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A pseudo-Lagrangian model study of the size distribution properties over Scandinavia: transport from Aspvreten to Värriö

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A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

Abstract

The evolution of the aerosol size distribution during transport between Aspvreten (58.8° N, 17.4° E) and Värriö (67.46° N, 29.35° E) was studied using a pseudo-Lagrangian approach. Aerosol dynamic processes were studied and interpreted utilizing a state-of-the-art aerosol dynamic box model UHMA (University of Helsinki Multicomponent Aerosol model) complemented with OH, NO₃, O₃ and terpene chemistry. In the model simulations, the growth and formation of aerosol particles was controlled by sulphuric acid, ammonia, water and an unidentified low volatile organic compound. This organic compound was assumed to be a product of terpene oxidation with a yield of 13% in the base case conditions.

Changes of aerosol size distribution properties during transport between the stations were examined in twelve clear sky cases. On average, the modelled number agreed fairly well with observations. Mass concentration was overestimated by 10%.

Apart from dilution, the only removal mechanism for aerosol mass is dry deposition. A series of sensitivity tests performed revealed that the absolute magnitude of dry deposition effects on the aerosol size distribution is slow overall. Furthermore, nucleation does not leave a significant contribution to aerosol number in the selected cases. The sensitivity of the modelled size distribution to concentration of precursor gases and oxidants is, however, obvious. In order to explain observed mass increase during transport we conclude that a yield of low volatile products from oxidation of terpenes of 10–15% is required to explain observed growth rates. Coagulation is acknowledged to be highly important in modelled cases.

1. Introduction

The atmospheric aerosol is believed to cool the climate through direct and indirect forcing. The direct forcing result from the ability of aerosols to scatter and absorb incoming solar radiation, while the indirect effect is a result of the ability of aerosols to

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

A pseudo-Lagrangian model study of the size distribution properties over ScandinaviaP. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

act as cloud condensation nuclei (CCN) and thereby to alter the radiative properties of clouds (Twomey, 1974). An increased aerosol number concentration contributes to more cloud droplets and thus brighter clouds. Still, the climatic effects from particulate matter constitute the greatest uncertainty in climate models (IPCC, 2001).

Furthermore, aerosol particles are an important component of air pollution known to cause considerable public health effects. As the concentration of sulphur dioxide has decreased significantly in Europe during the recent decades attention is now turned to particulate matter. Epidemiological studies performed by e.g. Künzli et al. (2000) in three European countries attribute 6% of total mortality and a significant increase of morbidity to air pollution.

On regional scale, the aerosol is associated with large temporal and spatial variability. In order to reduce the uncertainty associated with assumed effects of aerosols on climate we need to improve our understanding of the important dynamical processes controlling the aerosol size distribution on large spatial and temporal scales.

In order to perform studies of the importance of different source and sink processes affecting the aerosol it is favourable to make use of simultaneous measurements of aerosol properties on a regional scale. This type of measurement approach gives a relevant connection both in space and time, often lacking in previous investigations.

During recent years, regional long-term measurements of aerosol size distribution have been evolving in the Nordic countries. So far, measurements from four stations in Finland and Sweden have resulted in a database comprising continuous and simultaneous aerosol size distribution and meteorological data since autumn 1999. Some of the stations contribute with longer records of size distribution measurements.

The stations in the existing Nordic measurement network are representative for the background boreal environment, characteristic for Northern Europe (see Tunved et al., 2003 for a description of the network). The European boreal region covers more than 2 900 000 km². Outside Europe, the type of ecosystem and climate characteristics encountered in this region find counterparts in especially north-western parts of USA and western Canada as well as parts of Northern Asia. Thus, the European boreal

region is representative for economically important areas, including both sparsely and highly populated regions.

Recent investigation of the aerosol properties at these stations has mainly focused on the local properties of the aerosol and source-receptor relationships. Particularly well studied is the role of nucleation in the boreal environment. Measurements performed at especially Hyytiälä have greatly improved our understanding of new particle formation in this type of environment (e.g. Mäkelä et al., 1997, 2000; Nilsson et al., 2001a, b; Kulmala et al., 1998, 2001, 2004a).

From previous investigations of changes of aerosol size distribution properties during transport in-between the stations in the network several features was exposed. Firstly, the processes acting on the aerosol over the time scale of transport between the stations are rapid. This further implies that constant aerosol fields may not be used to characterize the aerosol over scales defined by Northern Europe (Tunved et al., 2003). Tunved et al. (2003) concluded that during southerly transport the air masses experience a source of aerosol number, while transport from south to north often is associated with a significant decrease in aerosol number and mass concentration. The increase of aerosols during southerly transport is likely to be explained by gas to particle formation. Nucleation is often observed in air-masses arriving from north, probably due to low pre-existing aerosol surface and typical mixing and temperature profiles in this kind of air-masses (Nilsson et al., 2001a, b).

In the present study we utilize a box model approach to explore the role of different processes shaping the aerosol during transport under clear sky conditions. The investigation is limited to studying the transport from south to north making use of two background stations as reference points for our model study (Aspvreten, some 70 km south of Stockholm (58.8° N, 17.4° E) and Värriö, Finnish Lapland (67.46° N, 29.35° E)). We utilize a pseudo-Lagrangian box model approach to explain the observed decrease in particle mass and number concentration and to explore the role of different processes in shaping the aerosol population under clear sky conditions. Twelve different transport situations were modelled and compared to observations. Sensitivity tests

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

are performed for one selected case in order to address the importance of different dynamical processes affecting the aerosol.

2. Methods

2.1. Stations and size distribution measurements

5 Simultaneous size distribution measurements from two stations during a period of one year (1 June 2000 to 31 May 2001) are utilized in this study. Both stations utilized similar observational methods by means of Differential Mobility Particle Sizer (DMPS).

The SMEAR I station (67.46° N 29.35° E, 400 m a.s.l.) in Värriö is located in Finnish Lapland. The station is far from any pollution sources. However, emissions on the
10 Kola Peninsula give rather strong signals (especially high SO₂ and particle number and mass) when winds are transporting air from this region (e.g. Kulmala et al., 2000). The Differential Mobility Particle Sizer (DMPS) system set-up observes a complete size scan between 8 and 460 nm approximately every 10 min.

Aspvreten (58.80° N, 17.40° E, 25 m a.s.l.), is located in the county of Sörmland, some 70 km south west of Stockholm. The station is situated 2 km inland from the Baltic
15 Sea. Influence from local anthropogenic activities is minimal (Tunved et al., 2003). The DMPS instrument set-up observes one size distribution spectrum every 6 min in the size ranges 10–452 nm.

2.2. Pseudo-Lagrangian assumption

20 In this work, we investigated the importance of different dynamic and chemical processes affecting the aerosol during transport from south to north with the help of a pseudo-Lagrangian approach. A detailed aerosol dynamics model was run along trajectories passing through both a southern source region defined by Aspvreten and the northern receptor region characterised by Värriö. The continuous particle size distribu-

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

tion measurements at both stations enable us to test our understanding of the factors shaping the size distribution during the transport.

The 120 h trajectories with one-hour resolution were calculated with the HYS-PLIT4 model (Draxler and Hess, 1997, 2003). The trajectory calculation relies on NCEP/GDAS FNL reanalysis data. The trajectory gives transport time and the time dependent meteorological output data for each of the endpoints. This allows us to run the aerosol dynamic model along each trajectory with adequate meteorological input.

Trajectories are calculated arriving to the receptor site 10 m a.g.l. Trajectories were used only if spending a majority of time in the boundary layer.

