particle sizes <100 nm) should be investigated more thoroughly.
- The Nordic intercalibration efforts should be coordinated within the EU FP6 EUSAAR aerosol super-sites, of which several are Nordic (SE: Vavihill, Aspvreten; N: Birkenes, Spitzbergen; FIN: Hyytiälä, Pallas).
- Number closure studies should be performed for all Nordic sites. This would require the operation of a particle counter (CPC) in parallel with the DMPS/SMPS system, and provide information on whether the particle number concentration retrieved from the DMPS/SMPS system equals that of the particle counter which continuously counts particles of all sizes. A “Nordic reference CPC” might be used for this purpose and circulated together with the size intercalibration unit.
- Mass closure studies should be attempted wherever possible. This would yield valuable information as to how much of the Nordic PM<sub>2.5</sub> (or PM<sub>1</sub>) can be found in the DMPS/SMPS submicrometer size range. A “Nordic reference aerosol collection unit” might be used for this purpose and circulated with the size intercalibration unit. This could consist of a PM<sub>1</sub> size-selective inlet and a filter unit. The aerosol mass collected on the filter could be determined gravimetrically at a reference laboratory to avoid ambiguities in the mass determination.

#### 4.1.5 References and reports

- Birmili, W., Stratmann, F., Wiedensohler, A., 1999. Design of a DMA-based size spectrometer for a large particle size range and stable operation. *J. Aerosol Sci.*, 30, 549-554.
- Massling, A., Löndahl, J., Swietlicki, E., Ketzel, M., Jensen, M., Wahlin, P., Bilde, M., Kristensson, M., Hansson, H.-C., Ströhm, J., Jonsson, A., Hallquist, M., Lunder, C. (2006) DMPS/SMPS intercomparison in terms of particle sizing, Reports on BACCI activities.
- Massling, A., Löndahl, J., Ketzel, M., Jensen, M., Wahlin, P., Bilde, M., Rosenbohm, E., Hansson, H.-C., Hallquist, M., Lunder, C., Swietlicki, E. (2005a) An easy set up for intercalibration of DMPS/SMPS systems in terms of particle sizing and total particle number, NOSA proceedings, Gothenburg, Sweden, Poster.
- Massling, A., Löndahl, J., Swietlicki, E. (2005b) Intercomparison of particle number size spectrometers, 2<sup>nd</sup> Joint BACCI meeting, Kuopio, Finland, Poster.
- Massling, A., Löndahl, J., Swietlicki, E., Ketzel, M., Jensen, M., Wåhlin, P., Bilde, M., Kristensson, M., Hansson, H.-C., Ströhm, J., Jonsson, A., Hallquist, M., Lunder, C (2007): DMPS/SMPS intercomparison in terms of particle sizing. Technical note in preparation.

## 4.2 Soot photometers

### Objective:

The objective of this work is to provide cheap custom build instrumentation facilitating measurements at Nordic key sites, urban and background, to provide high quality soot measurements of concentrations in different environments as well as better emission estimates.

### Summary:

ITM has designed and build the basic instrument. The instrument is based on the integrating plate method where the light transmission through a particle filter is continuously monitored as particles are collected on the filter also having a reference signal allowing corrections for electronic noise and drift. 6 soot photometers have been built and are placed at selected stations in Stockholm and Copenhagen. The instruments has been placed at a street canyon, urban background and regional background site in respective city together with other instrumentation as PM<sub>2.5/10</sub>, size distribution and NO<sub>x</sub> monitors allowing e.g. to determine soot emission factors and their dependence on surrounding factors. First estimate is 20 mg/veh km, based on half a year measurements. Continuous comparisons with commercial instruments have shown excellent agreement. Further automated filter changes and more photometers will be built in co-operation with other project participants.

### 4.2.1 Introduction

Carbonaceous aerosol particles consist of elementary carbon (EC), organic carbon (OC), metals, sulphuric acid and nitrates. Sometimes EC is referred to as black carbon (BC) or soot. When the term black carbon is used it is mostly its optical properties there is of interest. BC/soot is a strong absorber of visible light.

Soot particles are often a result of incomplete combustion. This is however not a clear definition and soot particles are not pure EC can contain a mixture of OC, metals, sulphuric acid and nitrates. Soot particles are mostly emitted as ultrafine particles from diesel- or gasoline engines, power- and heat production or wild fires.

Soot is not measured routinely and thus the knowledge on concentrations and emission factors are rather sparse, even though soot is one of the major primary pollutants. The major source is traffic.

Soot has a longer lifetime in the atmosphere, about 4-8 days, than most other particles due to when emitted soot is mostly hydrophobic and thus not susceptible to wet deposition. Aerosol dynamics, coagulation and condensation, give mixed particles containing the hydrophobic soot and hygroscopic compounds thus making the soot containing particle susceptible to wet deposition. The long life time implies a transport distance from source to deposition in the range 2000 - 4000 km. Soot is emitted from combustion engines as 30-70 nm particles. The soot particles constitute a major part of the number of particles close to the streets and due to the large emissions the soot particles might strongly affect different atmospheric processes as e.g. particle and cloud formation. Soot usually constitutes about 5-25% of PM<sub>2.5</sub>, with the lower contributions (about 5%) found in regional background and the higher values at kerbside sites. Recent modelling studies with the EMEP-model suggest an error in the soot/EC emission data of a factor 2-5.

### 4.2.2 Results:

The custom-built particle soot absorption photometer constructed at the Department of Applied Environmental Science, Stockholm University (herein after referred to as PSAP-ITM) is shown in the version with automated filter change in figure 1. It measures the absorption coefficient by

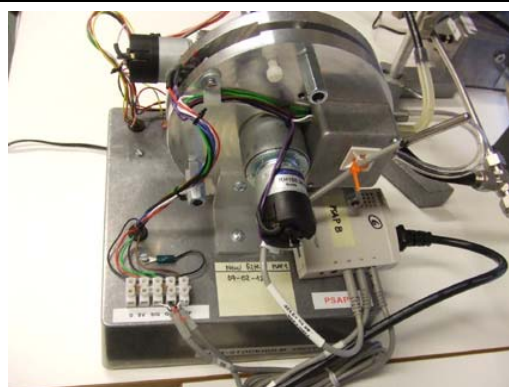


Figure 1: custom-built particle soot absorption photometer including automated filter change

using a 525 nm LED. Aerosol particles are collected on 47 mm Tissuglass E70-2075W filters (glass fibres with a cellulose baking, Pall Corporation, USA) within a 0.088 cm<sup>2</sup> circular area. Correction for drift and electronic noise is facilitated through an extra light path through a non exposed filter spot similar to the exposed filter spot.

#### 4.2.3 Intercalibration

Evaluation and intercalibration of the PSAP-ITM has been done in some different ways. The absorption coefficient of the aerosol is calculated from the transmissions measurements. The performance was controlled by comparing with a commercial aethalometer showing a linear relationship (see figure 2). Several PSAP-ITM instruments were run in parallel showing a very good agreement.

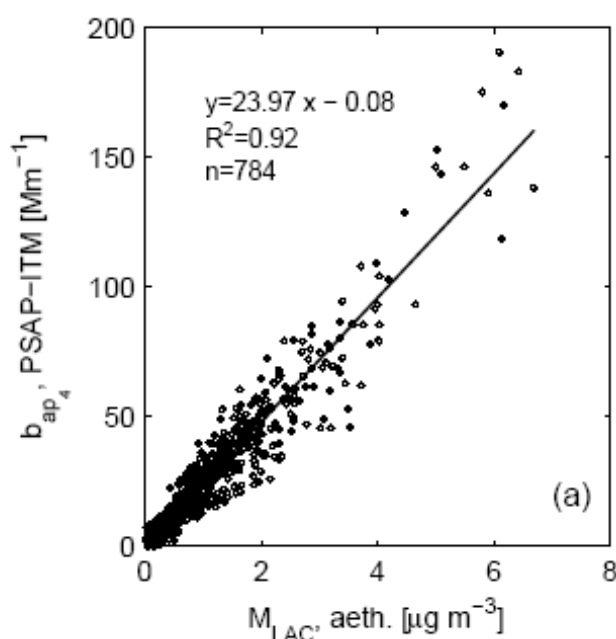


Figure 2: Scatter plot of simultaneous measurements with PSAP-ITM and a commercial aethalometer series 8100 (Magee Scientific, USA) in small city in Northern Sweden strongly influenced by wood smoke (Krecl, et al., 2007).

To facilitate converting the absorption data into mass BC, the mass absorption coefficient has to be determined. It is varying between types of BC aerosol, and is also found to vary as the aerosol is ageing. By comparing with instrumentation giving the mass it is possible to deduce a mass absorption coefficient. However as no standardized method exist and there is a considerable difference between different methods the determination of the mass absorption coefficient is dependent on BC mass method used. Using a Sunset thermal optical instrument the mass absorption coefficient was determined to 20 m<sup>2</sup>/g for a wood combustion dominated urban aerosol (Krecl et al., 2007). Using an Ambient Carbon Particle Monitor (ACPM, R&P Inc.) for reference the mass absorption coefficient was about 12 m<sup>2</sup>/g for traffic dominated aerosol (Andersen, 2006). Literature values are reported in the range 6-20 m<sup>2</sup>/g (Johansson and Hansson, 2007).

#### 4.2.4 Examples of measurements

Measurements simultaneously in a street canyon, a close urban background and a regional background site give a possibility to determine the local emissions, their influence on the urban air quality. Measurements in e.g. in Copenhagen, see figure 3, show clearly how strongly the local sources dominates the soot concentrations in street canyons, but also

substantially adds to the urban background. This is a different picture situation compared with  $PM_{2.5}$  or  $PM_{10}$ , where the regional background is often dominating the concentrations in the whole urban area. The reason that long range transport of PM so strongly influence the urban concentrations compared with soot, that is a primary PM component, is that secondary formation of PM during the transport adds strongly especially to  $PM_{2.5}$ , which by time becomes dominated by secondary components.

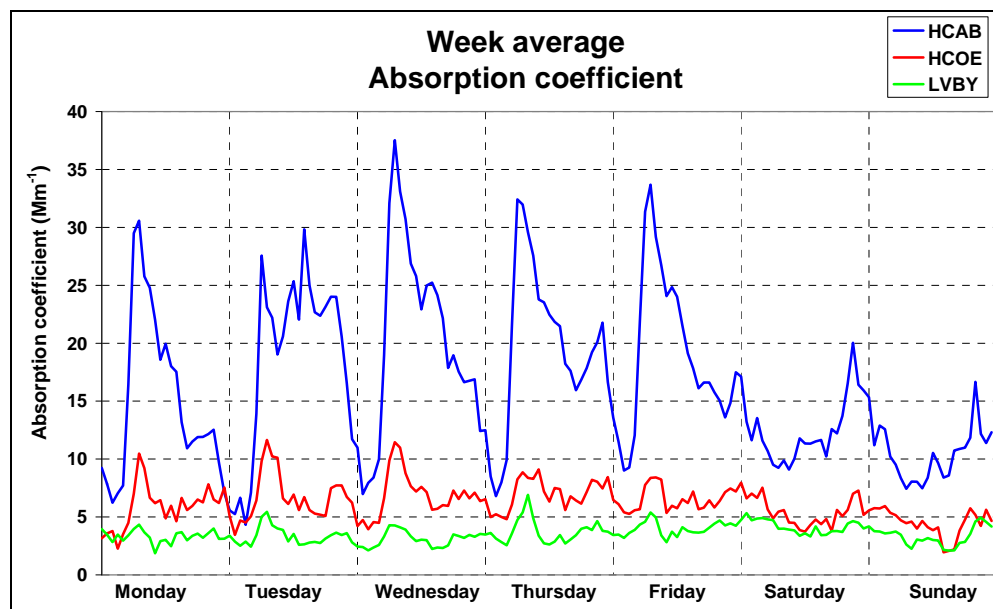


Figure 3: The levels of the absorption coefficient, as averaged weekly variation, at a kerbside site (HCAB), an urban background site (HCOE) in Copenhagen and a regional background site (LVBY), representative for the Copenhagen area.

Going into the Arctic we find strongly varying BC concentrations. The highest values reach the same values as the background concentrations outside Copenhagen, are observed during occasions of direct transport e.g. as regularly happening during Arctic Haze (see figure 4). The BC concentrations and total number of particles correlate very strongly. Simultaneous measurements with volatility tandem differential mobility analysers show that all particles contain soot, explaining the strong correlation. This is important and useful information when evaluating models. The PSAP-ITM has also been used in the Danish Galathea expedition measuring maritime soot concentrations supporting other atmospheric measurements on a cruise around the world (Glasius et al. 2007).

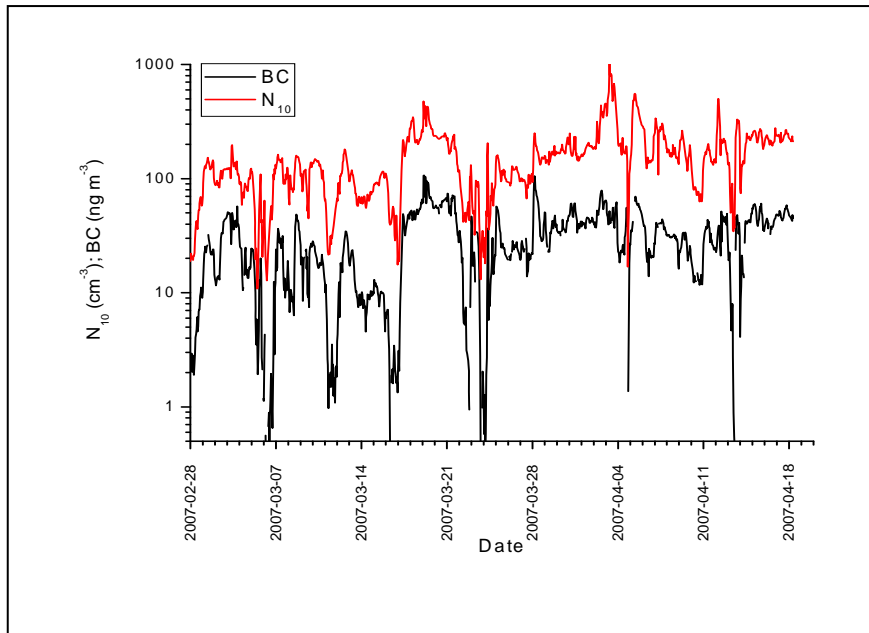


Figure 4: Seven weeks of hourly averages of particle number concentration ( $N_{10}$ ), size range  $D_p > 10\text{nm}$ , and light absorbing particles (BC) equivalent from the Zeppelin station at Spitzbergen.

Measurements over a 6-month period from the regional background site Aspvreten show a high correlation between BC and  $\text{PM}_{2.5}$ , mainly showing that soot is coming from the same source areas as  $\text{PM}_{2.5}$  (see figure 5).

