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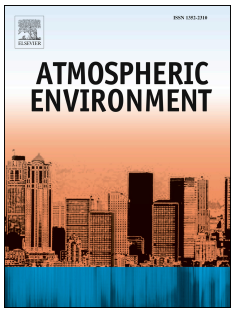

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Titel: Modelling ultrafine particle number concentrations at address resolution in Denmark from 1979-2018 – Part 1: Regional and urban scale modelling and evaluation

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Abstract

The integrated model system DEHM/UBM/AirGIS, developed at Aarhus University, Department of Environmental Science, has been extended with the dynamic aerosol module M7 to account for particle number concentrations of particles with diameters below 1 µm in the atmosphere. The aim of this development is to quantify the spatial and temporal distribution of particle number concentration across Denmark and evaluate the results with available measurements. This article presents model results for particle number concentrations from the regional scale model DEHM and the urban scale model UBM, for comparison with measurements of particle number concentrations from European and Danish measurement stations. The deterministic modelling of particle number concentration has been vitiated by the lack of consistency between emission inventories, and the evaluation of the models is challenged by the lack of consistent long-term measurements data. The performance evaluation of the DEHM and UBM models shows that both models overestimate the level of the particle number concentrations at all stations, however, the results for the correlation coefficients are 0.86 for DEHM and in the range from 0.86 to 0.87, for UBM, for annual mean particle number concentrations at Danish measurement stations. We conclude that the inclusion of particle number concentration in DEHM and UBM shows some capability of reproducing observed patterns, when comparing the results of the models with available measurements, but that there is also room for improvement, especially with respect to the emission inventories and preprocessing of emissions and to the treatment of volatile organic compounds based on natural emissions during summer time.

Introduction

Air pollution – especially ambient particulate matter – is one of the largest environmental risks for human health according to e.g. the Global Burden of Disease study (WHO, 2016) estimating a total number of 2.9 million annual premature deaths attributable to fine particulate matter ($PM_{2.5}$ – particulate matter with a diameter smaller than 2.5 µm) in 2012. Burnett et al. (2018) estimated the global number of premature deaths associated with long-term exposure to ambient $PM_{2.5}$ to be 8.9 million in 2015. The difference between the WHO estimate and the estimate by Burnett et al. can be attributed to the implementation of non-linear relative risk factors in the latter study. The Health Effects Institute characterises air pollution as the 4th leading factor of early death worldwide, and estimates the global number of premature deaths in 2019 to be 6.67 million (HEI, 2020). Numerous international studies have found associations between the concentration levels of gaseous or particulate air

pollutants and various health endpoints including both morbidity and mortality (e.g. Pope et al., 2019; Hoek et al., 2013; Atkinson et al., 2018). Similar studies have recently been conducted in Denmark (e.g. Hvidtfeldt et al., 2019; Raaschou-Nielsen et al., 2020) showing a higher relative risk for mortality at lower concentration levels. With respect to atmospheric particles, most of these studies include the metric particulate matter (PM) measured as mass per volume. This measure is monitored routinely across the world, and is a target of regulation in air quality guidelines and directives. However, it is less relevant for, and does not describe the burden of, the smallest particles with diameters below 1 µm, as their contribution to mass is relatively small compared to the less abundant, but larger particles that constitute e.g. $PM_{2.5}$.

Over the last decades, these small particles have been attracting attention with respect to health effects (e.g. Zhang et al., 2016; Schraufnagel, 2020) and there is a need to investigate the association between ambient concentration levels of these particles and mortality and morbidity. These particles are measured as number of particles per volume, and the result is denoted particle number concentration (PNC). Ultrafine particles (defined as particles with a diameter smaller than 100 nm) constitute a part of PNC, often the vast majority, especially at locations close to the emission sources. In Europe these sources are mainly road and non-road transport (Paasonen et al., 2012; Kukkonen et al 2016). In this study, we use the term PNC/UFP to describe the smallest particles with diameters below 1 µm. In order to achieve a robust metric with respect to measured and modelled concentrations, we in this study follow the approach by Kukkonen et al. (2016) and focus on particles with diameters in the size range 30 - 250 nm, which is here denoted $PNC_{30\text{-}250}$. In this way we both exclude the particles below 30 nm, which is the most uncertain part of the measurements (see below), and make sure to include most of the particles when not considering concentrations very close to important urban emission sectors, such as e.g. traffic in a street canyon.

Indoor contributions to particle pollution in terms of PNC/UFP are significant and come e.g. from cooking, candles and other sources of small-scale combustion inside buildings or cars (see e.g. Bekö et al., 2013; Wallace et al., 2014). Indoor concentrations of PNC/UFP can be of much higher significance for human exposure, than outdoor concentrations, however, the focus of this study is solely on ambient air pollution, i.e. outdoor concentrations. Concentrations of PNC/UFP in the remainder of the paper, therefore refers to outdoor/ambient concentrations.