2.3. Aerosol dynamic model

The aerosol dynamic box model run along the trajectories was the University of Helsinki Multicomponent Aerosol model (UHMA) described in detail in Korhonen et al. (2004). The model incorporates the major microphysical processes that affect the aerosol under clear sky conditions, namely nucleation, coagulation, multicomponent condensation and dry deposition.

In this study, the vapours condensing onto aerosol particles were sulphuric acid, ammonia, water and a low-volatile organic compound following the nano-Köhler mechanism, suggested by Kulmala et al. (2004b), and thus capable of contributing to the growth of newly formed particles after a threshold size of few nanometres was reached. Although such low volatile organic compounds have not been identified in the atmosphere, analysis of new particle formation events in boreal forest have shown that sulphuric acid can explain only a small fraction of the observed growth of nucleation mode particles (Boy et al., 2003). Furthermore, indirect experimental evidence indicates that the dominant compounds contributing to the growth of newly formed particles in forested areas are organic (O'Dowd et al., 2002).

The dry deposition rate of particles was calculated according to Rannik et al. (2003) over land and according to Slinn et al. (1978) over sea. Nucleation was simulated according to the kinetically limited nucleation mechanism, which essentially gives the

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

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

5 The particle size distribution was approximated with 55 size sections in the size range of 0.7 nm–2 μ m. Of the three size distribution descriptions available in UHMA, this study utilized the hybrid structure. In this structure, the particles are split into fixed size sections based on their core volume of sulphuric acid and condensable organic matter. The advantage with the approach is that it eliminates the numerical diffusion associated with the condensation of ammonia and water, the amount of which in the
10 particle phase is determined by equilibration at every model time step.

2.4. Treatment of chemistry

In the model we assume two species, except water vapour and ammonia, to control the growth and formation of particles: sulphuric acid, which is formed by oxidation
15 of SO₂ by hydroxyl radical (OH) and some unidentified product from degradation of terpenes formed by reaction between terpenes and hydroxyl radical (OH), ozone (O₃) and nitrate radical (NO₃). Concentration of OH and NO₃ is calculated assuming steady state following Janson et al. (2001).

Concentration of OH is determined by assuming balance between formation and
20 degradation of HO_x (OH, HO₂). Sources of HO_x are reaction of singlet oxygen (O¹D), produced from photolysis of O₃, and water vapour. Photolysis of HCHO and ozonolysis of terpenes also contributes to OH production. Photolysis constants are taken from Derwent and Jenkin (1990). Clear sky conditions are assumed. Solar zenith angles are calculated along trajectories from geographical coordinates, day of year and time
25 of day. The sinks of odd oxygen are HO₂-HO₂ self-reaction, reaction with terpenes, OH-HO₂ reaction and formation of nitric acid from combination of OH and NO₂. Daily average of calculated OH concentration was adjusted to be of same magnitude as reported by Hakola et al. (2003) using an observationally constrained box model. Slightly

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

higher values were however used in the present study.

Nitrate radical is formed by reaction of NO_2 and O_3 . NO_3 is highly sensitive to photolytic degradation and reaches near zero concentration during daytime. Temperature dependent dissociation and photolysis of N_2O_5 also serves as source of nitrate radical.

5 NO_3 is consumed due to photolysis, NO_3 -NO reaction, NO_3 - NO_2 reaction and reaction of NO_3 with terpenes.

To account for terpene emissions a parameterisation of this process, as presented by Lindfors et al. (1999) was implemented. The approach includes temperature dependent emissions of terpenes in grams per gram dry weight foliar biomass. Terpene emissions are exponentially dependent on temperature. The parameterisation relies on a latitude dependent biomass density which is valid for Finland over latitudes from 60° N to 69° N. As the Swedish source site investigated lies on lower latitude, we extrapolated the data behind the parameterization for latitudes down to 58.8. Furthermore, we assumed that the parameterisation is also representative of Swedish forested areas.

15 Once emitted, mono-terpenes are subjected to degradation by ozone, hydroxyl radical and nitrate radical. The rate constants for initial chemical reactions are rather well known for the most commonly encountered terpenes. We do not discriminate between different terpene species. Average rate constants for primary oxidation steps for the dominating terpene species (Janson et al., 2001) were used. Average molar mass of oxidation products is set to 150 g/mol.

2.5. Data usage and design of base case

Some 60 trajectories describe transport between Aspvreten and Värriö during the period of investigation. This investigation focuses on transport during clear sky conditions only. By investigating satellite images and relative humidity (RH) history of the single trajectories we are rather certain that at least 20 of the selected trajectories actually describe transport without influence of clouds or precipitation.

However, further precautions need to be included in this analysis. One of the main

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

concerns is the selection of an input aerosol at Aspvreten. Since the observed aerosol properties are dependent on source area we need to make certain that the conditions do not change significantly during a period of at least 6 h when the trajectory crosses Aspvreten. We only accept cases where small variability of aerosol number in the Aitken and accumulation mode size ranges is present during a 6 h period. This means that the aerosol reaching Aspvreten during this period of time is uniform and the influence from local sources is small. It is also necessary to assure that the advection is stable throughout at least 6 h by investigating the grouping of subsequent trajectories. Figure 1 displays a typical case which fulfils the homogeneity criterion for the size distribution. The 6 h median size distribution and the quartile ranges observed at Aspvreten on 29 August 2000 at the time when the trajectory passed the site are shown.

Figure 2 illustrates typical transport situations during our selected cases. 4 trajectories are calculated with some 100 km around the arrival point, defining a square around the receptor station. The trajectory arriving Värriö is indicated in the middle of this square. Three levels are considered, 10, 100 and 200 m, to assure a stable boundary layer transport. The picture reveals a stable flow from Aspvreten to Värriö. Due to vertical wind-shear a westward shift in trajectory orientation is observed. This shift is however small. By further investigating the flow situation at ± 3 h around the selected transport occasions we assure that no significant changes in transport pattern occur over the period of sampling at the receptor station.

In the base case of the model calculations we use input of gases as given in Table 1. Sulfur dioxide, ozone and nitrogen oxides are estimated as the monthly average concentration of these species during southerly winds as observed at Hyytiälä (data from Hyytiälä, 2001–2003). Hyytiälä is located in the middle of Finland. Since no measurements of gases except ozone is performed at Aspvreten we have to rely on observations from Hyytiälä, assumed to be somewhat representative for gas concentrations observed at Aspvreten. NO_x is partitioned between NO_2 and NO following typically observed diurnal variation. SO_2 , NO_x , O_3 , NH_3 , HCHO , CH_4 and CO is assumed constant during the model runs. HCHO , CH_4 and CO are taken from Janson

et al. (2001). Water vapour is estimated from relative humidity and temperature along the trajectory. Meteorological input such as temperature, pressure and boundary layer height was taken from trajectory model.

Terpenes are emitted from vegetation according to parameterisation in Sect. 2.4 and a yield of condensable products of 0.13 was assumed in the base case. In the base case only chemistry including the major oxidants OH, O₃ and NO₃ is considered. We do not know the actual vapour pressure of the wide range of possible products from terpene oxidation. The saturation vapour pressure over flat surface for the condensable species is therefore approximated to be 3×10^6 (molecules \times cm⁻³). According to Kulmala et al. (1998) similarly low vapour pressure is required to explain the observed growth of freshly formed nano-particles.