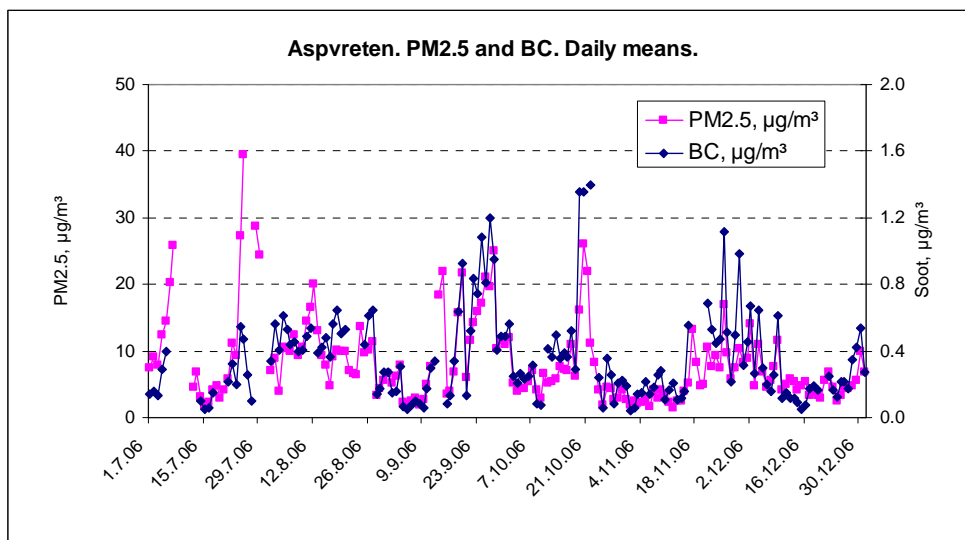


Figure 5: BC and  $\text{PM}_{2.5}$  concentrations at Aspvreten, a regional background site in mid Sweden, during the last 6 months of 2006.

#### 4.2.5 Conclusions and recommendations

The PSAP-ITM has shown to have the similar performance compared to a commercial aethalometer. The instrument works in very different environments, from heavily polluted kerbside sites to clean remote sites as the Arctic or in marine environments on the Danish research expedition Galathea. The PSAP-ITM is now in continuous use at several background site, regional as urban and at some kerbside sites.

#### 4.2.6 References and reports

- Andersen, P. (2006): Measurement of particulate carbon in relation to traffic and validation of a Particulate Soot Absorption Photometer (PSAP). MSc thesis. University of Southern Denmark and National Environmental Research Institute, Roskilde, Denmark.
- Glasius, M., Merete Bilde, Matthias Ketzel, Sara Pryor, Thomas Kristensen, Johan Ström and Liselotte G. Sørensen. (2007): Global marine aerosol studies on the Galathea 3 expedition. 8th Informal Conference on Atmospheric and Molecular Science, Helsingør, Denmark, 8-10 June 2007.
- Ketzel, M., Glasius, M., Wählin, P., Jensen, B., Andersen, P., Ström, J., Hansson, H.C. & Palmgren, F.: A simple optical soot sensor - laboratory tests and first field data on wood stove emissions. Nordic Society for Aerosol Research (NOSA), Göteborg, Sweden, 3-4 November 2005.
- Krecl, P., Ström, J., Johansson, C., (2007): Carbon content of atmospheric aerosols in a residential area during the wood combustion season in Sweden. Accepted for publication in *Atm. Env.*
- Johansson, C. and HC Hansson, PM10 och sot i Sverige, ITM rapport 165, 2007.

### 4.3 Particle database NORDIC

#### Objectives:

The final purpose of this model exercise is to establish high quality emission factors for PM<sub>10</sub>, PM<sub>2.5</sub> and UFP, taking into account their dependence on meteorological and street related parameters. These factors may later be used for regional modelling purposes.

For that purpose a common database NORDIC has been created.

#### Summary

A major concern in modelling PM, number or size distribution of particles in any environment is a sufficiently accurate emission data base. It has been concluded in several studies and reviews that present particle data bases lack considerably in accuracy, especially concerning particle number but also in PM.

A Nordic database on urban particle data has been set up for four cities (Stockholm, Copenhagen, Helsinki and Oslo), containing matching simultaneous regional background, urban background and kerbside measurements. Measurements of total particle number (size range 3-900 nm), PM<sub>10</sub> and NO<sub>x</sub> performed simultaneously at urban, near-city and rural locations are used to investigate the contributions from urban versus regional sources of particle number and mass concentration.

#### 4.3.1 Background

At the Copenhagen meeting (March 2004), it was decided that the Nordic model exercise during 2004-2005 should focus on the street and urban environment. Good data sets from street canyons and open roads from the Nordic countries should be commonly used in different model implementations. One main purpose of the project is to establish high quality emission factors for PM<sub>10</sub>, PM<sub>2.5</sub> and total particle number, taking into account their dependence on meteorological and street related parameters. The emission factors together with the understanding on how the emissions are modified when transported within the source area is needed for the regional modelling.

For that purpose a common database NORDIC has been created. The database presently consists of checked and complete PM data sets from street canyons and open roads in Stockholm, Helsinki, Oslo and Copenhagen. Complete means that except for PM<sub>10</sub>/PM<sub>2.5</sub> there is also NO<sub>x</sub> data, in order to use the tracer method. Urban background and rural background data are also given, so the local contribution can be analysed. Traffic counts – whenever available - and meteorological data are also stored.



### 4.3.2 Database overview (format, login, get started)

The database includes an Internet tool for time series analysis, which means that all data can be compared and analysed through the web (downloading of the raw data or elaborated variables is of course possible). Data are loaded in local time (including the summer time shift) with 1 hour time resolution and the time stamp indicating the end of the averaged hour (e.g. “1400” means average between 1300 and 1400). PM10/2.5 measurements by TEOM are provided as “direct readings” without any factor or correction for losses due to evaporation of volatile compounds. The units for all compounds in the database are  $\mu\text{g}/\text{m}^3$ .

The link to the NORDIC database is found via <http://www.luftkvalitet.se>, clicking on “NORDIC” in the blue coloured top menu. The username and password for each NORPAC group has been distributed by Email. The database will be open for interested users for research purposes providing proper referencing of the data source and information of the data providers about the planned use of data.

All settings and selection can be saved under 'macro' in one personal directory per user.

All users can see and use macros (= plots, analysis etc.) of other users.

A detailed technical manual to the graphic interface (IAirviro) can be downloaded from <http://www.luftkvalitet.se>, FORSKNING “NORDIC användarmanual”.

### 4.3.3 Database content

There are four Nordic datasets from street measurements:

#### **STO\_street (Stockholm)**

Period: 2002 – 2004

Stations:- Hornsgatan – street canyon  
- Rosenlundsgatan – urban background PM  
- Torkel Kn.20 – urban background NO<sub>x</sub>  
- Asprvreten – regional background  
- Maria Pol – meteorological station

#### **COP\_street (Copenhagen)**

Period: 2002 – 2004

Stations:- Jagtvej – street canyon  
- HC Andersens Boulevard – street canyon  
- HC Orsted Institute – urban background and meteorology

#### **HEL\_street (Helsinki)**

Period: 2003 – 2004

Stations:- Runeberg – street canyon  
- Vallila – open road  
- Kallio 2 – urban background  
- Kaisaniemi – urban background  
- Luukki – rural background  
- Runeberg – meteorology (urban)  
- Kaisaniemi – meteorology (urban)  
- Helsinki-Vantaa – meteorology (rural)

#### **OSL\_street (Oslo)**

Period: 2003

Stations:- Kirkeveien – street canyon  
- Loren – open road  
- Skoyen – urban background  
- Bakgrunn – rural background  
- Valle Hovin – meteorology

### 4.3.4 General features

From Figure 1, which displays the overall PM<sub>10</sub> levels registered during 2003 in the Nordic street canyons, it is clearly demonstrated that:

- Stockholm and Helsinki have very similar (springtime) peak PM<sub>10</sub> levels
- Copenhagen PM<sub>10</sub> peak levels are much lower
- During more than half of the year (mostly summertime), PM<sub>10</sub> levels are very similar in the Nordic countries, HC Andersens Boulevard having slightly higher levels than the other.

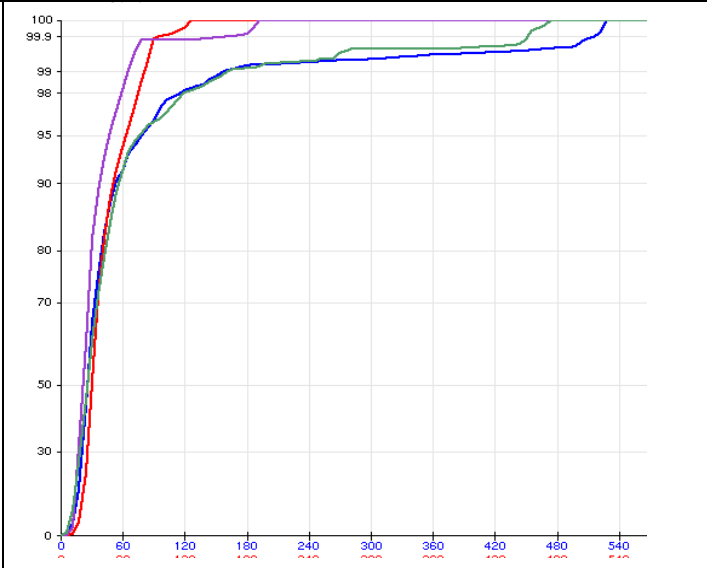
**Figure 1**

Accumulated frequency plots of PM<sub>10</sub> data from four street canyons. PM<sub>10</sub> measured with TEOM and no correction factor was applied

Running 24 hour averages.

Period: 2003

— Hornsg. part, PM-10, scN[M], Value (ug/m3)  
 m=34.1 s=39.5 min=5.4 max=524.2 n=8150  
 — HC Andersen (HCAB), PM-10, scN[M], Value (ug/m3)  
 m=33.2 s=13.7 min=10.1 max=123.4 n=8678  
 — Jagtvej (JGTV), PM-10, scE[M], Value (ug/m3)  
 m=24.2 s=12.1 min=6.0 max=187.4 n=8733  
 — Runeberg Street, PM-10, scE[M], Value (ug/m3)  
 m=33.7 s=35.4 min=3.3 max=468.6 n=7373



Another way of comparing the PM<sub>10</sub> levels in the 4 Nordic data sets contained in the database is shown in Figure 2 illustrating the annual variation at the different locations.

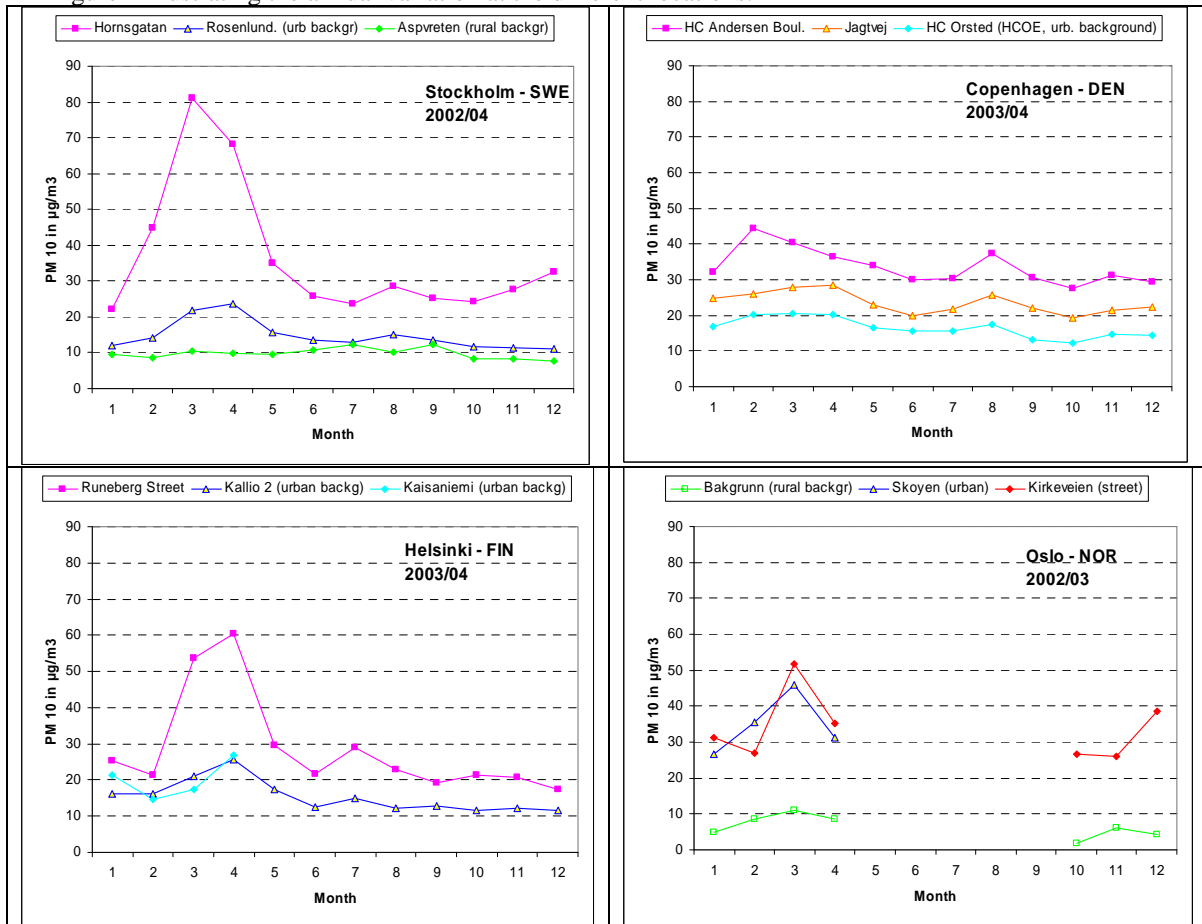


Figure 2: Annual variation of PM<sub>10</sub> at street and background stations in Stockholm, Copenhagen, Helsinki and Oslo. Measurements are performed with TEOM instruments that are operated at 50°C and show values of about 10µg/m<sup>3</sup> less PM<sub>10</sub> than the reference method due to evaporation of volatile compounds (mostly ammonium nitrate from long range transport).

### 4.3.5 Conclusions and recommendations

A Nordic database on urban particle data has been set up for four cities (Stockholm, Copenhagen, Helsinki and Oslo), containing matching simultaneous regional background, urban background and kerbside measurements. Measurements of  $PM_{10}$ ,  $PM_{2.5}$ ,  $NO_x$  and particle number concentration (size range 3-900 nm) performed simultaneously at urban, near-city and rural locations can now be used e.g. to investigate the contributions from urban versus regional sources of particle number and mass concentration as well as estimation of traffic emission factors.