Measurements of PNC/UFP are scarce, especially with respect to a better spatial distribution over a smaller area (larger city and surrounding area with more than one or two measurement sites). Size-resolved measurements are still relatively expensive, but also the harmonization between measurements in terms of lower (and upper) cut-offs makes intercomparability difficult. In particular, the lower boundary is very important for the number counts, and PNC/UFP particles are more complex to measure. There have been a few approaches presented to model deterministically the PNC/UFP while taking into account the processes that govern particle formation and transformation. These modelling studies have been conducted for different areas of interest and at different spatial resolution. Examples of such efforts on the regional scale are Fountoukis et al. (2012) and Kukkonen et al. (2016) and for the urban or local scale recent examples are also reported by the study of Kukkonen et al. (2016) where PNC/UFP were modelled for five European cities based on the implementation of the M7 particle module (Vignati et al., 2004) and the LOTOS-EUROS model. Karl et al. (2020) modelled PNC/UFP in plumes of ships close to large harbors.

Over the last 30 years, a Danish air pollution modelling system (DEHM/UBM/AirGIS, Brandt et al., 2001a; Jensen et al., 2017; Khan et al., 2019) has been developed, and it is now a standard application for health impact assessment studies involving the Danish population. The system consists of three coupled air pollution models (DEHM: Danish Eulerian Hemispheric Model; UBM: Urban Background Model and OSPM: Operational Street Pollution Model, see details below) operating on regional, urban and street scale, and the system is in this study extended to model particle number concentrations and estimate address-level concentrations for the first time. The model development, setup and evaluation is documented in two accompanying papers. The present paper is Part 1, describing the implementation of the dynamical particle module M7 (Vignati et al., 2004), the methodology for implementing particle number emissions, the coupling between the regional model DEHM and the urban model UBM, and the evaluation of these two models with available measurements of PNC/UFP on regional and urban scale. Part 2 describes the model setup of the OSPM model at street scale level, the extraction of address-level concentrations with the AirGIS system and the evaluation of these local scale model results with measurements on urban and street scale (Ketzel et al., 2021). Part 2 also includes a detailed description of the challenges with respect to measurements and evaluation procedures.

Methods and models

The DEHM/UBM/AirGIS model system forms the basis of the modelling of concentrations at address-level for PNC/UFP in this study. The model system has been developed by the atmospheric modelling group at Aarhus University over the last decades with focus on air pollutants that are relevant and important for human health and the environment. The output of the DEHM/UBM/AirGIS model system, as well as the DEHM/UBM model system without the AirGIS part, has served as estimates for human exposure to ambient concentrations of nitrogen-dioxide (NO_2), ozone (O_3), sulphur-dioxide (SO_2), carbon-monoxide (CO) and $PM_{2.5}$ in a large number of recent epidemiological studies focusing on various mortality and morbidity outcomes (see e.g. Hvidtfeldt et al., 2020; So et al., 2020; Amini et al., 2020; Poulsen et al., 2020; Taj et al., 2020; Cramer et al., 2020 and Raaschou-Nielsen et al., 2020).

The model system consists of a meteorological model and the three above mentioned air pollution models that are run in succession (DEHM -> UBM -> OSPM) to produce modelled air pollution concentrations on street scale level for all major streets (>500 vehicles per day) in Denmark, and on 1 km x 1 km spatial resolution for the rest of Denmark. Emissions for the models are comprised of national and international emission inventories, which are described in detail in the following together with the architecture and setup of the regional and urban scale models.

The performance of the regional scale model DEHM was evaluated by comparing modelled values with available measurements of PNC/UFP from one Danish and three European measurement stations (http://ebas.nilu.no) operating in the regional background. Similarly, the local scale model UBM was evaluated by comparing with the available measurements obtained from three Danish stations measuring PNC/UFP.

Meteorological model

Meteorological input data for the air pollution models are calculated with the Weather Research and Forecast model (WRF) version 3 (Skamarock et al., 2008), which is run for a setup with respect to spatial and temporal coverage and resolution that matches the setup of the air pollution models (see next section for details). The WRF model is driven by EVA-interim meteorological reanalyze datasets (Dee et

al., 2011) from the European center for medium-range weather forecast (ECMWF, https://www.ecmwf.int/) and run locally at computing facilities at Aarhus University.