In the base case a two-layer structure of the lowermost troposphere was assumed. The lower layer was defined by the boundary layer height as given by the trajectory model. Above this layer a residual layer was assumed to exist. For simplicity, this residual layer height is constant at 2000 m and defines a volume given by the difference in mixing layer height and the upper limit of the residual layer. This approach is reasonable. When the temperature profile alters and boundary layer height decrease parts of the aerosol will be left in this residual layer. Since the mixing in the lowermost free troposphere only is slow, it is rather safe to assume that the aerosol is confined within the residual layer at least during the time scale of days. This residual layer serves as a second reservoir of aerosols. The aerosol will partly be fumigated back in the mixing layer once the mixing layer height increase again. Within the residual layer, the aerosol is only affected by coagulation nucleation and condensation. The air in the mixing layer is in direct connection with sources of organic compounds, and also subjected to dry deposition. The air in the residual layer only get supply of condensable organic vapours and precursors thereof via vertical pumping associated with the diurnal variation in boundary layer height. This means that the growth of the nucleated particles in the residual layer will be slower as compared with mixing layer and further. This treatment naturally is associated with some portion of uncertainty, but as we will

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

see much more realistic than assuming e.g. mixing with zero air or attempting to define an aerosol in the free troposphere where measurements for the location currently not are available. The approach neglects any entrainment from the free troposphere.

3. Results

5 3.1. General size distribution properties

Of the 60 trajectories describing transport between Aspvreten and Värriö, only 12 fulfilled the strict criteria stated in Sect. 2.5. The major branch of trajectories describes transport from a sector NE-SE, over the Baltic states before crossing the area around Aspvreten and from there transport occurs following a somewhat straight line up to Värriö. 4 of the selected trajectories initially describe transport from NW, but the air parcels changes direction in southern Sweden and is subsequently transported north over Aspvreten up to Värriö.

The transport time between Aspvreten and Värriö in the selected cases vary between 50 and 120 h with an average transport time of 85 h (~3.5 days). Data from size distribution measurements at both stations is presented in Fig. 3. Quartile ranges and median distribution for the receptor Värriö and the source station Aspvreten are depicted. At Aspvreten the number concentration is on average 2400 cm^{-3} . Corresponding average mass concentration, assuming a density of 1.5 g/cm^3 , observed at Aspvreten is $3.1 \mu\text{g/m}^3$. The air observed at Värriö after transport contains a substantial lower number concentration, on average 1500 cm^{-3} . Average integrated mass concentration observed at Värriö was $4.1 \mu\text{g/m}^3$. Increase in mass is observed in 60% of the cases. Significant number decrease is observed in most cases. In those cases where number increase is observed, this increase is typically small.

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

3.2. Base case conditions

In the ten selected cases, we ran the aerosol dynamic model UHMA along the trajectories using the 6-hour median size distribution measured at Aspvreten as an input. Since no measurements of the particle composition are available, the initial ratio of sulphate and soluble organics in nucleation, Aitken and accumulation modes is assumed to be 7:3, 1:1, and 3:2, respectively.

The bulk concentration of assumed low volatility terpene oxidation products in these runs are on average (over land) 1.67×10^7 (molecules \times cm $^{-3}$). Corresponding concentration of terpene precursors over land was on average 4.3×10^9 (molecules \times cm $^{-3}$).

Assuming standard conditions, this corresponds to a terpene mixing ratio of 150 pptv on average. Source rate of condensable products from terpene oxidation by OH, O₃ and NO₃ is on average 6.5×10^4 (molecules \times cm $^{-3}$). No discrimination between saturation vapour pressures of different products is done. Considering that the modelled cases are confined to summer and comparing with observed terpene concentrations at different heights in Hyytiälä August 1999 (Janson et al., 2001) and spring 1998–1999 (Spanke et al., 2001) the modelled concentrations are well in agreement with measurements. Daytime reaction of terpenes and SO₂ are dominated by reactions with OH. The diurnal variability of OH is governed by fluctuations in water vapour and solar zenith angle (sza). Maximum of OH is predicted during noon. Daily maximum OH concentration was typically estimated to be $2\text{--}8 \times 10^6$ cm $^{-3}$. Daily maximum of H₂SO₄ was typically $1\text{--}3 \times 10^6$ cm $^{-3}$, with variations governed by latitude and water vapour concentration, pre-existing aerosol concentration and input concentration of O₃, SO₂ and NO_x.

In Fig. 4 four different cases from the model run is shown. In some cases the model result is completely in agreement with observations as in case A (upper frame Fig. 4). The accumulation mode concentration is slightly underestimated for particles of size >250 nm, but the model manages to accurately capture the characteristic features of both nuclei mode and Aitken mode size range. The deficiency of simulated number concentration above 250 nm compared with observations might be result of primary

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

A pseudo-Lagrangian model study of the size distribution properties over ScandinaviaP. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

emissions, unaccounted for by the model. In this case the evolution of the size distribution is characterised by significant growth of the initial Aitken mode. Recalling no primary sources are included in the model, it is obvious that growth by condensation is capable of moving Aitken mode particles up to accumulation mode size range over a time period of a few days. Further, it is obvious that even if nucleation does not result in an overall number increase, addition of freshly formed particles is necessary in order to capture observed features.

This is also evident in case B. The modelled size distribution excellently represents the observed aerosol size distribution for both Aitken and nuclei mode particles, but fails in reproducing the observed accumulation mode. This might indicate an underestimated growth rate in the model or possible primary emissions.

On the other hand, case C excellently captures the accumulation mode concentration, but in doing so the nuclei mode and particles in the lower Aitken mode size range is underestimated. This indicate that the modelled nucleation and subsequent growth is too slow compared with observations, leading to the predicted particle depletion in the sub 50 nm size range.

In Case D the accumulation mode is clearly overestimated, leading to significant mass-increase during transport. In this case it is obvious that the condensation growth is overestimated. Also number representation by the model in nuclei and Aitken size range is poor.

Average model output is depicted in Fig. 5. Median and quartile ranges for Värriö observational size distribution data and modelled results for the 12 selected cases are displayed. The location of the Aitken mode is on average excellently captured by the model. The number concentration is slightly underestimated for smaller size aerosols and overestimated in the 100 nm size range. The number concentration is overestimated on average by 10% and the mass concentration is overestimated 10% on average. The good agreement between average properties of simulated and observed aerosol size distribution indicate that the representation and balance between different dynamical processes is fairly good.

A pseudo-Lagrangian model study of the size distribution properties over ScandinaviaP. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

When investigating single cases, however, it is found that although the model overestimates the mass in most cases, it is underestimated in a few cases. General statistics for the measured and modelled particles in the size range 10–450 nm are shown in Table 2. The four selected case clearly indicate the weaknesses associated with the model. The shortcomings are mainly the result of failure in reproducing concentration of condensable species, as is clearly indicated by miscalculated growth rates. In order to better understand the models response to changes of different parameters the following part of the investigation is dedicated to sensitivity analysis.

3.3. Sensitivity tests

In the following we focus on a single transport case to perform a more detailed investigation of the importance of different dynamical processes controlling the evolution of the size distribution. As reference we use an air-parcel arriving Värriö on 29 August 2000 at 05:00 UTC. The transport time is 77 h. The transport pattern for this case is depicted for three arrival heights in Fig. 2, Sect. 3.1.

Size distribution data for Värriö $t=0$ and Aspvreten $t=-77$ h is shown in Fig. 6. The size distribution observed at Värriö is dominated by accumulation mode particles. The size distribution observed at Aspvreten 77 h earlier contains large amount of Aitken mode particles. A significant shift in size distribution occurs during transport.

In Fig. 7, the modelled time evolution of the aerosol size distribution under base case conditions is shown. Initially the air is transported over the Baltic Sea. Therefore, it is not affected by emissions from underlying vegetation. During this time, there are indications of new particle formation. However, the growth is too slow to significantly alter the number of particles larger than 10 nm. As the air reaches the Finnish coast after 48 h, emissions of terpenes begin to influence the air mass. Over the sea, the lack of organics limited the growth of the freshly formed particles. As the air is exposed to the source of terpenes, an increased growth rate is observed. The organics are able to shift some of the particles up to around 20–30 nm before these particles are scavenged by coagulation. During the last day of the simulation a new nucleation is initialised.