### 4.3.6 References and reports

NORDIC data base: [www.luftkvalitet.se](http://www.luftkvalitet.se) and click on *NORDIC* in the top menu or <http://www.slb.nu/airviro/> Contact for Log-in details: lars.gidhagen@smhi.se

Gidhagen, L. Christer Johansson, Lars Törnquist (2004): NORDIC - A database for evaluation of dispersion models on the local, urban and regional scale. SMHI report, September 2004. (Download via [www.luftkvalitet.se](http://www.luftkvalitet.se))

Backström, H. (2003): Airviro User's Reference. Vers. P2.4-1. SMHI report, Nov. 2003. (Download via [www.luftkvalitet.se](http://www.luftkvalitet.se))

## 4.4 Evaluation and Inter-comparison of Open Road Line Source Models currently being used in the Nordic Countries

### Objective

The aim of this study is to inter-compare and evaluate operational open road line source models (ORLS), currently in use in the Nordic countries of Finland, Denmark, Norway, and Sweden. Datasets from four measurement campaigns, from each of the mentioned countries, will be distributed to the institutes involved and used for the inter-comparison. As the main objective is to assess the quality and variability of the transport and dispersion models, we will be able to determine under which conditions the models perform well or perform poorly. These conditions include wind speed and direction, atmospheric stability and inversion height, road geometry, traffic volume, traffic flow, and levels of background concentrations. In addition, less well defined parameters or methodologies for which the models are most sensitive are also to be identified, such as traffic-induced turbulence, Lagrangian time scales, meteorological pre-processing, and numerical integration methods.

### Summary

There are five ORLS models frequently used in the Nordic countries; these are CAR-FMI, OML, WORM, Dispersion\_ROAD, and HIWAY 2. The institutes responsible for these models are FMI (Finland), DMU (Denmark), NILU (Norway), SMHI (Sweden), and EPA (USA), respectively. These five models will be applied to the four different datasets from four measurement campaigns in the mentioned Nordic countries, and each institute will be responsible for their own dataset. In addition to the WORM model, NILU will also be responsible for the HIWAY 2 model.

The Danish and the Norwegian datasets have been made available for the study, and calculations using the OML and WORM models have been performed. Although PM is the major topic of interest in NORPAC,  $NO_x$  is the primary compound for this study since  $NO_x$  emissions are better defined than for  $PM_{10}$  and  $NO_x$  as inert tracer is best suited for validation of a dispersion model. The aim of this study is to assess the quality and variability of the dispersion models rather than the emissions themselves.

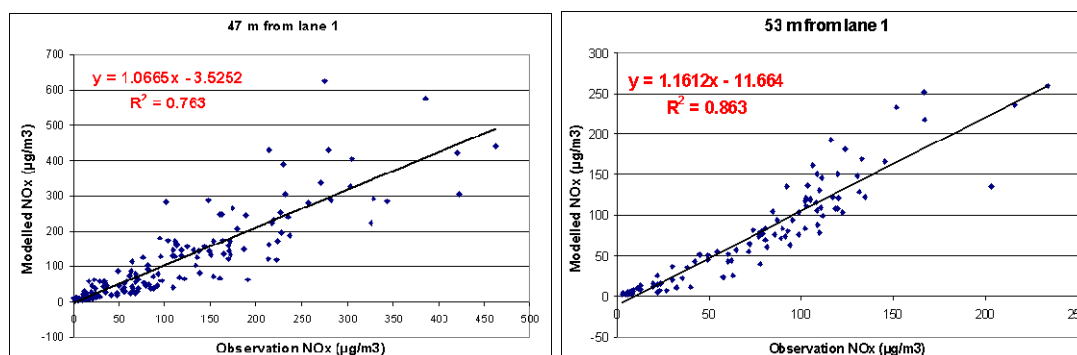
Results show, for example, that WORM performs well on both datasets for higher wind speed conditions. However, low wind speeds tend to significant positive biases in the model. In addition, it has been shown that the use of improved numerical integration

techniques provides better results for winds blowing near parallel to the road. For the WORM model, it has also been shown that best results are achieved when long Lagrangian time scales are used. Further results of the intercomparison will be assessed and discussed.

#### 4.4.1 Results

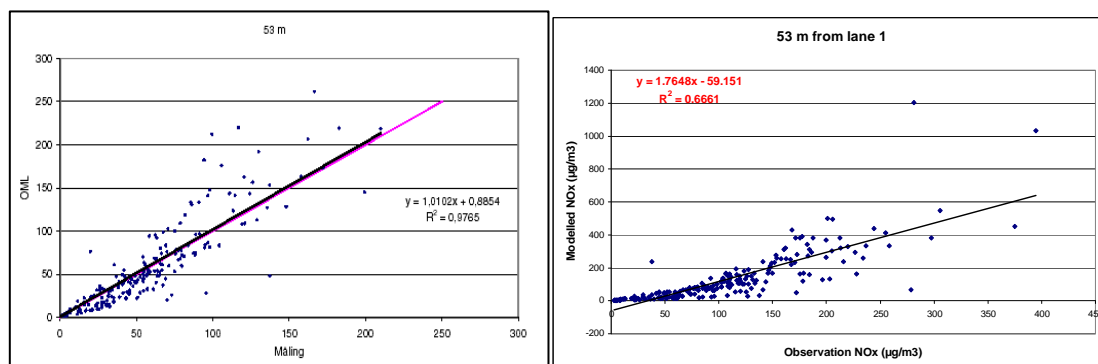
The Norwegian dataset is from a measurement campaign at an open road at Nordbysletta (NBS), outside Oslo, which was conducted 1 January – 15 April 2002. The Danish data set is from a campaign at Køge Bugt highway south of Copenhagen, which was performed 17 September – 18 December 2003. Both sites have three measurement stations on one side of the road, and one background station on the other side.

Figure 1 shows WORM applied to the NBS data and Danish data for the station that is situated approximately 50 m from the road. The data filter used to select data is described in the figure caption. The results are good for both datasets, as the correlation lies above 0.7 and the slope is close to 1 (similar results are obtained for the other stations). The filter is applied chiefly to match similar runs made with the OML model. Thereby, it is easier to compare the models in terms of the calculation of the transport and the dispersion, which is of major interest, without considering the effect of different filters.



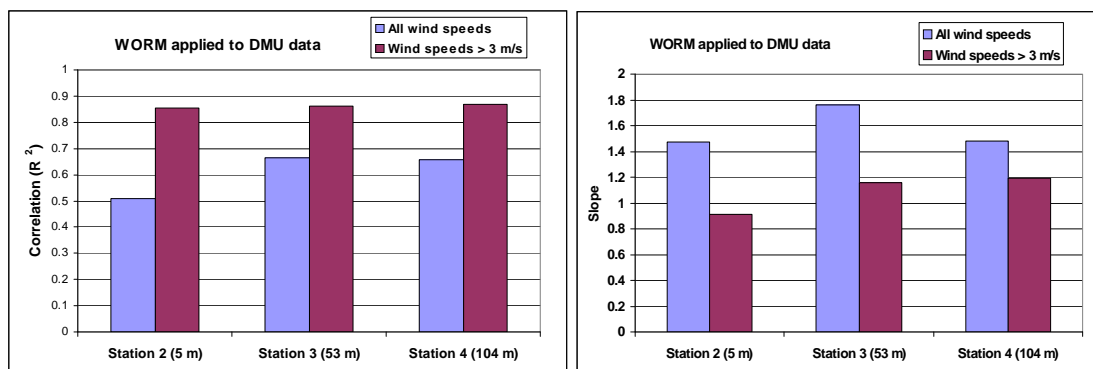
**Figure 1:** Scatter plot of modelled versus observed concentrations of  $\text{NO}_x$  for WORM applied to the Norwegian (NBS) data (left) and the Danish data (right). For both, only data where the wind direction is perpendicular to the road  $\pm 30$  degrees are used. For the Norwegian (NBS) data, only data for which wind speeds  $> 0.5$  m/s are included; for the Danish data, only data for which wind speeds  $> 3.0$  m/s are included.

Figure 2 shows the OML and the WORM model applied to the Danish dataset. Both models perform well.



**Figure 2:** Scatter plot of modelled versus observed concentrations of  $\text{NO}_x$  for OML applied to the Danish data (left) and WORM applied to the Danish data (right). For both, only data where the wind direction is perpendicular to the road  $\pm 30$  degrees are used. For OML and WORM, data for all wind speeds are used.

There are some parameters that need to be studied in greater depth. Figure 3 shows correlation and slope, respectively, using different wind regimes, for WORM as applied to the Danish data (further details are as described in the caption). It is evident that the results are better when data for wind speeds below 3 m/s are excluded. When they are included, the resulting concentrations are strongly overestimated. However, this is not the case for the OML model, which uses all wind speeds, as mentioned under figure 2. This may be due to the fact that WORM model uses only the initial turbulent intensity to describe the turbulence, whereas OML also has traffic produced turbulence implemented.



**Figure 3:** Correlation (left) and slope (right) between modelled and observed  $NO_x$  concentrations for WORM applied to the Danish data for three stations. The blue column represents results when all wind speeds are included; the red column represents results as data for wind speeds below 3 m/s are excluded. For both, only data where the wind direction is perpendicular to the road +/- 30 degrees are used.

#### 4.4.2 Conclusion

Up to now two open road line source models, OML and WORM, have been compared and evaluated based on their application to two datasets from measurement campaigns in Denmark and Norway. In general it can be seen that both models perform quite well on the Danish data set with the WORM model overestimating the concentrations the most by an average of around 12%. Both models, on the other hand underestimate concentrations for the Norwegian dataset.

The significance of the additional turbulence produced by the traffic itself (TPT) on the dispersion could be demonstrated by model runs using the OML model. Results for calculations without TPT indicate a significant increase in model concentrations, by a factor of 2 or more compared to the case when TPT is included.

The study demonstrates the usefulness of comparing models on differing datasets and will lead to improved and more robust models in the future

The current status of the inter-comparison can be seen in table 1 below. It is the intention to continue this study into the future independent from the NORPAC project including the other datasets and models.

**Table 1:** Status of the NORPAC Intercomparison study showing data availability and calculations made by each model on each dataset.

Institute			NILU	NERI	FMI	SMHI
		<b>Dataset</b>	Nordby-Sletta	Køge Bugt highway	Elimäki	??
		<b>Dataset availability</b>	Available	Available	Available	Not yet available
	<b>Model</b>					
NILU	WORM		Calculations made	Calculations made	Calculations in progress	
NERI	OML		Calculations made	Calculations made		
FMI	CAR-FMI				Calculations made	
SMHI	Dispersion-Road					Calculations made
NILU	Highway2					

#### 4.4.3 References and reports:

- Berger, Jane, Sam-Erik Walker, Bruce Denby, Per Løfstrøm, Matthias Ketzel, Ruwim Berkowicz (2007a): Evaluation and inter-comparison of open road line source models currently being used in the Nordic countries- A NORPAC project. Presentation at the Urban Air Quality Conference UAQ-6, Cyprus, 27-29 March 2007.
- Berger, Jane, Ruwim Berkowicz, Sam-Erik Walker, Bruce Denby, Per Løfstrøm, Matthias Ketzel, Jaakko Kukkonen (2007b): Evaluation and inter-comparison of open road line source models currently being used in the Nordic countries- A NORPAC project. Proceedings of the 11<sup>th</sup> International Conference on Harmonisation within Atmospheric Dispersion Modelling, Cambridge, UK, 2-5 July 2007. pp.119-123.
- Jensen, S. S., Løfstrøm, P., Berkowicz, R., Olesen, H. R., Frydendal, J., Fuglsang, K., Hummelshøj, P., 2004 Air quality along motorways – Measurement campaign and model calculations. National Environmental Research Institute, Denmark, 67 p. – NERI report No. 522. <http://fagligerapporter.dmu.dk> (In Danish).
- Härkönen, J., Valkonen, E., Kukkonen, J., Rantakrans, E., Lahtinen, K., Karppinen, A., Jalkanen, L. 1996. A model for the dispersion of pollution from a road network. Finnish Meteorological Institute, Publ. on Air Quality 23, Helsinki.
- Kukkonen, Jaakko, Jari Härkönen, Jari Walden, Ari Karppinen and Kaisa Lusa, 2001. Evaluation of the dispersion model CAR-FMI against data from a measurement campaign near a major road. Atmos. Environ. 35-5, pp. 949-960.
- Levitin J, Härkönen J, Kukkonen J and Nikmo J, 2005. Evaluation of the CALINE4 and CAR-FMI models against measurements near a major road. Atmos. Environ., 39 (2005) 4439–4452.
- Öttl, D., J. Kukkonen, R.A. Almbauer, P.J. Sturm, M. Pohjola and J. Härkönen, 2001. Evaluation of a Gaussian and a Lagrangian model against a roadside dataset, with focus on low wind speed conditions. Atmos. Environ. 35, pp. 2123-2132.
- Walker, S.E. 2006. Air4EU case study D 7.1.4. Data assimilation in open road line source modelling. [www.air4eu.nl/reports\\_products.html](http://www.air4eu.nl/reports_products.html)

## 4.5 Aerosol dynamics modelling exercise

### Objectives:

This activity was aimed at getting a common Nordic understanding of the following questions:

1. Which aerosol dynamic processes are important in various spatial scales? What can we learn from measurements, time scale analysis and model studies?
2. Can we model the aerosol dynamics in order to predict total particle number concentration and size distribution?
3. Can we define a test case to evaluate the performance concerning aerosol dynamics in different models?

### Summary

An Aerosol Modelling Workshop was organized and a common understanding was reached concerning the importance of aerosol dynamical processes both for urban and regional scale and separated for particle mass and particle number concentrations.

In common for all scales it was agreed that emission and dilution are very crucial processes and have to be modelled with detail before aerosol dynamics can be considered. Major uncertainties still exist regarding the emission inventories for ultrafine particle number emissions despite the progress in getting consistent vehicle emission factors.

For urban scale air pollution modelling particle dynamics can be neglected concerning the total particle mass. Also for particle number concentration the dilution is totally dominating the concentrations, even though it can be observed that the particle number emissions can double when decreasing temperature from +20 to +5 °C due to particle formation in the immediate exhaust plume. Dry deposition might be strongly enhanced due the traffic motion and can reduce the total number with about 20% in a street canyon. The dilution dominates over the aerosol dynamical processes as implied by the very high correlation observed between particle number concentrations at all size classes and NO<sub>x</sub>, as the latter pollutant is inert in street scale. However, aerosol processes can be important in case of very high concentrations, and in the dispersion regime in the immediate vicinity of the tailpipe. Aerosol processes can also change the size distribution of particles in the ultrafine particle size range.

At regional scale aerosol dynamic processes, as nucleation and coagulation to make a relatively little effect on the calculated concentrations of **PM<sub>2,5</sub>** and **PM<sub>10</sub>**. Condensation may be of major importance for the organic fraction of the aerosol. Dry deposition is the dominating removal process for coarse particles, but is less important for fine particles. Wet scavenging is the most important particle sink for fine particles, depending on fine particles acts as cloud condensation nuclei's. In the cloud droplets a crucial aerosol formation process occur as absorption of water soluble gases and subsequent cloud water chemistry forms new non-volatile compounds as e.g. inorganic compounds.

With respect to **particle number**, the comparison of model results with Nordic measurements shows that adequate model description of nucleation and further particle growth can be important for accurate calculation of Aitken particle number (which determines the total number). Dry deposition is the dominating removal process. Still, the largest uncertainty in modelled particle numbers is related to particle emissions. It should be noted however that not all processes are included in the aerosol dynamic description in the models. The cloud processes, one of the major secondary formation processes and the major sink process, is not included.