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

In the presence of low volatile organic products, the nucleated aerosol grows rather rapidly. The concentration of pre-existing aerosols is however large, and the growth rate is not sufficiently rapid to move the freshly formed particles into sizes where they will remain as individual particles. The model predicts contributions of particles larger than 10 nm at the end of the run as indicated in Case A (Fig. 4). However, due to the strong sinks present, nucleation only contributes with a small fraction to the total particle number as the air reaches Värriö.

Comparison of the modelled time evolution with time series of aerosol size distribution observations performed at Värriö before and after the arrival of the trajectories is shown in Fig. 8. There is no sign of well defined nucleation as described by Mäkelä et al. (2000) going on during either of the days. However, contributions of small particles are noticed sporadically. Especially, a peak in sub-20 nm particles appears in the afternoon on 29 August. Contribution of small particles is also noticed during morning hours on 29 August. Considering the time scale of removal for such small aerosols, these particles are likely to have been formed recently. This means that the model is in agreement with observations. Even if no pronounced nucleation event is indicated in observations, new particle formation obviously occur close to the receptor site.

Using this day as a reference, a number of different tests were performed in order to examine the processes determining the size distribution dynamics and to test the sensitivity of the model. The selected runs are summarized in Table 3. All these cases should be compared to the modelled base case. In the base case run, the mass concentration is $3.7 \mu\text{g}\times\text{m}^{-3}$ and the integrated number concentration is 1980cm^{-3} . Figure 9 provides a graphical summary of the modelled cases explained in detail below.

3.3.1. Case 1: Dependence on OH/SO₂ concentration

In order to determine the role of photochemical reaction, we tested the model sensitivity to an increased concentration of OH radicals. The source term of OH was increased an order of magnitude. Under these selected conditions, OH concentration reached daily maximum of 5×10^7 . This significantly increases the concentrations

A pseudo-Lagrangian model study of the size distribution properties over ScandinaviaP. Tunved et al.

of both sulphuric acid and especially daily concentration of condensable organic oxidation products. The higher condensable vapour concentrations naturally lead to an increased growth rate of the particles but also to an increased nucleation rate. These changes have dramatic effects on the resulting size distribution: both number and mass increase significantly. The resulting size distribution bears no similarities with the observational data or modelled base case conditions (see Fig. 9, upper frame) due to the major increase in nucleation rate. The resulting number concentration in this case is predicted to 5000 cm^{-3} compared with observed integral concentration of 1400 cm^{-3} . The ternary ($\text{H}_2\text{O}-\text{H}_2\text{SO}_4-\text{NH}_3$) nucleation mechanism used in this model is nonlinear, and even small changes in the input concentration can give rise to large responses in nucleation rate. Similar test was done to SO_2 concentration whose concentration was increased tenfold (corresponding to 1.4 ppb in this particular case). This gave results similar as ten-fold increase of OH concentration.

3.3.2. Case 2: Dependence condensable product yield from terpene oxidation

The yield of condensable products from terpenes is crucially important for an accurate description of the growth of both pre-existing and recently formed particles. It is generally assumed that some additional condensable species besides sulphuric acid is required to explain the growth rate observed in Eularian experiments such as performed at Hyytiälä (e.g. Kulmala et al., 2001). Low volatile products from terpene oxidation are often argued to be possible contributors to the observed growth rate (e.g. Janson et al., 2001). In the base case we used a yield 13%. This is in agreement with estimates from Leaitch et al. (1999) (Canadian boreal forest) who estimated the aerosol mass yield from terpene oxidation to be at least 13%.

Since the terpene aerosol yield is highly uncertain it is necessary to investigate how the system responds to changes thereof. Two cases were selected varying the yield over almost an order of magnitude: one case in which the yield was assumed to be 3% based on the formation on low volatile di-acids from terpene oxidations (Boy et al., 2003 and references therein) and one with a major increase in aerosol yield (25%). In

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

the case of lowering the yield to 0.03 the resulting mass at Värriö would be $1.7 \mu\text{g}\times\text{m}^{-3}$. The mass at Aspvreten ($t=-76$ h) was integrated between 10–450 nm and corresponds to $1.9 \mu\text{g}\times\text{m}^{-3}$. The modelled increase in mass value should be compared with the observed value $6.0 \mu\text{g}\times\text{m}^{-3}$ (assuming $\rho=1.5 \text{g}/\text{cm}^3$). It is therefore determined that if terpenes are responsible for the observed mass, the yield of condensing species must be higher than 0.03. As discussed by Boy et al. (2003) di-acids are necessary to explain the growth of thermodynamically stable clusters but larger saturation vapour pressures may support the growth of larger particles.

Applying the yield of 0.25 gives a resulting mass at Värriö of $6.5 \mu\text{g}/\text{m}^3$. The resulting size distribution is associated with higher number concentration in both Aitken and accumulation mode, obviously different from observational data. The test clearly displays the sensitivity of the model to the estimated aerosol yield. Base case conditions capture the appearance of the observed size distribution fairly well. Assuming correct representation of terpene emissions and accurate estimates of concentration of major oxidants it is therefore likely that yield of condensable products from terpene oxidation is close to 10–15%.

3.3.3. Case 3: Role of dry deposition

In an idealised case, deposition and dilution are the only processes capable of lowering the mass concentration of aerosols in the absence of wet deposition. This means that an accurate description of deposition is required to capture the aerosol properties during transport under clear sky conditions. In order to investigate the sensitivity to dry deposition we tested the model performance by shutting off deposition completely. As a result, the modelled mass at Värriö increased to $4 \mu\text{g}/\text{m}^3$, compared to $3.7 \mu\text{g}/\text{m}^3$ in the base case. Beside the small increase in mass achieved when shutting of deposition there is only minor differences present comparing this case with model runs under base case conditions.

It is concluded that deposition is important; however by neglecting deposition rate we

do not drastically change the result as compared with the base case. This indicates that deposition is of minor importance during this kind of transport and that the sensitivity to moderate changes in deposition rate is small.

3.3.4. Case 4: Role of nucleation

5 In this sensitivity case, we ran the model with nucleation disabled in order to evaluate the contribution of newly formed particles to the resulting size distribution observed at Värriö.

It is found that the resulting mass concentration in this test was $4.0 \mu\text{g}/\text{m}^3$ and the predicted number concentration was 1600 cm^{-3} . Under base case conditions, integral number concentration was predicted to be 1980 cm^{-3} . The main difference in number concentration comparing the two cases is located in the nuclei-lower Aitken size range. It is thus rather clear that nucleation does not significantly change the result compared to the base case, which in turn implies that nucleation does not affect particles of larger size in the modelled case. However, this is not self evident. As shown in Sect. 3.3.1, an increase of the sulphuric acid source term by a factor of ten changes both aerosol mass and number significantly compared to the base case. Thus, the contribution of nucleation to the final distribution is highly dependent on the ratio of precursors and pre-existing aerosol. It is also clear that nucleation likely contributes to the population of small particles during transport.

20 3.3.5. Case 5: Role of coagulation

In order to study the evolution of the aerosol size distribution in the absence of coagulation, we need to assume that no nucleation occurs. Scavenging to pre-existing particles is clearly the most important loss mechanism of newly formed clusters, and thus including nucleation without coagulation would result in unrealistically high particle concentrations. On the other hand, disabling nucleation does not significantly alter the resulting size distribution compared to the base case as was seen in previous section.

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

Thus, in this case we disabled both nucleation and coagulation.

When coagulation and nucleation are switched off in the model, the predicted number concentration is significantly higher than in the base case: 2400 cm^{-3} compared to 1980 cm^{-3} . The location of the predominating Aitken mode is further shifted to smaller sizes. As can be expected, the mass concentration is not significantly changed in this test. In the modelled case, coagulation seems to be the most important mechanism controlling the evolution of the number concentration.

3.3.6. Case 6: Role of residual layer and boundary layer exchange

The boundary layer evolution is crucial for a more realistic representation of aerosol size distribution evolution. In the base case simulation, we applied a rather simplified, yet realistic model of the boundary layer dynamics. The use of a residual layer above the boundary layer along with corresponding fumigation and dilution effects is intuitively reasonable. Furthermore, it is much more accurate than assuming that all aerosol particles are confined to the boundary layer or than mixing with air devoid of particles when the boundary layer evolves during day time. In this section we test how the aerosol size distribution would evolve in absence of mixing.

As shown in lowermost frame of Fig. 9 the resulting size distribution derived from this test is significantly biased towards larger sizes. This results in overrepresentation of aerosol mass compared with base case simulations and produces an aerosol size distribution significantly apart from observations. This is mainly the result of higher terpene concentration since no dilution by mixing occurs. This results in increased aerosol growth if we assume similar yield of low volatile products from terpene oxidation as in base case conditions.

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

4. Discussion

We have explored how the aerosol size distribution evolves as northerly transport over Scandinavia take place. In the selected box-model approach we assume growth of the aerosol to be controlled by low volatile products from oxidation of terpenes and sulphur dioxide. It is obvious that some condensable species in addition to sulphuric acid is necessary to accurately describe the size distribution evolution. In this study this species was represented by some condensable product from terpene oxidation. The idea of a link between aerosol mass accumulation and emissions from the forest offers straightforward connection between the characteristic environment over the Nordic boreal region and observed aerosol properties. Organics may play an important role in contributing to aerosol mass and number with important implications for climate change. Already in 1960 Went recognized the fact that biogenic emissions may significantly contribute to the atmospheric aerosol burden. The importance of biogenic secondary organic aerosol (SOA) in atmospheric chemistry is further emphasized by e.g. Andrea and Crutzen (1997).

During recent years efforts have been put down to quantify the role of naturally emitted organics in the boreal region. Low volatile oxidation products of mono-terpenes have aroused as the most promising candidate for significant gas to particle conversion over the boreal forest (Janson et al., 2001; Spanke et al., 2001). Although it seems rather clear that the mono-terpenes do not contribute to the actual nucleation mechanism, it is likely that degradation of mono-terpenes constitute a significant source of low volatile hydrocarbons necessary to explain the observed growth rate of freshly formed particles (Jansson et al., 2001; Kulmala et al., 2001). Knowledge about the products formed from terpene oxidation however needs to be improved, regarding density, surface tension and saturation vapour pressure. Also to be considered is the actual yield of condensable species from the oxidation of monoterpenes. Controlled experiments indicate a wide range of aerosol mass yields depending on oxidant and type of terpenes (e.g. Odum et al., 1996; Hoffmann et al., 1997). Yield is generally in the order of

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

A pseudo-Lagrangian model study of the size distribution properties over ScandinaviaP. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

a few percent up to 50–60%. Since an explicit knowledge concerning the mass yield is lacking we are thus confined to estimates of the yield under conditions valid over the Nordic countries. Assuming that terpenes actually participate in aerosol formation, it is evident from this study that an accurate determination thereof is mandatory.

5 Nucleation is not predicted contribute to number increase of >10 nm particles to a large extent during northerly transport under selected base case conditions. This was shown both by simulations and observations. Nucleation does occur during the transport cases modelled, but does not result in integral number increase during transport. The potential of nucleation to contribute significantly to the overall number may in simple terms be defined by the relation of precursors of nucleating species versus the
10 pre-existing aerosol (or condensational sink) an increased concentration of either OH or SO₂ would favour a high apparent nucleation rate as shown in Sect. 3.3.1. This in consequence would lead to actual increase in integral aerosol number during transport. However, characteristic values of the SO₂ concentration observed at Hyytiälä in southerly winds ranges from 0.05–0.35 ppbv (25th and 75th percentile, respectively).
15 Thus, concentrations of SO₂ in the order of several ppbv's are not often encountered.

5. Summary and conclusions

We have investigated the evolution of aerosol size distribution as transport over Finland and Sweden from south to north occurs. The analysis is based on a pseudo-Lagrangian assumption. As reference points for model input and output size distribution measurements from two stations have been used. Aspvreten (58.8° N, 17.4° E) served as starting location for our analysis and Värriö (67.46° N, 29.35° E) as the receptor station. The average transport time between the two stations is 85 h. The aerosol dynamics was modelled using University of Helsinki Multicomponent Aerosol model
20 (UHMA, Korhonen et al., 2004). Twelve well-defined transport cases were selected from a one year data set. All cases describe transport under clear sky conditions. The transport mainly occurs over sparsely populated areas. Anthropogenic emissions
25

are likely to give only small contributions along the transport path. As a main subject of interest we have studied what processes determine the size distribution during the transport.

Simulations under base case conditions performed in this investigation show good agreement with observations on average. Number concentration and mass concentration was overestimated by 10% on average. This indicates that aerosol growth as well as other dynamical processes is well represented by the model.

By performing sensitivity analysis of selected parameters we conclude following:

- The effect from dry deposition on the aerosol size distribution is small. By disabling dry deposition only minor changes in simulated aerosol size distribution was observed. We recognize dry deposition as being the only mechanism for removal of mass except dilution under clear sky conditions, but however conclude that the absolute magnitude of dry deposition effects on the aerosol size distribution over all is low.
- By investigating the response to changes in yield of condensable products from terpene oxidation we conclude that the sensitivity to changes in the yield is high. Under base case conditions a yield of condensable products from terpene oxidation of 13% was applied. This gave in many cases a nice agreement between simulated and observed size distributions. Assuming correct representation of terpene emissions and accurate estimates of concentration of major oxidants it is therefore likely that aerosol product yield from terpene oxidation is close to 10–15%.
- Simulations and observations indicate that nucleation continuously occur during northerly transport, showing maximum amplitude during noon. The concentration of pre-existing particles is however large in modelled cases. This result in high condensation and coagulation sink, and consequently only a smaller fraction of newly nucleated particles will reach a size where they will remain as individual particles. Nucleation is however in many cases necessary to explain presence of

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

small particles associated with observational data, but does not contribute to overall increased number concentration of larger particles during northerly transport under modelled conditions. As shown, the sensitivity to sulphuric acid generation rate is however obvious.

- Coagulation seems to be the main processes in controlling the observed reduction in aerosol number concentration in virtually all cases.

In total, although nucleation occasionally occurs, it does not generally lead to number increase during transport from Aspöreten to Värriö. Instead, as indicated by the modelled cases and observational data, transport from south to north will result in a number decrease in the Aitken size range, generally in conjunction with increased mass due to condensation growth of existing particles. The most important mechanism in reducing aerosol number is coagulation. Modelled cases with coagulation disabled showed poor agreement with measured size distributions at Värriö. Although we believe that the aerosol dynamic aspects of the model study are correct, we are likely to need improvement the representation of the boundary layer and the exchange of air within the boundary layer as well as the exchange with the free troposphere.

Conclusively, this investigation have explored the possibilities of using rather simple approaches in order to achieve a Lagrangian perspective to a unique data set comprising size distribution measurements at several stations in the Nordic countries. The approach proved successful. Besides the existing size distribution observations, equally ambitious measurements of chemical properties of the aerosol would greatly improve our current understanding of the determining processes controlling aerosol size distribution, and pave way for a more detailed investigation of the interplay between water vapour and aerosols.

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A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model used in this publication.