### 4.5.1 Background

Particle dynamics of vehicle exhaust in street and urban environment has been recently investigated by several groups participating in NORPAC (DMU, SMHI, FMI, UHEL) with different kind of dispersion and aerosol dynamic models. During discussions in the NORPAC meetings in Copenhagen, March 2004 and Stockholm, November 2004 it was decided to arrange a workshop in order to facilitate a comparison between the approaches used by these groups. The workshop was organised and held on

April 21, 2005 in Helsinki. Focus of the workshop was the aerosol dynamics of ultrafine particles at local (street), urban and regional level.

#### 4.5.2 Outcome of the Helsinki workshop

The discussion about the relevance of the different aerosol dynamics and other processes is separated in the local/urban and regional scale. All processes are discussed: condensation, coagulation, dry deposition, wet deposition, nucleation, dilution and emission.

In common for all scales it was agreed that emission and dilution are very significant processes and have to be modelled with detail. Major uncertainties still exist regarding the emission inventories for ultrafine particle number emissions despite the progress in getting consistent vehicle emission factors.

The presentations and discussion during the workshop with respect to the importance of various aerosol dynamic processes at different scales are summarised in Table 1 (taken from Ketzel et al. 2005) and are discussed in the following.

Local and urban scale:

Based on the Ph. D. thesis's of Lars Gidhagen, Matthias Ketzel and Mia Pohjola, the following conclusions can be drawn. Particle dynamics can be neglected for most of the practical air pollution modelling assessments concerning the total particle number concentration and particle mass at street scale. However, aerosol processes can be important in case of very high concentrations, and in the dispersion regime in the immediate vicinity of the tailpipe. Aerosol processes can also change the size distribution of particles in the ultrafine particle size range.

This is more certain for coagulation and deposition, and less certain for condensation / evaporation, since a large gap of knowledge exists in both composition measurements for the organic fraction of the ultrafine particles and gas phase measurements of condensable species in traffic exhaust. In addition, large-scale nucleation events were not regarded at local scale. The importance of such particles for health effects has not yet been investigated. Nucleation plays probably a role during the rapid dilution in the plume directly after the exhaust pipe. This initial nucleation process is likely to depend on the ambient temperature and might be responsible for the observed temperature dependence of the emission factors.

A method for coupling aerosol process models and a roadside dispersion model has been developed for the simulation of aerosol processes for the dispersion of traffic exhaust particulate matter in local scale. The modelling systems containing the aerosol process models MONO32 (UH/Stadia) and UHMA (UH), both of these coupled with the roadside dispersion model CAR-FMI (Hussein, 2007, Pohjola 2007). The predicted results have been evaluated against two datasets from field measurement campaigns; the first campaign has been conducted using a mobile laboratory in Helsinki (Pirjola et al., 2004; 2006), and the second using a stationary site.

Regional scale:

At regional scale aerosol dynamic processes, as nucleation and coagulation to make a relatively little effect on the calculated concentrations of **PM<sub>2.5</sub>** and **PM<sub>10</sub>**. Condensation may be of major importance for the organic fraction of the aerosol. Dry deposition is the dominating removal process for coarse particles, but is less important for fine particles. Wet scavenging is the most important particle sink for fine particles, depending on fine particles acts as cloud condensation nuclei's. In the cloud droplets a crucial aerosol formation process occur as absorption of water soluble gases and subsequent cloud water chemistry forms new non-volatile compounds as e.g. inorganic compounds.

With respect to **particle number**, the comparison of model results with Nordic measurements shows that adequate model description of nucleation and further particle growth can be important for accurate calculation of Aitken particle number (which determines the total number). Dry deposition is the dominating removal process. Still, the largest uncertainty in modelled particle numbers is related to particle emissions. It should be noted however that not all processes are included in the aerosol dynamic description in the models. The cloud processes, one of the major secondary formation processes and the major sink process, is not included in the models discussed.



**Table 1:** Summary of the presentations and discussion during the workshop with respect to the importance of various aerosol dynamic processes at different scales. The legend below the table explains the used symbols, references are listed below. The values in percent give the estimated change in particle number (ToN) or particle mass (PM) that is caused by the given process(es).

scale	Local				
case study / REF	tunnel, rush hour (1)	Street canyon (2)	open road (3)	Time scale analysis (4)	open road (5)
Process \ ToN or PM	ToN	ToN	ToN	ToN	ToN
condensation	n.s.	n.s.	n.s.	shift to larger pt. diameters, no loss	x (p.i.)
coagulation	80% loss for part < 10nm 40% loss for part 10-29 nm	<15% loss (<30% for low w.s.)	÷	÷ (except for road tunnels)	÷
dry deposition			10%		n.s.
wet deposition	÷	÷	÷	n.s.	n.s.
nucleation	(s,td)	(s,td)	(s,td)	(s,td)	(s,td)
Dilution	X!	X!	X!	X!	X!
Emission	x	X!	X!	X!	X!

**Legend:**

- ÷ not important
- x important
- X! very important / dominating
- n.s. not studied
- (s,td) important as source term directly after the vehicle exhaust pipe, later probably not, probably responsible for the observed temperature dependence of emissions
- w.s. wind speed
- (p.i.) possibly important, but data for the condensing vapour are not available
- ToN Total particle number
- PM Particle mass

**Table 1 cont.:**

scale	urban			regional		
case study / REF	Stockholm (6)	Copenhagen (7)	Helsinki (10-13)	EMEP (8)	EMEP (8)	FMI (9)
process \ ToN or PM	ToN	ToN	ToN	ToN	PM	PM
condensation	n.s.	Minor shift to larger pt. sizes	p.i.	n.s.	÷ 5-7 % (diff. between aer. dyn. and mass model)	÷
coagulation	<5% (<10% for low w.s.)	15 % - 30% loss	p.i.	n.s.	X! – coarse particles x – fine part.	÷
dry deposition	<25% (<50% for low w.s.)		x	X!		
wet deposition	÷	n.s.	x		X!	X!
nucleation			X!	x	÷	÷
Dilution	x	X	x	x	X	x
Emission	X!	X!	X!	X!	X!	X!

**4.5.3 Aerosol dynamics model inter-comparison**

As one further outcomes of the workshop, an aerosol dynamics model inter-comparison was conducted aiming at the evaluation of the process parameterisation towards a harmonisation between the Nordic modelling groups.

A test case on urban scale was designed, i.e. Copenhagen area including a westerly located station (Lille Valby). This test case was modelled with AEROFOR (Pirjola, 1999; Pirjola and Kulmala, 2001)

and DMU's Multi-plume Aerosol dynamics and Transport (MAT) model (Ketzel and Berkowicz, 2005).

The inter-comparison included the processes: emission from a near ground source, dilution with background air, deposition, coagulation and condensation.

The simulation starts at time=0s with a background distribution (measured at Lille Valby) given as sum of 3 log normal modes. The air mass is then transported over an (urban traffic) area source with a constant wind speed. The emission density of the homogeneous area source and the size distribution of the emitted particles are prescribed. The evolution of the size distribution will be followed for a certain simulation time. Model runs are performed with sensitivity analysis for specific processes included or excluded in the simulations. Results are shown in Figure 1 indicating a good agreement between both models considering the complexity of the modelled case. Both models treat aerosol dynamics in a similar way. However, due to the implementation of a different parameterisation in the AEROFOR model dry deposition so small that we cannot distinguish the case EmDi (not shown in the graphs) from the case EmDi+dep. This is also the reason why the curves EmDi+coa and EmDi+DeCoa overlap.

Largest deviations between both models are observed for the simulations including treatment of condensation. This is expected since the chemical composition and properties of the aerosol are different in the two models.

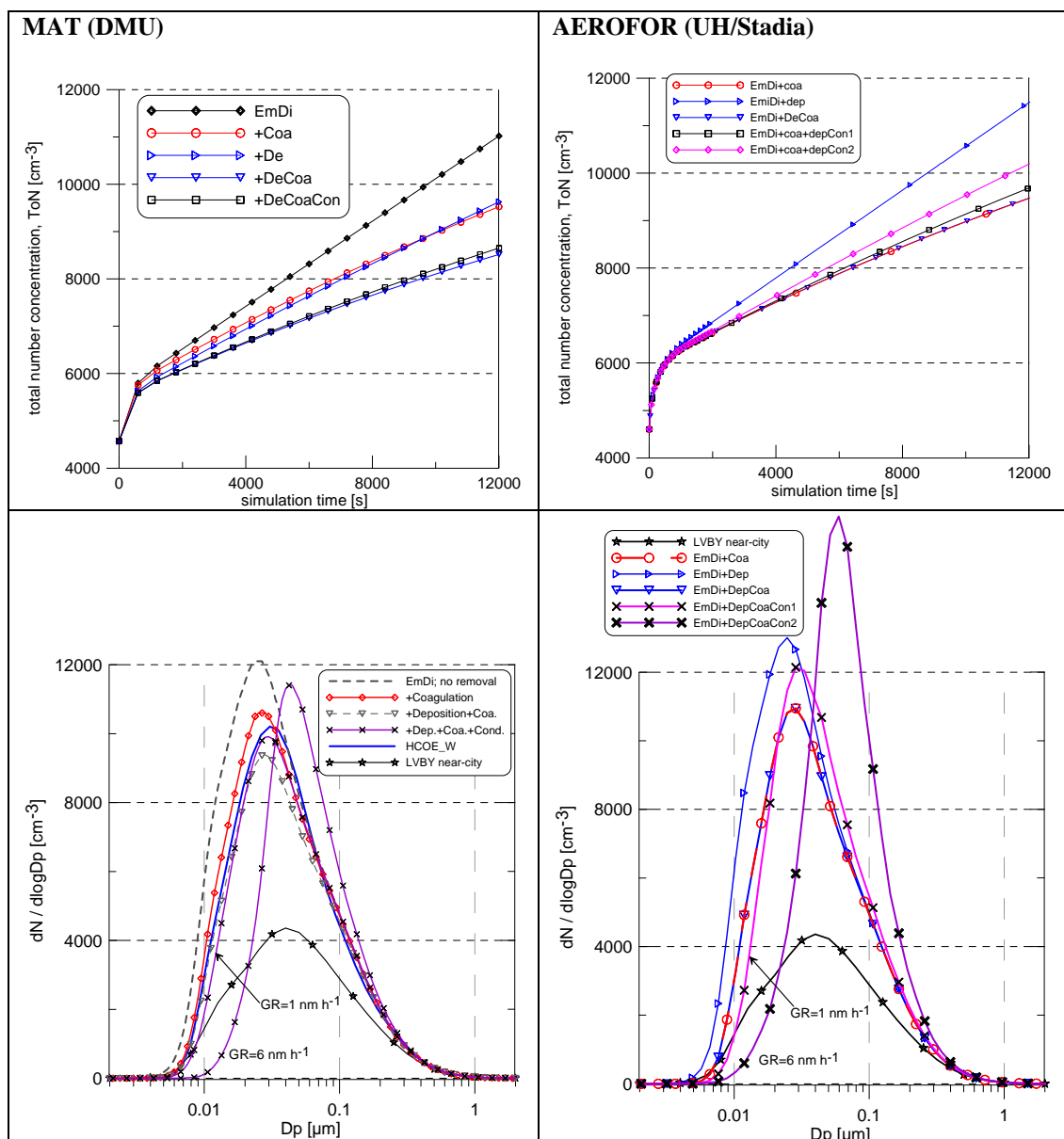


Figure 1: Inter-comparison between model results from MAT (Left) and AEROFOR (Right) for the test case in Copenhagen. Upper graph: Time dependence of the total particle concentration at ground level. Results from 5 simulations including different aerosol dynamics processes.

Lower graph: Size distribution calculated with different particle dynamics processes considered. Two different growth rates (GR=1 or 6 nm h<sup>-1</sup>) considered for the simulation with condensation. For comparison the measured size distributions at LVBY (near-city) and HCOE (urban rooftop). Simulation time 12000 s.

#### 4.5.4 Conclusions and recommendations

An Aerosol Modelling Workshop was organized and a common understanding was reached concerning the importance of aerosol dynamical processes both for urban and regional scale and separated for particle mass and particle number concentrations.

In common for all scales it was agreed that emission and dilution are very crucial processes and have to be modelled with detail before aerosol dynamics can be considered.

Major uncertainties still exist regarding the emission inventories for ultrafine particle number emissions despite the progress in getting consistent vehicle emission factors. Here more effort has to be invested in the future.

Two aerosol dynamics models have been applied to a common test case showing a very similar performance of the models with respect to the influence of aerosol dynamics on shaping the size distribution at urban background level.

The temperature dependence of the particle number emission factors has to be investigated further and should be included in the Nordic models.

Particle dynamics should be incorporated in regional models both for particle mass and especially for particle number.

#### 4.5.5 References and reports

- Hussein, T., J. Kukkonen, H. Korhonen, M. Pohjola, L. Pirjola, D. Wraith, J. Härkönen, K. Teinilä, I.K. Koponen, A. Karppinen, R. Hillamo and M. Kulmala, 2007. Evaluation and modeling of the size fractionated aerosol particle number concentration measurements nearby a major road in Helsinki – Part II: Aerosol measurements within the SAPPHIRE project. *Atmos. Chem. Phys.* 7, 4081–4094
- Ketzel, M. and Berkowicz, R. (2005): Multi-plume aerosol dynamics and transport model for urban scale particle pollution. *Atmospheric Environment* 39, 3407-3420.
- Ketzel, M. et al. (2005): Summary of the NORPAC Workshop on Aerosol Dynamics Modelling in the Nordic Countries on Local, Urban and Regional Scale, 21 April 2005 at FMI, Helsinki. (Download from <http://NORPAC.dmu.dk>)
- Pirjola L. (1999) Effects of the increased UV radiation and biogenic VOC emissions on ultrafine aerosol formation, *J. Aerosol. Sci.*, 30, 355-367.
- Pirjola, L. and Kulmala, M. (2001) Development of particle size and composition distribution with a novel aerosol dynamics model. *Tellus*, 53B, 491-509.
- Pohjola, M., L. Pirjola, A. Karppinen, J. Härkönen, H. Korhonen, T. Hussein, M. Ketzel, and J. Kukkonen, 2007. Evaluation and modelling of the size fractionated aerosol particle number concentration measurements nearby a major road in Helsinki – Part I: Modelling results within the LIPIKA project. *Atmos. Chem. Phys.* 7, 4065-4080.

##### References used in Table1:

1. Gidhagen, L., Johansson, C., Ström, J., Kristensson, A., Swietlicki, E., Pirjola, L. and Hansson, H.-C. (2003): Model simulation of ultrafine particles inside a road tunnel. *Atmospheric Environment* 37, 2023-2036.
2. Gidhagen, L., Johansson, C., Langner, J. and Olivares, G. (2004): Simulation of NO<sub>x</sub> and ultrafine particles in a street canyon in Stockholm, Sweden. *Atmospheric Environment* 38, 2029-2044.
3. Gidhagen, L., Johansson, C., Omstedt, G., Langner, J. and Olivares, G. (2004): Model simulations of NO<sub>x</sub> and ultrafine particles close to a Swedish highway. *Environmental Science & Technology* 38, 6730-6740.
4. Ketzel, M. and Berkowicz, R. (2004): Modelling the fate of ultrafine particles from exhaust pipe to rural background: an analysis of time scales for dilution, coagulation and deposition. *Atmospheric Environment* 38, 2639-2652.
5. Pohjola M A, Pirjola L, Kukkonen J, Karppinen A, Härkönen J, Ketzel M: MODELLING OF DISPERSION AND AEROSOL PROCESSES IN A ROADSIDE ENVIRONMENT, AND

EVALUATION WITH MEASURED DATA. Presentation at the 5th Internatl. Conf. on Urban Air Quality (UAQ 2005), Valencia, Spain, 29-31 March 2005.