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- 30

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

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A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

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A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Table 1. Base case conditions. All units in ppb. (¹Data from monthly average concentration of gases observed at Hyytiälä, ²Terpene emissions from pine and spruce according to Lindfors et al., 1999. ³From Janson et al., 2001).

Parameter	O ₃	NO ₂	NO	HCHO	CH ₄	CO	SO ₂	NH ₃	Terpenes	Meteorological parameters
	30–40 ¹	1–2 ¹	from NO ₂	0.4 ³	1700 ³	150 ³	0.1–0.2 ¹	0.025	temperature dependent emissions ² . 15% yield of condensable products assumed	from trajectory model

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Print Version](#)
[Interactive Discussion](#)

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Table 2. Observed and modelled number (cm^{-3}) and mass concentration ($\mu\text{g}/\text{m}^3$) at Värriö for model runs under base case conditions.

		Case A	Case B	Case C	Case D	Case E	Case F	Case G	Case H	Case I	Case J	Case K	Case L
Mass, mg/m^3 ($\rho=1.5\text{g}/\text{cm}^3$)	Observation	6.0	4.6	4.0	2.2	6.0	4.7	4.0	2.7	6.6	4.9	1.3	2.4
	Model	3.7	3.5	3.8	4.1	2.8	3.0	3.6	4.3	3.2	3.7	3.5	3.8
Number (cm^{-3})	Observation	1435	1182	1777	1364	1172	1378	1554	1238	2035	1799	606	610
	Model	1980	1161	1086	968	1768	1386	1394	1114	1752	1980	1106	1009

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

Table 3. Summary of sensitivity tests performed.

	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
Deviation from base case	OH or SO ₂ increased an order of magnitude	Yield of COV altered between 0.03 and 0.25	Deposition disabled	Nucleation disabled coagulation disabled	Nucleation and dynamics modified	Boundary layer

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

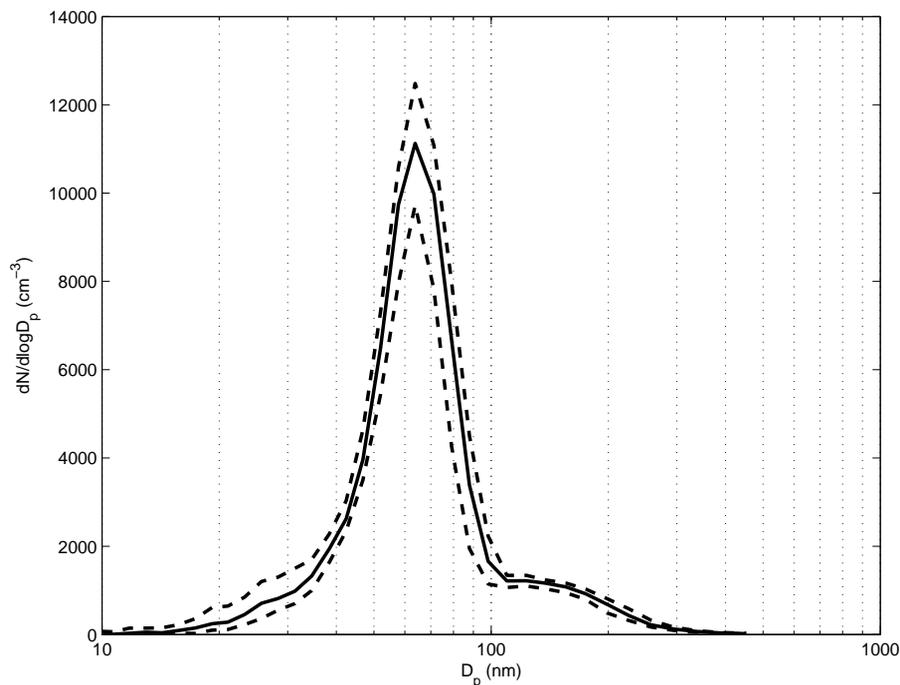


Fig. 1. 6 h aerosol size distribution ($dN/d\log D_p$, cm^{-3}) observed at Aspvreten 02:00 UTC \pm 3 h, 26 August 2000. (75 h before the air arrives Värriö according to trajectory analysis). Quartile ranges indicated by dashed lines.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

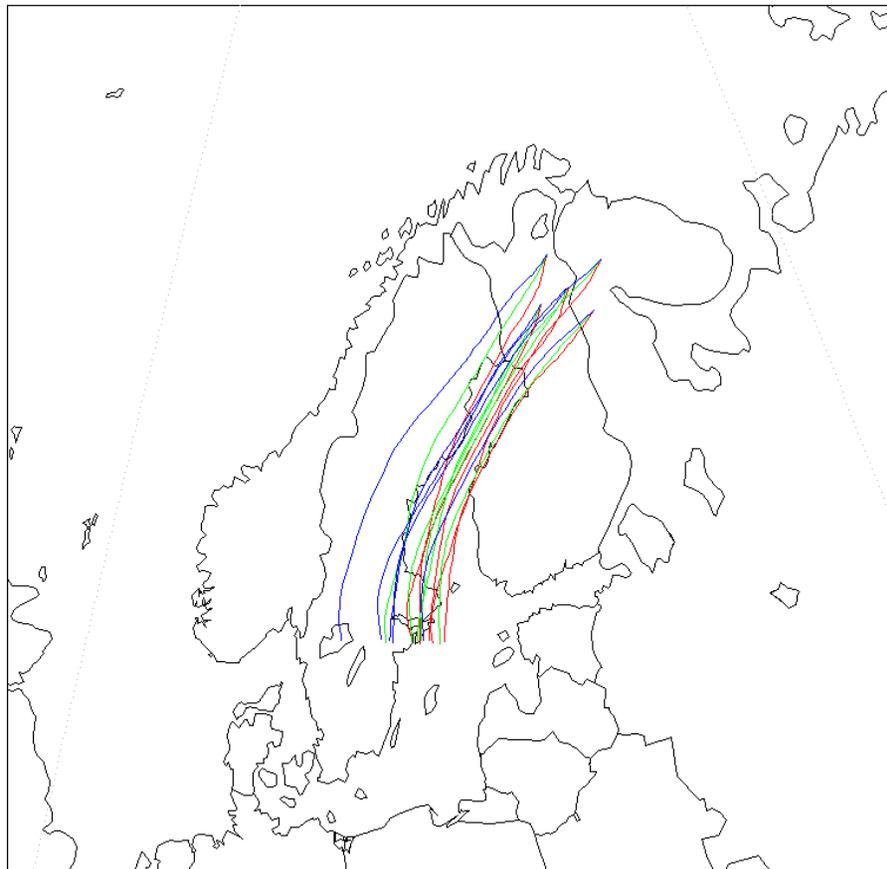


Fig. 2. Transport to Värrö and surroundings during 05:00 UTC, 29 August 2000. Trajectories are calculated at 10 (red), 100 (green) and 200 m (blue) to cover boundary layer transport pattern. Displayed are trajectories calculated for the receptor station Värrö as well as four other trajectories for the same arrival time defining a square around the station.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

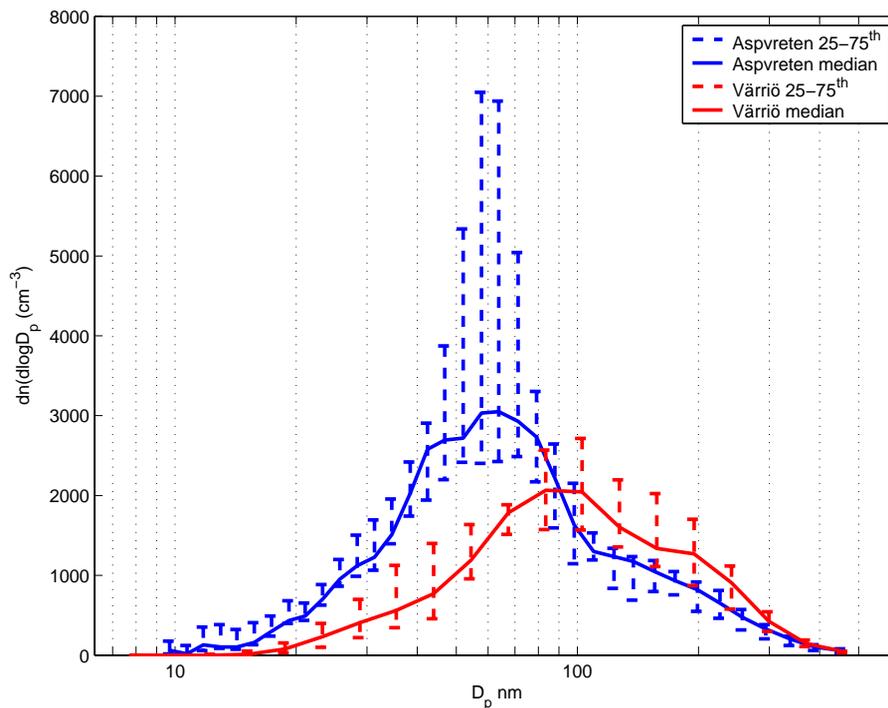


Fig. 3. Size distribution comparison ($dN/d\log D_p$, cm^{-3}) of inter-station transport occasions between Värriö and Aspvreten. Median and quartile ranges are indicated for both stations.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

**A pseudo-Lagrangian
model study of the
size distribution
properties over
Scandinavia**

P. Tunved et al.

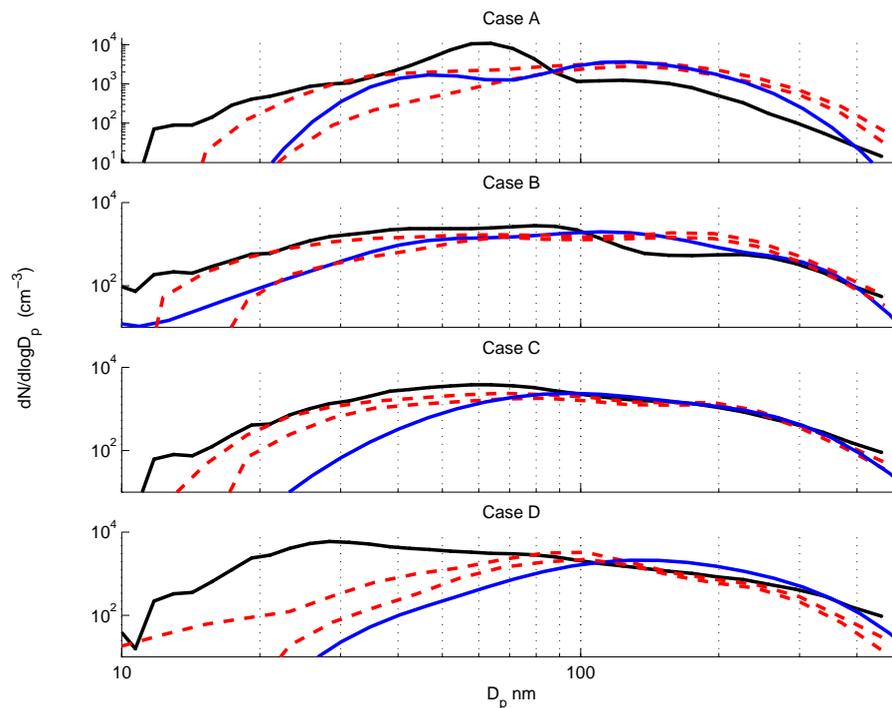


Fig. 4. Three selected cases from the modelled and observed size distribution data ($dN/d\log D_p$, cm^{-3}) resulting from transport between Aspvreten and Värriö. Red dashed line indicate quartile ranges of observational data, black solid line correspond to the initial aerosol size distribution and blue solid line corresponds to simulated aerosol size distribution.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

**A pseudo-Lagrangian
model study of the
size distribution
properties over
Scandinavia**

P. Tunved et al.

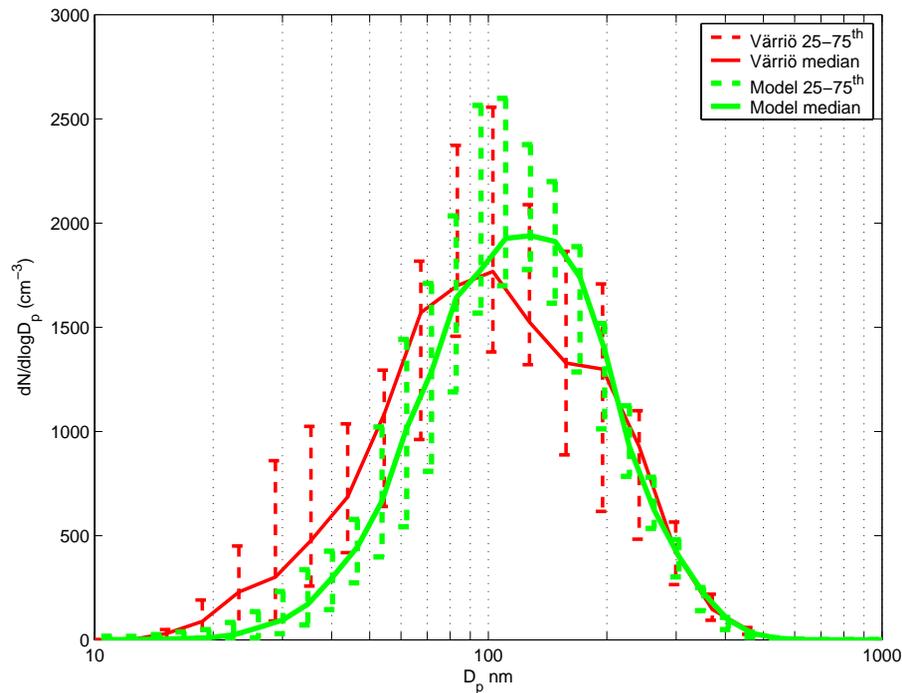


Fig. 5. Median size distribution properties ($dN/d\log D_p$, cm^{-3}) and model prediction of the resulting aerosol size distribution at Värriö. 12 cases considered. Average transport time is 85 h.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

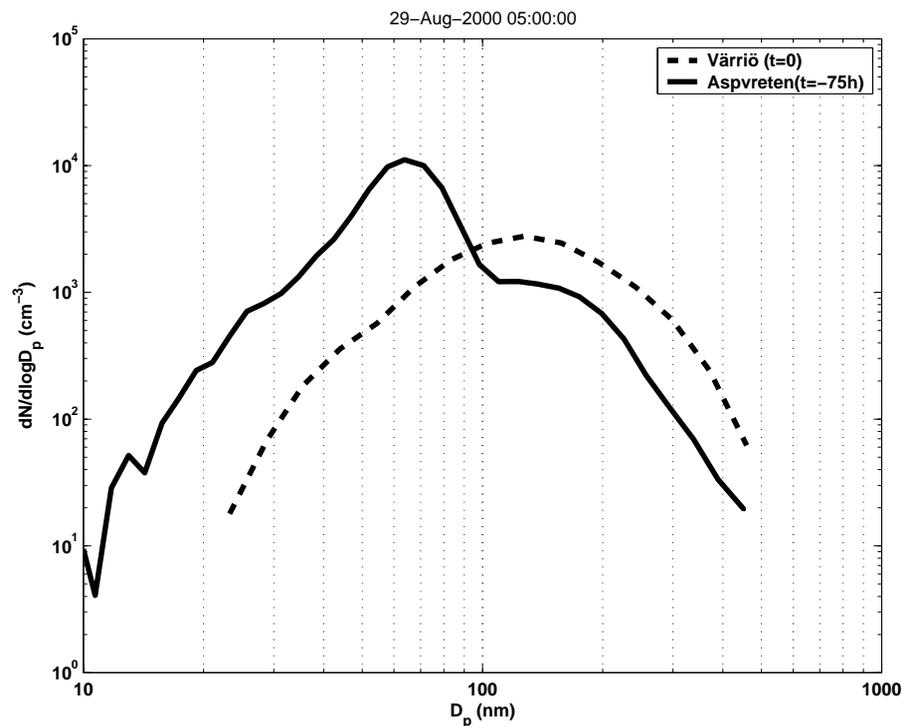


Fig. 6. Observed size distribution at Aspvreten (solid line) 77 h before it arrives Värriö (dashed line).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