6. Gidhagen, L., Johansson, C., Langner, J. and Foltescu, V. L. (2005): Urban scale modeling of particle number concentration in Stockholm. *Atmospheric Environment* 39, 1711-1725.
7. Ketzel, M. and Berkowicz, R. (2005): Multi-plume aerosol dynamics and transport model for urban scale particle pollution. *Atmospheric Environment* 39, In Press,
8. Tsyro, S.(2004) Model assessment of particulate matter in Europe in 2002 .In EMEP Status report 4/2004.Transboundary particulate matter in Europe. Available on <http://www.emep.int>)
9. Sofiev M, P. Siljamo, I. Valkama, M. Ilvonen and J. Kukkonen, 2006. A dispersion modelling system SILAM and its evaluation against ETEX data. *Atmos. Environ.* 40 (2006) 674–685.
10. Hussein, T., Hämeri, K., Kulmala, M., 2002. Long-term indoor-outdoor aerosol measurement in Helsinki, Finland. *Boreal Environment Research* 7, 141-150.
11. Hussein, T., Puustinen, A., Aalto, P. P., Mäkelä, J. M., Hämeri, K., Kulmala, M., 2004. Urban aerosol number size distributions. *Atmospheric Chemistry and Physics* 4, 391-411.
12. Hussein, T., Hämeri, K., Aalto, P. P., Kulmala, M., 2005. Modal structure and spatial-temporal variations of urban and suburban aerosols in Helsinki-Finland. *Atmospheric Environment* 39, 1655-1668.
13. Hussein, T., Hämeri, K., Aalto, P., Asmi, A., Kakko, L., Kulmala, M., 2004. Particle size characterization and indoor-to-outdoor relationship of atmospheric aerosols in Helsinki. *Scandinavian Journal of Work, Environment, and Health* 30 (Supplement 2), 54-62.

## 4.6 Particle emission factors, including non-exhaust emissions

### Objectives:

In this activity we addressed the following questions:

- Are our methods for particle mass (PM) emission estimation good enough for the prediction of air quality standards according to the EU legislation?
- What type of processes do we need to describe in our emission models?
- Is it possible to “harmonise” on a common Nordic emission method at least for summer conditions?

### Summary

A specific Workshop was dedicated to *Modelling of PM emissions in different Nordic traffic environments*. Hereby emission factors for traffic-generated PM during the winter/spring PM maximum and as annual mean could be estimated separately for fine and coarse road dust and road salt, using a source-receptor model based on the elemental composition of the aerosol. The measured emission factors were used to validate the Swedish SMHI PM<sub>10</sub> model. The different models used in Nordic countries consistently indicate that a large part (about 50% - 85% depending on the location) of the total PM<sub>10</sub> emissions originates from non-exhaust emissions. This implies that measures relying on reducing the exhaust part of the vehicle emissions can have a very limited effect on ambient PM<sub>10</sub> levels.

A mobile measurement system has been developed and used in the Stockholm region by ITM to quantify the relative importance for the road particle emission and suspension of accumulated dust versus direct pavement wear, tire type, pavement type and vehicle speed. Highest particle mass concentrations were always observed behind the studded tire and lowest behind the summer tire. This was regardless of the road type and indicates that studded tires lead to higher emissions than friction and summer tires. Studded-to-summer tire ratios were 4.4–17.3 and studded/friction ratios were 2.0–6.4.

Progress has been made in estimating and comparing particle number emission factors between several places in the Nordic countries. Further, it has been shown how the emission factors for the number of particles emerging from engine exhaust particles depend on ambient temperature.

It was also shown that the largest number of particles generated from wear of the road-tire interface is found in the nano- and ultrafine particle size range (< 100 nm) with number emission factors ranging between 1-0.1 % of the tail-pipe exhaust emissions. The conclusion is that even cars with zero-emission engines will emit considerable quantities of nanoparticles.

### 4.6.1 Background

Modelling of Ultra Fine Particles (UFP) and Particle Mass (PM) in the urban environment is important but difficult. The EU legislation (Council Directive 1999/30/EC) regulates PM<sub>10</sub> to protect the health of the population and this requires tighter controls on the sources of PM<sub>10</sub> in order to achieve the limit values. Several cities in the Nordic countries have severe problems meeting the limit values for PM<sub>10</sub>. The Nordic countries have rather similar climate and traffic conditions but also with important differences e.g. the use of studded tyres, climate differences, differences in antiskid treatments. Comparing data and models used in the Nordic countries can therefore improve our understanding of these problems. A comparison between different methods used in some Nordic countries was started at the NORPAC meeting in Stockholm November 2004 and later presented by Ketzel et al., 2005 at the International Conference on Urban Air Quality at Valencia, Spain. This work was continued during a workshop on modelling of PM emissions in different Nordic traffic environments organized

by SMHI in October 2005. A publication on the results of the workshop has been accepted for publication (Ketznel et al. 2007).

The work on emission factors for UFP has started with a short meeting in Helsinki in August between FMI, DMU, Stadia with a discussion of the use the data set from the LIPIKA campaign (2003). Several emission factor estimates have been made across the Nordic countries showing relatively consistent emission factors for the average vehicle fleet. A summary of UFP emission factor estimates, including size distribution and their observed temperature dependence is ongoing at the ITM group, this work is closely related to the aerosol dynamic modelling activity (Chapter 4.5).

#### 4.6.2 Results

Current available emission factor estimates for  $PM_{10}$  and  $PM_{2.5}$  from emission databases and different emission models have been compared and validated against 8 high quality street pollution measurements in Denmark, Sweden, Germany, Finland and Austria.

In Figure 1 the derived PM emission factors the 8 different streets are plotted; separated in summer month and full year for. For the emission factor estimation  $NO_x$  was used as a tracer and the data-set owner provided the  $NO_x$  emission factor.

The observed range of summer time  $PM_{10}$  emission factors is about 80-130 mg/km, with the German streets closer to the lower end of the range and one Danish street HCAB showing much higher emissions of about 150 mg/km. No comprehensive explanation for the elevated values at HCAB has been found (higher HDV share, higher vehicle speed, road conditions and leaf debris from nearby trees). The range of  $PM_{2.5}$  emission factors is 30-60 mg/km, highest again at HCAB and lowest value at a German site MEBG. The exhaust emission factors (estimated by the data set supplier) are also plotted in Figure 1. Also here a wide range of 20-50 mg/km is observed, highest value once more at HCAB and lowest at MEBG, reflecting the different vehicle fleet and models for PM exhaust emissions. Estimated exhaust emissions are very close to the total  $PM_{2.5}$  emissions indicating the relatively smaller contribution from the non-exhaust emission in the fine ( $PM_{2.5}$ ) fraction.

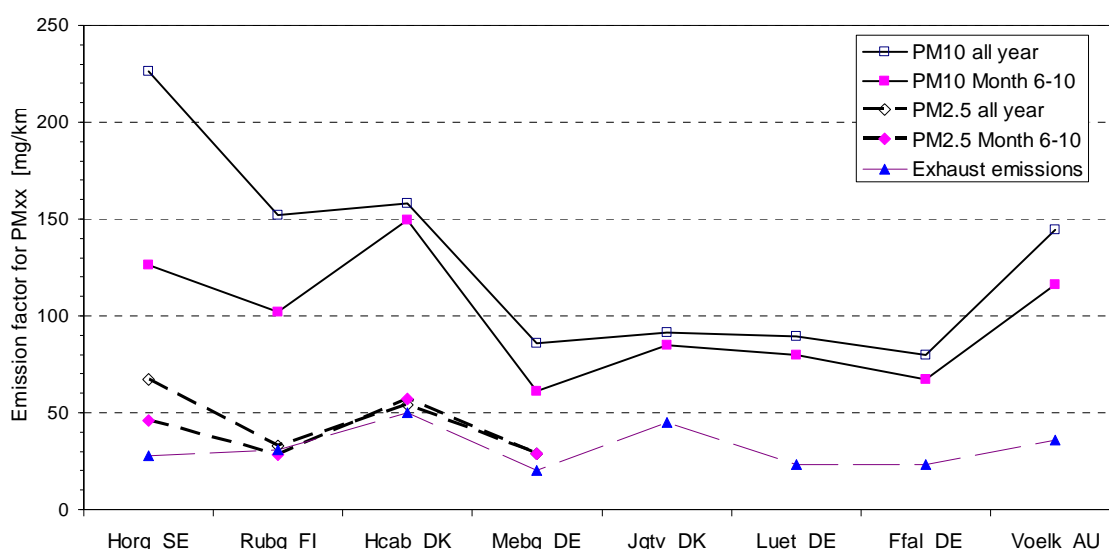


Figure 1:  $PM_{10}$  and  $PM_{2.5}$  emission factors separated for all year and summer month (6-10) for 8 locations in 5 North and Central European countries, estimated by using  $NO_x$  as tracer. The estimated values for exhaust emissions are given as well (Ketznel et al. 2007).

Figure 2 shows a comparison of the three emission factor models with results from measurements and receptor analysis for Swedish conditions and for Danish conditions (HCAB), where the contribution of the non-exhaust part is spitted into re-suspension, tyre brake and salt. All estimates are yearly averages, i.e. including the elevated winter/spring time

re-suspension emissions due to studded tyres in Sweden. This high emissions are well reproduced by the Swedish model.

The Danish method gives higher exhaust emissions compared to the other two methods (see Figure 6), but agrees with the receptor analysis for HCAB (Wåhlin et al., 2006). The reason for the higher Danish emissions could be in the vehicle composition (smaller share of newest cars). The measured total PM<sub>10</sub> emission factors at HCAB are elevated compared to other streets and also relative to the modelled estimates as discussed earlier. For total PM<sub>2.5</sub> emission factors the Danish method agrees well with the measurements.

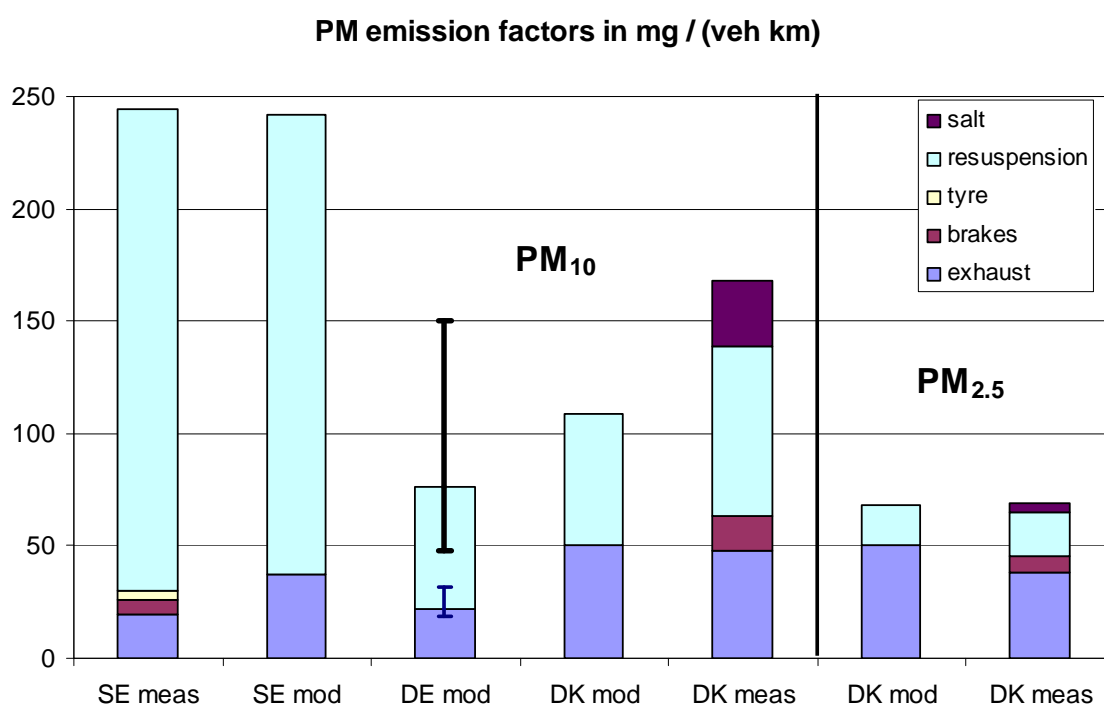


Figure 2: Comparison of the average emission factors for PM<sub>10</sub> / PM<sub>2.5</sub> modelled (marked with “mod”) with three methods (Swedish-SE, German-DE and Danish-DK); and estimates (“meas.”) based on measurements and receptor analysis (taken from Johansson et al., 2006 and Wåhlin et al., 2006). For the model estimates (‘mod’) the re-suspension fraction includes tyre and brake wear. The bars for the ‘German’ method represent the variability due to different urban driving conditions. (Source: Ketznel et al. 2007)

A mobile measurement system has been developed and used in the Stockholm region by ITM to quantify the relative importance for the road particle emission and suspension of accumulated dust versus direct pavement wear, tire type, pavement type and vehicle speed (Hussein, 2007). Measurements were performed during spring (May) and autumn (September) on selected roads with different pavements and traffic conditions in the Stockholm region. Highest particle mass concentrations were always observed behind the studded tire and lowest behind the summer tire. This was regardless of the road type and indicates that studded tires lead to higher emissions than friction and summer tires. Studded-to-summer tire ratios were 4.4–17.3 and studded/friction ratios were 2.0–6.4.

An important dependence on vehicle speed was also observed (Figure 3). During May the particle mass concentrations behind the studded tire at vehicle speed 100 km/h were about 10 times higher compared to 20 km/h. The speed dependence was not so pronounced in September, which could be due to less accumulated dust on the roads or to different pavement conditions compared to May.

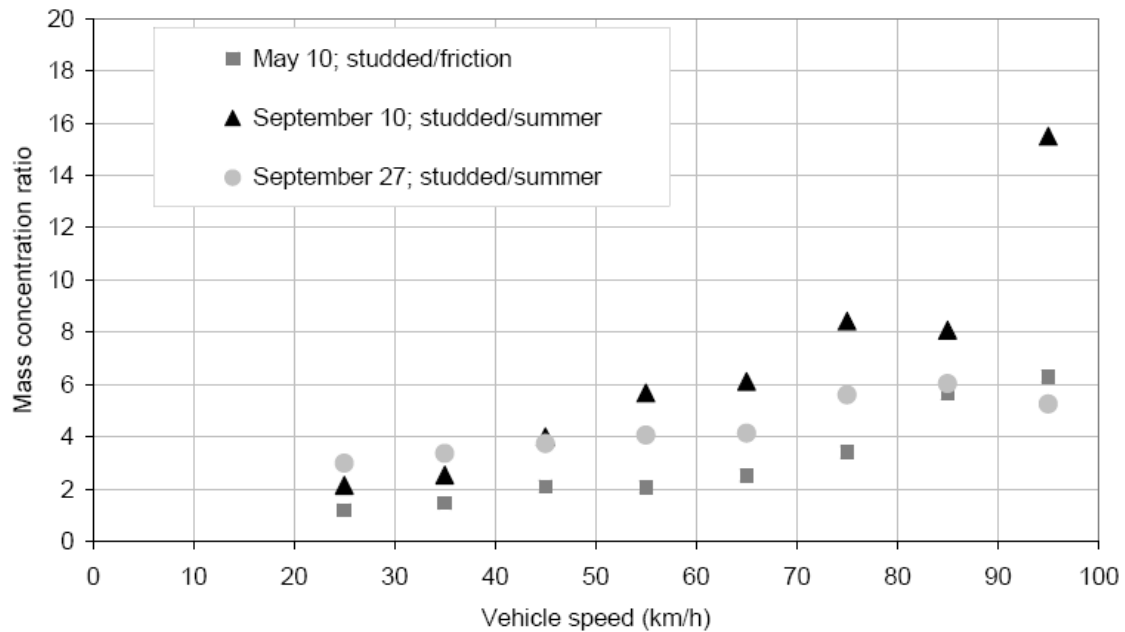


Figure 3: Vehicle speed dependence of the particle mass concentration ratios for studded versus friction and summer tires during May 10<sup>th</sup> and September 10<sup>th</sup> and 27<sup>th</sup>. The presented data is the overall median for all measurement points on different road surfaces. (Source: Hussein et al. 2007)

#### 4.6.3 Conclusions and recommendations

The different PM emission models used in Nordic countries consistently indicate that a large part (about 50% - 85% depending on the location) of the total PM<sub>10</sub> emissions originates from non-exhaust emissions. This implies that measures relying on reducing the exhaust part of the vehicle emissions can have a very limited effect on ambient PM<sub>10</sub> levels. As exhaust emissions continuously decrease due to after-treatment the non-exhaust emissions are gaining more importance.

A method to measure the re-suspension emissions directly on-road has been tested in the field and gives very promising results that will allow to study parameters as e.g. speed dependence of emissions and influence of road and tyre type.

It is necessary to gather more high quality PM data of to investigate and understand the differences and influencing factors at the different locations.

#### 4.6.4 References and reports

- Hussein, T., Johansson, C., Karlsson, H., Hansson, H.C. (2007): Factors affecting particle emissions from paved roads: on road measurements in Stockholm, Sweden. Accepted for publication in *Atmospheric Environment*.
- Johansson, C., Norman, M., and Gidhagen, L., 2006: Spatial & temporal variations of particle mass (PM<sub>10</sub>) and particle number in urban air – Implications for health impact assessment. *Environ. Monitoring and Assessment*, Published online. (<http://dx.doi.org/10.1007/s10661-006-9296-4>)
- Ketzel, M., G. Omstedt, C. Johansson, I. Düring, M. Pohjola, D. Oettl, L. Gidhagen, P. Wählin, A. Lohmeyer & R. Berkowicz (2007): Estimation and validation of PM<sub>2.5</sub>/PM<sub>10</sub> exhaust and non-exhaust emission factors for practical street pollution modelling, Submitted for publication in *Atmospheric Environment*.
- Omstedt, G., Bringfelt, B. and Johansson, C. 2005: A model for vehicle-induced non-tailpipe emissions of particles along Swedish roads. *Atmospheric Environment* 39, 6088-6097.
- Wählin, P., Berkowicz, R. and Palmgren, F., 2006: Characterisation of traffic-generated particulate matter in Copenhagen. *Atmospheric Environment* 40, 2151-2159.



## 4.7 OC/EC parameterisation and Implementation in the EMEP model

### Objectives:

To implement a better description of elemental and organic carbon (EC / OC) in the EMEP Unified Eulerian model, improving its capability to address sources, concentrations in the atmosphere, transformation and deposition.

### Summary

A new and more detailed emission inventory for EC and OC has been implemented in the EMEP model, with explicit incorporation of wood-burning emissions. Comparison of model runs with observations from the EMEP OC/EC campaign, and the CARBOSOL campaign, suggest that the EMEP model reproduces the primary components of particulate carbonaceous matter (PCM=EC+OC) to a reasonable degree. Model results for total PCM show good performance at the Nordic sites. However, calculations at more southerly sites show much significant underpredictions, mainly related to problems with secondary organic aerosol (SOA). Such underpredictions are typical for even the most advanced models today. Despite these uncertainties, model results suggest that biogenic volatile organic compounds (BVOC) dominate SOA formation, and this highlights the need for a proper evaluation of emissions from this source.

Monitoring of the particle size distribution at the Nordic background super sites has been used to estimate the particle formation of BVOC over the boreal forest. The result agree well with the EMEP model. Further a box model with full aerosol dynamical process description has shown to agree relatively very well with observed particle size distribution. This work must be considered a scientific break through as it so clearly describe and quantify the influence by the boreal forest on the atmosphere. A part of the work was published in Science.

### 4.7.1 Introduction

The main modelling activities within NORPAC have involved testing different model formulations against the results of two measurement campaigns which were conducted during 2002-2004.

The EMEP EC/OC campaign measured EC, OC, TC and PM at 15 sites, and many of these sites have complementary measurements of other species. In addition, some of the samples from the EMEP campaign have been analysed for levoglucosan. Details of the EMEP EC/OC campaign have been reported previously (Yttri and Kahnert, 2004; Yttri and Facchini, 2005).

The EU CARBOSOL Project (Carbonaceous Aerosols over Europe, <http://www.vein.hu/CARBOSOL>), combined weekly measurements of EC, OC, inorganic ions, elemental composition, levoglucosan and radioactive tracers across a network of 6 sites in southern-central Europe.

Extensive details of the CARBOSOL measurements, as well as of model studies with the EMEP EC/OC model, will be published in a special-issue collection in the Journal of Geophysical Research (including Simpson et al., 2007, Tsyro et al., 2007).

The monitoring of the atmospheric particles have been developed and extend to presently now 8 sites in the Nordic countries. The longest record, Hyytiälä are now more than 10 years and several have records over 6 years. The measurements and the data evaluation are thoroughly described in Tunved et al, 2003, 2005, 2006a and 2006b.

### 4.7.2 Modelling

The model used in this work is an extended version of the EMEP MSC-W 3-D photo-oxidant model (Simpson et al., 2003; Fagerli et al., 2004). The extended model has a number of semi-volatile organic gases which are assumed to be able to condense onto (or evaporate from) pre-existing OC using the standard gas/particle partitioning ideas developed by Pankow (1994a,b) and Odum et al. (1997). These

SOA schemes have been described previously in previously by Andersson-Sköld and Simpson (2001) and Simpson and Makar (2004). Two versions of the model will be referred to here.

The Kam-2 version uses the gas/particle scheme as developed by Andersson-Sköld and Simpson (2001). The advantage of this scheme compared to others available in the literature is that it was evaluated against smog-chamber experiments covering a wide-range of conditions, with  $\alpha$ -pinene concentrations from 900 ppb to 20 ppb, NO<sub>x</sub> from 240 ppb to 0 ppb, and temperatures from 49°C to 11°C.

A second SOA module, Kam-2X has identical chemical reactions, but with increased partitioning coefficients, which leads to significantly greater SOA formation in some areas - particularly Northern Europe (where cold temperatures encourage condensation).

A new inventory for annual national BC and OC emissions developed by Kupiainen and Klimont (2004, 2007) has been used. Estimates of BC, OC and PM emissions were based upon an extensive review of the literature, and further checked for consistency with estimates of fine PM included previously in the model. For BC and OC the approach differs between stationary and mobile sources. An important advantage of this inventory is that it includes details for a very large number of sources. In particular, we were able to extract the emissions from wood-burning sources separately, which allowed verification against the levoglucosan measurements performed within EMEP and the CARBOSOL project.

EC ageing, i.e. transformation from hydrophobic to hydrophilic, has been accounted for in the model as described in Tsyro et al. (2007). Emissions from wild forest fires from Global Fire Emission Database (van der Werf et al., 2006) have been also included in EC calculations.

### 4.7.3 Results

A large number of results for sites across Europe are presented in Simpson et al. (2007) and Tsyro et al. (2007). Here we show some results for Nordic sites. As noted above, the EMEP model was run with two versions of the SOA chemical mechanism, denoted Kam-2 and Kam-2X, with the latter giving a higher partitioning from gas to aerosol phase. Figure 1 gives examples of the model results for elemental carbon (EC), and then total carbon (TC=OC+EC) for sites in Norway, Sweden, and Finland, using both of these SOA schemes. The comparison with TC should be regarded as the most robust, as this avoids the well-known problems of defining the OC/EC split (Schmid et al., 2001; ten Brink et al., 2004).

In Northern Europe the model reproduced observed TC levels to a fair degree, although with a tendency to over-predict in winter. In southern Europe (not shown) both model versions severely under-predict observed TC levels. The Kam-2X module (or, equivalently, the assumption of greater condensation rates) sometimes improves model performance in summertime, but can exacerbate problems in winter.

Levoglucosan is a useful marker of the wood-burning contribution to observed PCM. Results for levoglucosan at these sites are shown in Figure 2. In these plots the modelled levoglucosan (assumed levoglucosan/OC = 10% for emissions from wood-burning), is given together with lines covering an uncertainty range of factors of two (representing assumptions that levoglucosan was 5% or 20% of emitted OC). This range of uncertainty is used because of the known wide variability in the emission fraction of levoglucosan compared to OM (e.g. Fine et al., 2002; Simoneit, 2002; Yttri et al., 2005). In Simpson et al. (2007) it is shown that for some sites in southern Europe wood-burning emissions (at least in the vicinity of the sites) are severely underpredicted, and correction for this could substantially improve model comparisons with OC and EC (Tsyro et al., 2007). However, for Nordic sites the plots suggest that wood-burning emissions in the model are of the same order of magnitude as in the observations, although the number of data are too small to draw certain conclusions.

Further, we used EC as a (crude) tracer of primary PM from combustion sources, which are largely mobile sources in summer period and residential/commercial combustion in winter time in many countries. Analysis of model performance compared with measurements for winter period suggests that EC (PM) emissions from residential combustion might be overestimated at Nordic countries and underestimated in some south European countries. On the other hand, the model underestimation of EC for most of sites in summer period is probably due to inaccuracies in either emission estimates of EC/PM emissions from mobile sources or in their spatial distributions. Accounting for emissions from forest fires improved model results (bias and correlation) for several sites.

#### 4.7.4 Monitoring

Monitoring background sites specializing on the atmospheric aerosol have been established in background areas, mostly for research purpose. Presently 8 sites have been established in the Nordic countries ranging from Lille Valby in Denmark to Zeppelin in the Spitzbergen and from Birkenes in Norway to Vääriö in Finland. Collecting data over years on size distribution and chemistry have shown to be very effective in not only getting means of concentrations and size but also quantitative estimates of emission, formation and deposition processes.

The data could not only be used for monitoring the air quality and to detect changes but also to develop new descriptions of different processes and to evaluate models. This development facilitates more thoroughly tested models, as the models can be tested also on a detailed level.

The major results achieved show:

- The Lagrangian studies on particle size distributions from the Nordic particle super sites show the particle formation as a result of BVOC emissions from the boreal forest and in magnitude support the modelling results.
- The data as well show good agreement with the advanced dynamic aerosol process models, UHMA, developed by University of Helsinki. It shows clearly that the emissions from the boreal forest controls the size and chemistry of the natural aerosol over the boreal forest after about 1 day of transport of the air mass over the boreal forest.
- The boreal forest aerosol is dominated by organic compounds while the polluted aerosol in the European plume entering over the is mainly dominated by inorganic compounds. The anthropogenic aerosol is thus expected to have significantly different influence on the clouds and the radiation balance than the natural aerosol.

#### 4.7.5 Conclusions

The modelling work conducted over the last years suggests that:

- The EMEP model does a good job of reproducing concentrations of pollutants with well-known emissions and chemistry, for example of sulphate.
- In Northern Europe the model predicts TC levels which are in line with measured values, especially with the Kam-2X scheme. These predictions are dominated by modelled BSOA.
- In Southern Europe, including the CARBOSOL sites, both versions of the SOA model significantly under-predict TC levels, especially in wintertime. Comparison with the results of Gelencsér et al. (2007), and with levoglucosan, suggests that the model under-predicts both the biomass burning and SOA components of the measured TC at these sites, and the biomass component of EC.
- At all sites, the contribution of BSOA far exceeds that of ASOA (anthropogenic SOA). The relative contribution of BSOA compared to POA and background components varies significantly across Europe though, and at different times of year.
- Model results regarding SOA are extremely sensitive to assumptions about unknown variables (e.g. volatility assumptions and vapour pressures, stoichiometric coefficients, even basic mechanisms).
- The major uncertainties associated with modelling of PCM lie with the emission inventories and modelling of compounds from biomass-burning, and with SOA modelling.

The monitoring data evaluation work conducted over the last years suggests that:

- The Lagrangian studies on particle size distributions from the Nordic particle super sites show the particle formation as a result of BVOC emissions from the boreal forest and in magnitude support the modelling results.
- The data as well show good agreement with the advanced dynamic aerosol process models, UHMA, developed by University of Helsinki. It shows clearly that the emissions from the boreal forest controls the size and chemistry of the natural aerosol over the boreal forest after about 1 day of transport of the air mass over the boreal forest.
- The boreal forest aerosol is dominated by organic compounds while the polluted aerosol in the European plume entering over the is mainly dominated by inorganic compounds. The

anthropogenic aerosol is thus expected to have significantly different influence on the clouds and the radiation balance than the natural aerosol.

#### 4.7.6 Future work

A large number of factors make it difficult to conduct a true verification of modelled versus observed concentrations for BC and OC. One major problem has always been that measurements of OC actually contain thousands of different and largely unidentified molecules, usually with no information on the proportion of primary, secondary, anthropogenic, or biogenic sources. Without such a distinction model verification will always be severely limited. The possible reasons for problems in modelling SOA, biomass-burning and other emissions are discussed in detail in Simpson et al. (2007) and additionally for EC in Tsyro et al. (2007).

A major problem in the modelling of PCM is that too many steps in the calculation process are very uncertain, and there are too few constraints. It seems clear that much more work is needed to constrain the various steps of the PCM modelling process, including:

- Evaluation of the emissions of BVOC. These emissions have strong seasonal variations. Modelled concentrations of various monoterpenes can in principle be validated against measured concentrations, but very few data are available. Further, the short lifetime of many terpene species (and especially of sesquiterpenes) makes such comparisons tricky. Still, given the importance of BVOC to SOA formation, and the fact that emissions validation is possible with today's measurement methods, this step alone would act significantly improve the most important input for PCM modelling
- Evaluation of the emissions of EC+OC from anthropogenic sources. Mobile source as well as residential combustion emissions are still highly uncertain, but very amenable to near-source validation experiments.
- Developing accurate and time resolved data for wildfire EC+OC emissions
- Further use of tracers, such as C-14, levoglucosan and markers of primary biological OC (e.g. cellulose, sugars). Such tracers offer much greater possibilities to understand the sources of observed OC than measurements of simply the total OC or TC.