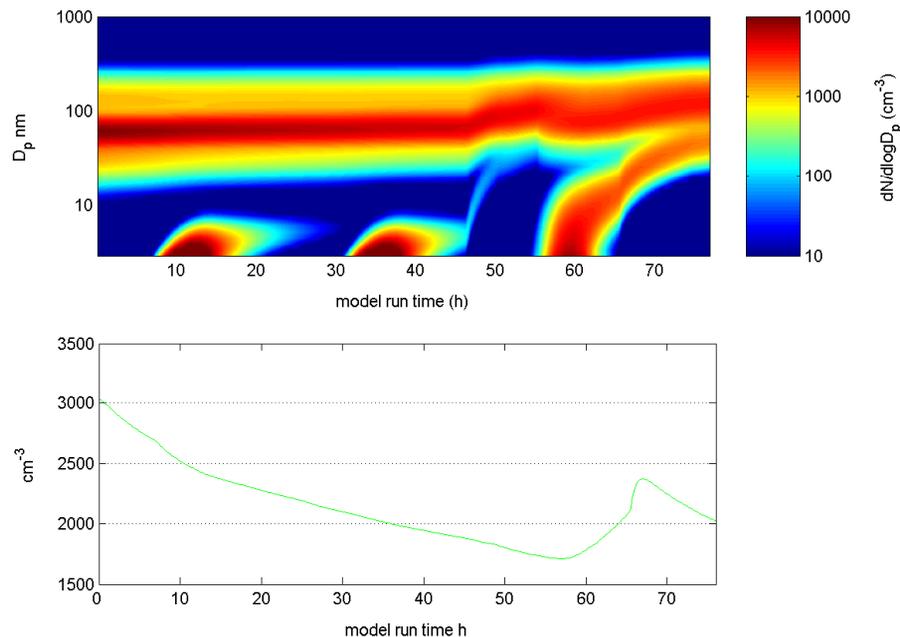


Fig. 7. Modelled evolution of size distribution during transport from Aspvreten to Värriö. The air arrive Värriö on 29 August 2000 at 05:00 UTC. Transport time is 77 h. Lower frame shows time dependent evolution of integral number concentration >10 nm.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

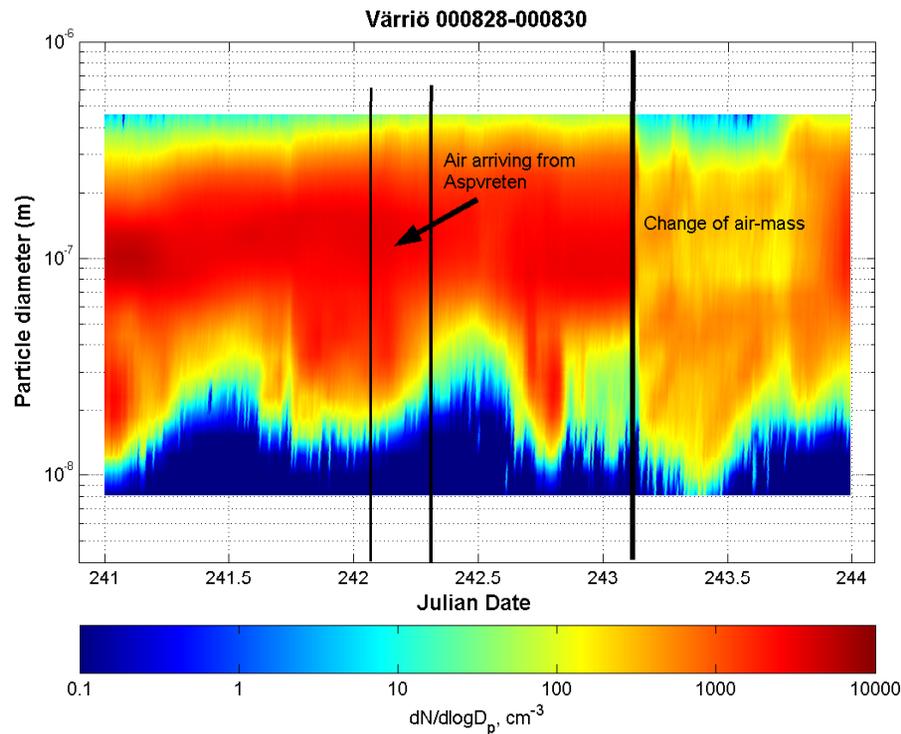


Fig. 8. Observed size distribution at Värriö 28–30 August 2000. The arrival of the air from Aspvreten is indicated by black lines. A change in air-mass is noted during morning hours on 30 August. Freshly formed particles are observed sporadically.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

A pseudo-Lagrangian model study of the size distribution properties over Scandinavia

P. Tunved et al.

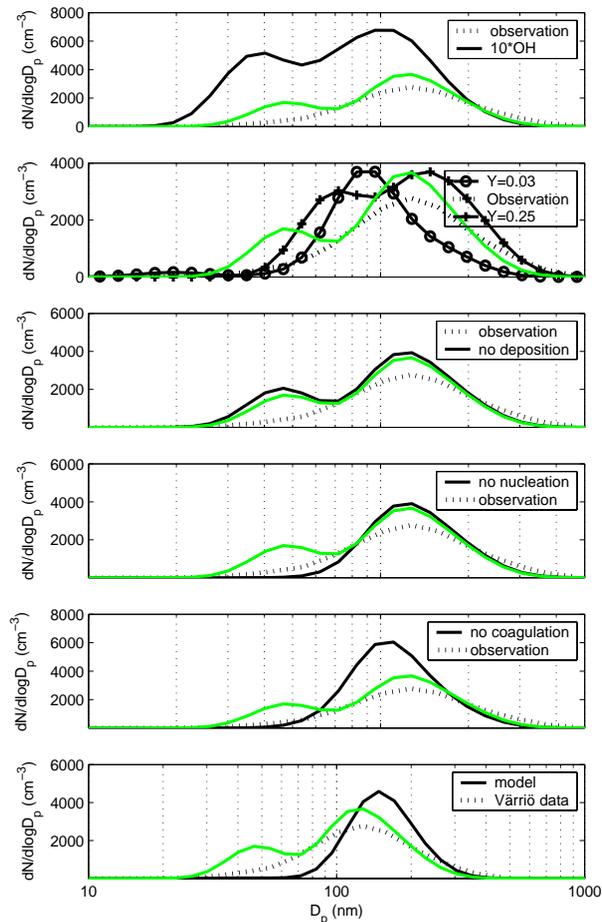


Fig. 9. Summary of results from the six different case studies. Dashed lines indicate observed data and solid line number size distribution as predicted by the model. Green line indicates size distribution derived from simulations under base case conditions.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Print Version](#)
[Interactive Discussion](#)