#### 4.7.7 Acknowledgments

The work was supported by the EU project CARBOSOL, and by the EMEP project under UNECE, the Nordic Council of Ministers, and the Swedish MISTRA Project on Organic Aerosols. Kaarle Kupiainen and Zbigniew Klimont are acknowledged for their great efforts with the emission inventory, and CARBOSOL colleagues for their ideas and support.

#### 4.7.8 References and reports

- Y. Andersson-Sköld and D. Simpson. Secondary organic aerosol formation in Northern Europe: a model study. *J. Geophys. Res.*, 106(D7):7357–7374, 2001.
- H. Fagerli, D. Simpson, and S. Tsyro. Unified EMEP model: Updates. In *Transboundary acidification, eutrophication and ground level ozone in Europe. EMEP Status Report 1/2004*, pages 11–18. The Norwegian Meteorological Institute, Oslo, Norway, 2004.
- P.M. Fine, G.R. Cass, and B.R.T. Simoneit. Chemical characterisation of fine particle emissions from the fireplace combustion of woods grown in the Southern United States. *Environ. Sci. Tech.*, 36:1442–1451, 2002.
- A. Gelencsér, B. May, D. Simpson, H. Puxbaum, D. Wagenbach, C. Pio, A. Kasper-Giebl, and M. Legrand. source apportionment of PM<sub>2.5</sub> organic aerosol over Europe: primary/secondary, natural/anthropogenic, fossil/biogenic origin. Accepted for publication in *J. Geophys. Res.*,
- K. Kupiainen and Z. Klimont. Primary emissions of submicron and carbonaceous particles in Europe and the potential for their control. Interim Report IR-04-079, International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria, 2004.
- K. Kupiainen and Z. Klimont. Primary emissions of submicron and carbonaceous particles in Europe and the potential for their control. In Press, 2007.

- H. Schmid, L. Laskus, H.J. Abraham, U. Baltensperger, V. Lavanchy, M. Bizjak, P. Burba, H. Cachier, D. Crow, J. Chow, T. Gnauk, A. Even, H.M. ten Brink, K.-P. Giesen, R. Hitzenberger, C. Hueglin, W. Maenhaut, C. Pio, A. Carvalho, J.-P. Putaud, and D. Toom-Sauntry. Results of the “carbon conference” international aerosol carbon round robin test stage I. *Atmospheric Environment*, 35:2111–2121, 2001.
- B.R.T Simoneit. Biomass burning - a review of organic tracers for smoke from incomplete combustion. *Applied Geochemistry*, 17:129–162, 2002.
- D. Simpson and P. Makar. Modelling SOA and OC in Europe. pages 86–92. The Norwegian Institute for Air Research (NILU), Kjeller, Norway, 2004. Transboundary Particulate Matter in Europe, EMEP Status report 4/2004.
- D. Simpson, H. Fagerli, J.E. Jonson, S. Tsyro, P. Wind, and J.-P. Tuovinen. The EMEP Unified Eulerian Model. Model Description. EMEP MSC-W Report 1/2003, The Norwegian Meteorological Institute, Oslo, Norway, 2003.
- D. Simpson, K.E. Yttri, Z. Klimont, K. Kupiainen, A. Caseiro, A. Gelencsér, C. Pio, and M. Legrand. Modelling of carbonaceous aerosol over Europe. *J. Geophys. Res.*, 2007, doi: 10.1029/2006JD008158, In Press.
- H. ten Brink, W. Maenhaut, R. Hitzenberger, T. Gnauk, G. Spindler, A. Even, X. Chi, H. Bauer, H. Puxbaum, J.-P. Putaud, J. Tursic, and A. Berner. INTERCOMP2000: the comparability of methods in use in Europe for measuring the carbon content of aerosol. *Atmospheric Environment*, 38:6507–6519, 2004.
- S. Tsyro, D. Simpson, L. Tarrasón, K. Kupiainen, Z. Klimont, K.E. Yttri, and C. Pio. Modelling of black carbon over Europe. *J. Geophys. Res.*, 2007, doi: 10.1029/2006JD008164, In press.
- P. Tunved, H.-C. Hansson, M. Kulmala, P. Aalto, Y. Viisanen, H. Karlsson, Kristensson, E. Swietlicki, M. Dal Maso, J. Ström, and M. Komppula, One year boundary layer aerosol size distribution data from five Nordic background stations, *Atmos. Chem. Phys.*, 3, 2183-2205, 2003
- P. Tunved, J. Ström and H-C. Hansson, An investigation of processes controlling the evolution of the boundary layer aerosol size distribution properties at the Swedish background station Aspvreten, *Atmospheric Chemistry and Physics*, Vol. 4, pp 2581-2592, 20-12-2004
- P. Tunved, E.D. Nilsson, H-C. Hansson, J. Ström, M. Kulmala, P. Aalto, Y., Viisanen, Aerosol characteristics of air masses in Northern Europe – influences of location, transport, sinks and sources, 2005, *J. Geophys. Res. (D Atmos.)*, vol:110 DOI:10.1029/20
- P. Tunved, H. Korhonen, J. Ström, H.-C. Hansson, K.E.J. Lehtinen, M. Kulmala, Is nucleation capable of explaining observed aerosol integral number during southerly transport over Scandinavia? 2006a, *Tellus*, Vol 58, 129-140
- P. Tunved, H-C. Hansson, P., Kerminen, V-M, Ström, J., Dal Maso, M., Lihavainen, H., Viisanen, Y., Aalto, P.P, Komppula M. And , M. Kulmala, High natural aerosol loading over boreal forests, *Science*, April 14, 2006b
- van der Werf, G.R., J.T. Randerson, L.Giglio, G.J. Collatz, and P.S. Kasibhatla (2006) Interannual variability in global biomass burning emission from 1997 to 2004. *Atmos. Chem. Phys.*, 6, 3423-3441.
- K.E. Yttri and M.C. Facchini. Levels and relative contribution of water-insoluble organic carbon at rural background sites in Europe. EMEP/CCC Report 5/2005, The Norwegian Institute for Air Research (NILU), Kjeller, Norway, 2005.
- K.E. Yttri and M. Kahnert. Preliminary results from the EMEP EC/OC campaign. EMEP/CCC-Report 3/2004, The Norwegian Institute for Air Research (NILU), Kjeller, Norway, 2004.
- K.E. Yttri, C. Dye, L.H. Slørdal, and O.A. Braathen. Quantification of monosaccharide anhydrides by negative electrospray HPLC/HRMS-TOF - application to aerosol samples from an urban and a suburban site influenced by small scale wood burning. *J. Air Waste Manage. Assoc.*, 55:1169–1177, 2005.

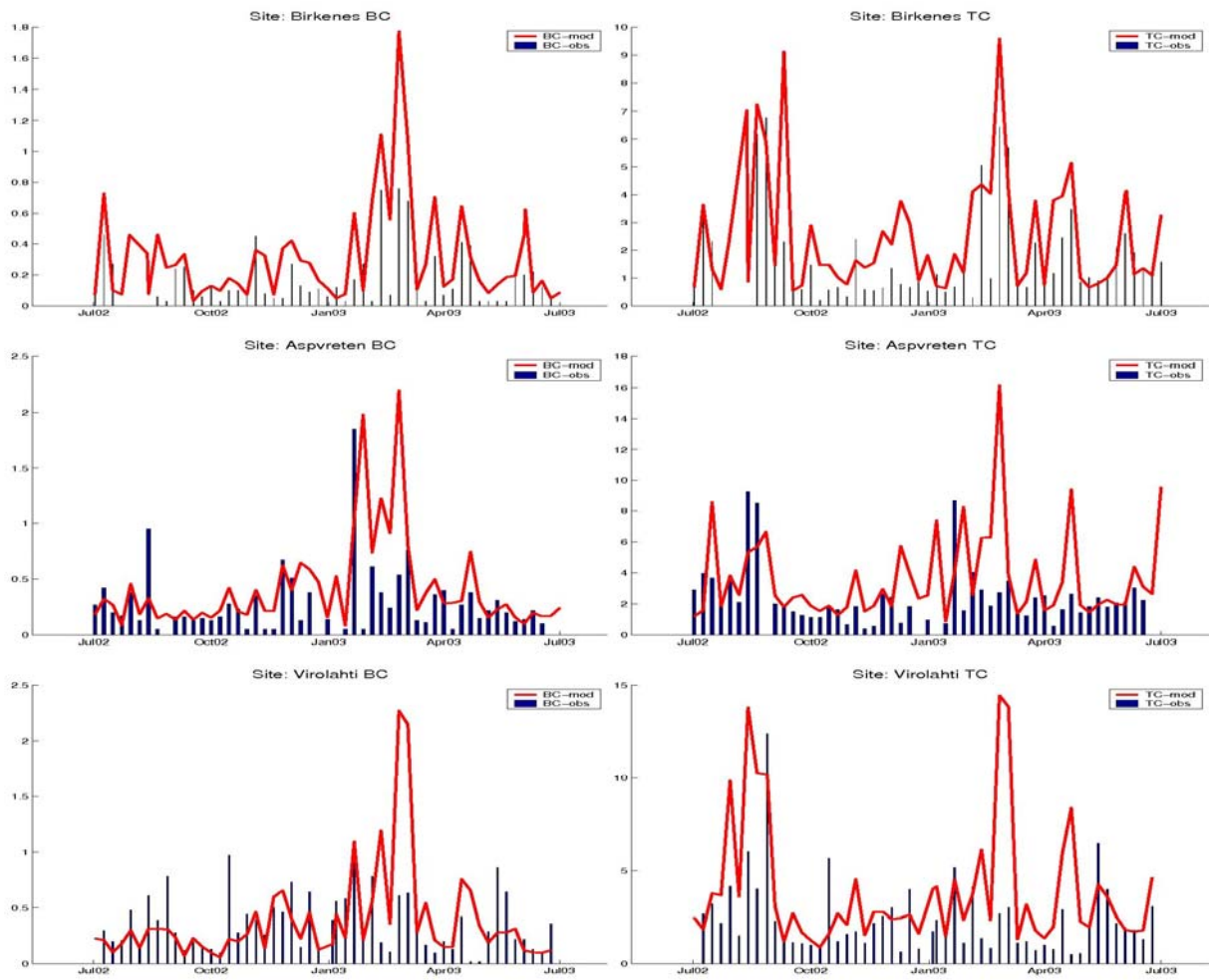


Figure 1: Modelled and Observed Elemental Carbon (here denoted BC, left column) and Total Carbon (TC, right column) Results ( $\mu\text{g}/\text{m}^3$ ), for Birkenes (Norway), Aspvreten (Sweden), and Virolahti (Finland). Uses Kam-2X SOA scheme.

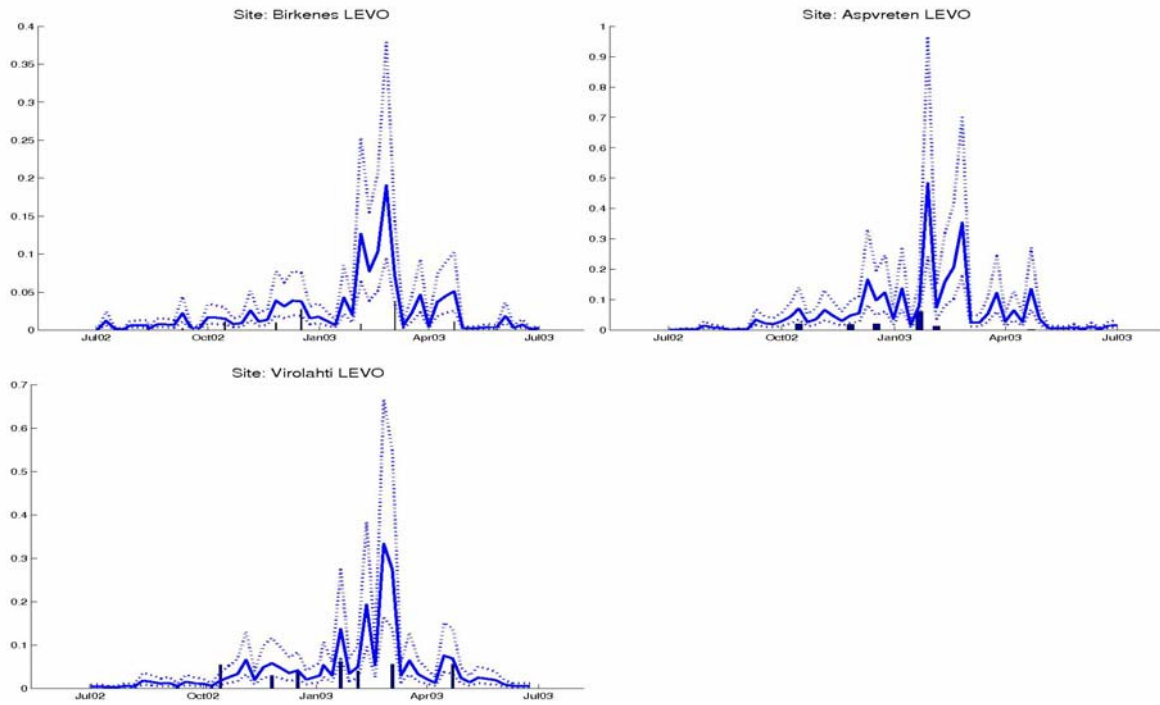


Figure 2: Modelled and Observed Levoglucosan Results ( $\mu\text{g}/\text{m}^3$ ) for Birkenes (Norway), Aspvreten (Sweden), and Virolahti (Finland). Bars indicate observations. Model results given as central value with dotted lines indicating factor of two range.

## 5 References

*Publications associated with NORPAC are marked (\*).*

- \*Andersen, P. (2006): Measurement of particulate carbon in relation to traffic and validation of a Particulate Soot Absorption Photometer (PSAP). MSc thesis. University of Southern Denmark and National Environmental Research Institute, Roskilde, Denmark.
- Hussein T, A. Karppinen, J. Kukkonen, J. Härkönen, P.P. Aalto, K. Hämeri, V-M Kerminen, M Kulmala, 2006. Meteorological dependence of size-fractionated number concentrations of urban aerosol particles. *Atmos. Environ.* 40 (2006) 1427–1440.
- \*Hussein, T., J. Kukkonen, H. Korhonen, M. Pohjola, L. Pirjola, D. Wraith, J. Härkönen, K. Teinilä, I.K. Koponen, A. Karppinen, R. Hillamo and M. Kulmala, 2007. Evaluation and modeling of the size fractionated aerosol particle number concentration measurements nearby a major road in Helsinki – Part II: Aerosol measurements within the SAPPHERE project. *Atmos. Chem. Phys.* 7, 4081–4094.
- IPCC (2007): Climate Change 2007 - The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the IPCC. (Download from: <http://www.ipcc.ch/ipccreports/ar4-wg1.htm>)
- Janhäll S, Jonsson AM, Molnar P, Svensson EA, Hallquist M, Size resolved traffic emission factors of submicrometer particles, *Atmospheric Environment*, 38 (26): 4331-4340 (2004)
- \*Ketznel, M., L. Gidhagen, M. Haakana, M. Hallquist, H.-C. Hansson, M. Kulmala, O.J. Nielsen, D. Simpson, L. H. Slørdal and E. Swietlicki: NORPAC - A Nordic Project on PM Measurements and Modelling: Overview and First Results. NOSA Symposium in Stockholm, 11-12 November 2004. (Abstract and Poster)
- \*Ketznel, Matthias, Gunnar Omstedt, Christer Johansson, Ingo Düring, Lars Gidhagen, Achim Lohmeyer, Ruwim Berkowicz, Peter Wählin: Estimation and validation of PM<sub>2.5</sub>/PM<sub>10</sub> exhaust and non-exhaust emission factors for street pollution modelling. Presentation at the 5th Internatl. Conf. on Urban Air Quality (UAQ 2005), Valencia, Spain, 29-31 March 2005. (Short Paper)
- \*Ketznel, M., G. Omstedt, C. Johansson, I. Düring, M. Pohjola, D. Oettl, L. Gidhagen, P. Wählin, A. Lohmeyer & R. Berkowicz (2007): Estimation and validation of PM<sub>2.5</sub>/PM<sub>10</sub> exhaust and non-exhaust emission factors for practical street pollution modelling, *Atmospheric Environment* 41: 9370–9385.
- \*Ketznel, M., Glasius, M., Wählin, P., Jensen, B., Andersen, P., Ström, J., Hansson, H.C. & Palmgren, F.: A simple optical soot sensor - laboratory tests and first field data on wood stove emissions. Nordic Society for Aerosol Research (NOSA), Göteborg, Sweden, 3-4 November 2005.
- \*Ketznel, M., Pagels, J., Wählin, P., Glasius, M., Jensen, B., Swietlicki, E. & Palmgren, F.: Volatility measurements of submicrometer particles in a street canyon in Copenhagen. Nordic Society for Aerosol Research (NOSA), Göteborg, Sweden, 3-4 November 2005.
- Kiss, Gyula, Etelka Tombácz, Hans-Christen Hansson, Surface tension effects of humic-like substances in the aqueous extract of tropospheric fine aerosol, *J of Atm Chem* in press
- Korhonen, H., Lehtinen, K. E. J., Kulmala, M. 2004. Aerosol dynamic model UHMA: Model development and validation. *Atmospheric Chemistry and Physics* 4, 757-771.
- Kukkonen J., Sokhi R.S., and M. Ketznel M., 2005. Modelling of urban air particulate matter – some future perspectives and policy implications. In: Sokhi, RS, Millán, MM, Moussiopoulos, N (eds.): Proceedings (CD) of the 5th International Conference on Urban Air Quality, Valencia, 29-31 March 2005, University of Hertfordshire, UK, 2005. ISBN 1-898543-92-5. (4 pages).
- \*Kukkonen, Jaakko, Ranjeet Sokhi, Lakhu Luhana, Jari Härkönen, Timo Salmi, Mikhail Sofiev and Ari Karppinen, 2007. Evaluation and application of a statistical model for assessment of long-range transported proportion of PM<sub>2.5</sub> in the United Kingdom and in Finland *Atmos. Environ.* (in print).
- \*Karppinen A, Kukkonen J, Kauhaniemi M, Härkönen J, Nikmo J, Sokhi RS, Luhana L, Kousa A, Alaviippola B, Koskentalo T and Aarnio P, 2005. Evaluation and application of a model for the urban and regional scale concentrations of PM<sub>2.5</sub>, In: Sokhi, RS, Millán, MM, Moussiopoulos, N



- (eds.): Proceedings (CD) of the 5th International Conference on Urban Air Quality, Valencia, 29-31 March 2005, University of Hertfordshire, UK, 2005. ISBN 1-898543-92-5. (4 pages).
- \*Massling, A., Löndahl, J., Ketzel, M., Jensen, B., Wählin, P., Bilde, M., Rosenbohm, E., Hansson, H.-C., Hallquist, M., Lunder, C. and Swietlicki, E. (2005): Intercomparison of Particle Number Size Spectrometers. 2nd Joint BACCI meeting, 13 – 15 June 2005, Kuopio, Finland.
  - \*Massling, A., Löndahl, J., Swietlicki, E., Ketzel, M., Jensen, M., Wählin, P., Bilde, M., Kristensson, M., Hansson, H.-C., Ströhm, J., Jonsson, A., Hallquist, M., Lunder, C (2006): DMPS/SMPS intercomparison in terms of particle sizing. Technical note in preparation.
  - Mønster J., T. Rosenørn, B. Svenningsson and M. Bilde “Evaporation of methyl- and dimethyl-substituted malonic, succinic, glutaric and adipic acid particles at ambient temperatures” *Journal of Aerosol Science*, **2004**, 35, 1453-1465.
  - Pirjola L. (1999) Effects of the increased UV radiation and biogenic VOC emissions on ultrafine aerosol formation, *J. Aerosol. Sci.*, **30**, 355-367.
  - Pirjola, L. and Kulmala, M. (2000) Aerosol dynamical model MULTIMONO. *Boreal Environ. Res.*, **5**, 361-374.
  - Pirjola, L. and Kulmala, M. (2001) Development of particle size and composition distribution with a novel aerosol dynamics model. *Tellus*, **53B**, 491-509.
  - Pirjola, L., Tsyro, S., Tarrason, L. and Kulmala, M. (2003) A monodisperse aerosol dynamics module – a promising candidate for use in the Eulerian long-range transport model. *Journal of Geophysical Research*, **108** (D9), 4258.
  - Pirjola, L., Parviainen, H., Hussein, T., Valli, A., Hämeri, K., Aalto, P., Virtanen, A., Keskinen, J., Pakkanen, T., Mäkelä, T., Hillamo, R. (2004) “Sniffer” – a novel tools for chasing vehicles and measuring traffic pollutants. *Atmospheric Environment* **38**, 3625-3635.
  - Pirjola, L., P. Paasonen, D. Pfeiffer, T. Hussein, K. Hämeri, T. Koskentalo, A. Virtanen, T. Rönkkö, J. Keskinen, T.A. Pakkanen and R.E. Hillamo (2006) Dispersion of particles and trace gases nearby a city highway: mobile laboratory measurements in Finland. *Atmospheric Environment*, **40**, 867-879.
  - Pohjola, M A, Pirjola, L, Kukkonen, J, Kulmala, M. 2003. Modelling of the influence of aerosol processes for the dispersion of vehicular exhaust plumes in street environment. *Atmospheric Environment*, 37, 3. pp. 339-351.
  - \*Pohjola M A, Pirjola L, Kukkonen J, Karppinen A, Härkönen J, Ketzel M: MODELLING OF DISPERSION AND AEROSOL PROCESSES IN A ROADSIDE ENVIRONMENT, AND EVALUATION WITH MEASURED DATA. Presentation at the 5th Internatl. Conf. on Urban Air Quality (UAQ 2005), Valencia, Spain, 29-31 March 2005.
  - \*Pohjola, M. A., Pirjola, L., Kukkonen, J., Kulmala, M. 2006a. Correction to modelling of the influence of aerosol processes for the dispersion of vehicular exhaust plumes in street environment. *Atmospheric Environment*, 40. pp. 311-314.
  - \*Pohjola, M A, Pirjola L, Kukkonen J, Karppinen A, Härkönen J, Ketzel M, 2006b. Roadside environment particle modelling and evaluation with measured data – coupling a dispersion model and an aerosol process model. In: Vehkamäki H, Bergman T, Julin J, Salonen M, Kulmala M (eds.): Proceedings of the NOSA 2006 Aerosol symposium, combined with the X Finnish National Aerosol Symposium, Finnish-Czech Aerosol Symposium, and BACCI workshop, Helsinki 8.-10.11.2006. pp. 292-296.
  - Pohjola, M., 2006c. Evaluation and modeling of the spatial and temporal variability of particulate matter in urban areas. Ph.D thesis. Finnish Meteorological Institute Contributions, 57. 143 p.
  - \*Pohjola, M A, Pirjola, L, Karppinen, A, Härkönen, J, Ketzel, M, Kukkonen, J, 2007. Evaluation and modelling of the size fractionated aerosol particle number concentration measurements nearby a major road in Helsinki – Part I: Modelling results within the LIPIKA project. *Atmos. Chem. Phys.* 7, 4065-4080.
  - Riipinen I., K. E. J. Lehtinen, B. Svenningsson, M. Bilde, A. Gaman, M. Kulmala ” A method for determining thermophysical properties of organic material in aqueous solutions: succinic acid” *Submitted to Atmospheric Research* **2005**
  - \*Saarikoski, Sanna, Markus Sillanpää, Mikhail Sofiev, Hilikka Timonen, Karri Saarnio, Kimmo Teinilä, Ari Karppinen, Jaakko Kukkonen and Risto Hillamo, 2007. Chemical composition of aerosols during a major biomass burning episode over northern Europe in spring 2006: Experimental and modelling assessments. *Atmos. Environ.* 41 (2007), 3577–3589.
  - \*Sillanpää M., Saarikoski S., Hillamo R., Pennanen A., Makkonen U., Spolnik U., Van Grieken R., Koskentalo T., Salonen R.O., 2005. Chemical composition, mass size distribution and source

analysis of long-range transported wildfire smokes in Helsinki. *Science of Total Environment* 350, 119–135.

- \*Simpson, D., K.E. Yttri, Z. Klimont, K. Kupiainen, A. Caseiro, A. Gelencsér, C. Pio, and M. Legrand. Modelling of carbonaceous aerosol over Europe. *J. Geophys. Res.*, 2007, In Press.
- E. Swietlicki, A. Kristensson, H. Areskoug and H.-C. Hansson, Source apportionment of PM10 and PM2.5 at a background site in southern Sweden, Accepted by *Atm Env.*
- \*Tsyro, S., D. Simpson, L. Tarrasón, K. Kupiainen, Z. Klimont, K.E. Yttri, and C. Pio. Modelling of black carbon over Europe. *J. Geophys. Res.*, 2007, In Press.
- P. Tunved, H. Korhonen, J. Ström, H.-C. Hansson, K.E.J. Lehtinen, M. Kulmala, A pseudo-Lagrangian model study of the size distribution properties over Scandinavia: transport from Aspvreten to Värriö, *Atmospheric Chemistry and Physics Discussions*, Vol. 4, pp 7757-7794, 29-11-2004
- P. Tunved, E.D. Nilsson, H-C. Hansson, J. Ström, M. Kulmala, P. Aalto, Y., Viisanen, Aerosol characteristics of air masses in Northern Europe – influences of location, transport, sinks and sources, Submitted to *JGR*
- P. Tunved, J. Ström and H-C. Hansson, An investigation of processes controlling the evolution of the boundary layer aerosol size distribution properties at the Swedish background station Aspvreten, *Atmospheric Chemistry and Physics*, Vol. 4, pp 2581-2592, 20-12-2004
- P. Tunved, H. Korhonen, J. Ström, H.-C. Hansson, K.E.J. Lehtinen, M. Kulmala, Is nucleation capable of explaining observed aerosol integral number during southerly transport over Scandinavia? Submitted to *Tellus*.
- WHO, 2004: Health aspects of air pollution - Results from the Project 'Systematic Review of Health Aspects of Air Pollution in Europe'. World Health Organisation Regional Office for Europe. Document E83080 (<http://www.who.int/> accessed 13 April 2006)

## Appendix 1: Report on related projects

*Selected list of major projects that NORPAC groups are involved in and that have a relation to NORPAC connection to international activities, contribution for funding.*

Activity/title	Participants	Funded by	Short Description / Main findings	Reference, publications
SAPPHIRE, 2002 - 2005	7 partners and 3 sub-contractors from the area of 5 countries?	EU, City of Tomorrow	Development of methods for source apportionment of airborne particulate matter and polycyclic aromatic hydrocarbons in urban regions of Europe, and measurements of the same.	<b>References:</b> <a href="http://www.gees.bham.ac.uk/research/sapphire/">http://www.gees.bham.ac.uk/research/sapphire/</a>
Nordic Centre of Excellence BACCI	Finland, Sweden, Denmark, Norway	Academy of Finland	The main objective is to study the importance of aerosol particles on climate change and on human health. The approach covers both experimental (laboratory and field experiments including development of novel instrumental techniques) and theoretical (basic theories, simulations, model development) approaches.	<a href="http://www.atm.helsinki.fi/BACCI/">http://www.atm.helsinki.fi/BACCI/</a>
OSCAR (2002-2005)	12 participants from 6 European countries, including FMI and NILU	EU/FP5	The overall aim of OSCAR is to develop and deliver an optimised system for the assessment of the impacts of road traffic on urban air quality in local scale.	<a href="http://www.eu-oscar.org">www.eu-oscar.org</a>
Workshop from 29.11.2004 – 03.12.2004 at the Leibniz-Institute for Tropospheric Research in Leipzig, Germany.	IfT Leipzig, TSI, Grimm, Lund University		Intercomparison of eleven particle number size spectrometers by comparing particle number size distributions of differently generated polydisperse and monodisperse aerosols with varying chemical composition.	
ASTA		Swedish Foundation – MISTRA	International and National Abatement Strategies for Transboundary Air Pollution	<a href="http://asta.ivl.se/">http://asta.ivl.se/</a>
CARBOSOL, 2002-2005	France, Austria, Germany, Portugal, Hungary, Norway	EU-FP5	Carbonaceous aerosol in Europe – assessment of OC in southern/central Europe, including historical assessment (ice/snow cores)	
GÖTE 2005		Swedish Foundation - MISTRA	A major collaborative measurement campaign during winter conditions (2 Feb, 2 Mar 2005 ), in Göteborg	<a href="http://www2.chem.gu.se/~hallq/Gote_2005.htm">http://www2.chem.gu.se/~hallq/Gote_2005.htm</a>
LIPIKA	6 Finnish partners: TUT,Stadia, UH/TTL, FMI, VTT,YTV	TEKES	Mobile laboratory measurement campaign of roadside site ultrafine/fine particle number concentrations and size distributions, NO,NO2,NOx,CO, Helsinki (2003-2004)	<a href="http://nuuskija.stadia.fi/">http://nuuskija.stadia.fi/</a>

## Appendix 2: List of NORPAC-organised workshops and meetings

- Kick-off workshop, 29 March 2004, Copenhagen
- 14 June 2004, status meeting in connection with the BACCI workshop in Lund (14.-15. June)
- 17 August 2004: working meeting on emission factors and local modelling (in Lund with SMHI and DMU)
- 19 August 2004: working meeting on aerosol modelling (at FMI/Helsinki with Stadia and DMU)
- 10 November 2004, 2nd project workshop in connection with the NOSA symposium at ITM / Stockholm University
- 21 April 2005 :Workshop on Aerosol Dynamics Modelling in the Nordic Countries on Local, Urban and Regional Scale (at FMI/Helsinki)
- 9 May 2005, 3rd project workshop in Copenhagen
- 4-5 October 2005: Workshop on modelling of PM emissions (at SMHI / Norrköping)
- 2 November 2005, 4th workshop in connection with annual NOSA symposium, Göteborg University
- 19 May 2006, 5th project workshop in Copenhagen
- 8 November 2006: 6<sup>th</sup> project workshop in Helsinki