Regional scale PNC/UFP model

The DEHM model (Christensen, 1997; Christensen et al., 2004; Frohn et al., 2002; Brandt et al., 2012; Brandt et al., 2013; Geels et al., 2012) is a three-dimensional Eulerian atmospheric chemistry-transport model developed to study long-range transport of air pollution in the Northern Hemisphere. DEHM is offline coupled to meteorological data from WRF, and set up for a domain defined on a polar-stereographic projection covering the Northern Hemisphere, true at 60° N (see Figure 1). The model domain has a horizontal spatial resolution of 150 km x 150 km (d01), and includes three two-way nested domains with higher spatial resolution over Europe (50 km x 50 km resolution; d02), Northern Europe (16.67 km x 16.67 km resolution; d03) and Denmark (5.56 km x 5.56 km resolution; d04). The purpose of this setup is to cover intercontinental and regional transport of air pollution while simulating air pollution levels at a relatively high resolution over Denmark. The vertical resolution has 29 layers and extends up to 100 hPa, corresponding to the lowest 12-15 km of the atmosphere. The thickness of the layers is smallest close to the surface and in the boundary layer (where important processes such as emission and deposition take place) and largest in the top of the domain at the high troposphere/lower stratosphere.

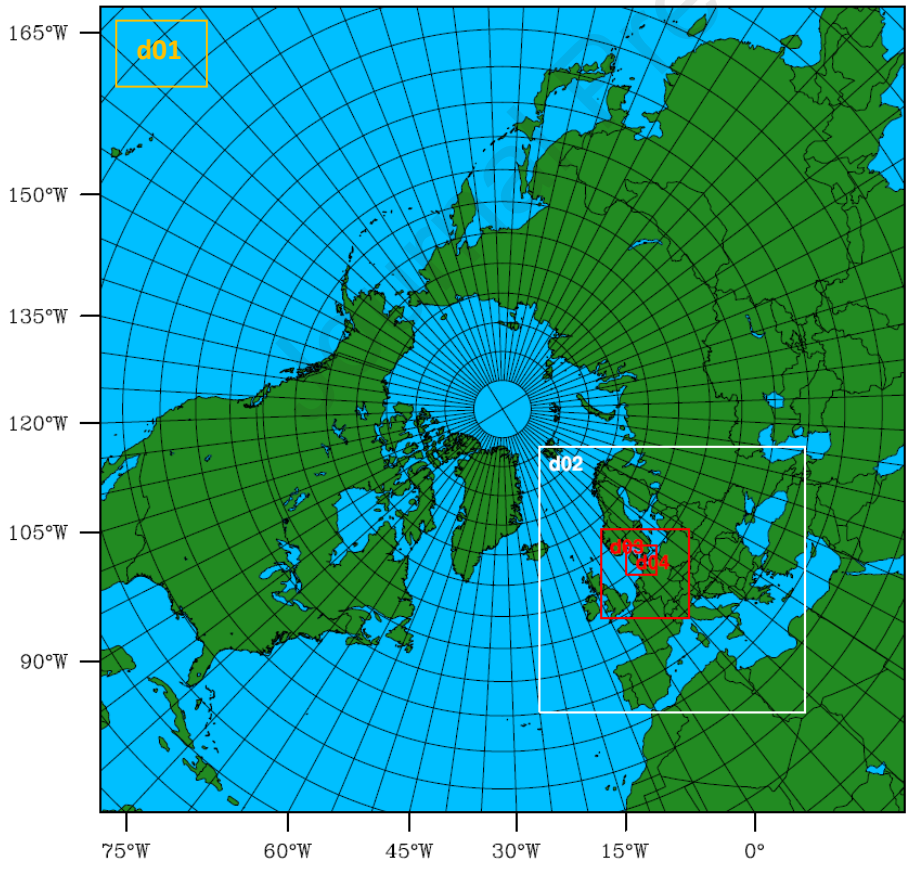

Figure 1: The geographical domains of the DEHM model in the polar stereographic projection. d01 is the main domain covering the Northern Hemisphere with a spatial resolution of 150 km x 150 km, d02 is a

nested domain covering the majority of Europe with a spatial resolution of 50 km x 50 km, d03 covers Northern Europe with a spatial resolution of 16.67 km x 16.67 km and d04 covers Denmark with a target spatial resolution of 5.56 km x 5.56 km.

In the basic setup (prior to implementation of PNC/UFP) the DEHM model calculates atmospheric transport, dispersion, chemical transformation and deposition of 80 chemical components in gas-phase or on particle form based on emissions from global, European and Danish emission databases. The anthropogenic emission data (apart from ship emissions) in the DEHM model are obtained from the Danish national emission inventory model SPREAD (Plejdrup et al., 2018), the EMEP database (Mareckova et al., 2008) and the Eclipse v6b database (Klimont et al., 2017). Emissions from wild fires are based on the REanalysis of the TROpospheric chemical composition inventory (RETRO) compiled by Schultz et al. (2007) for the time period prior to 2003, and from the Global Fire Assimilation System database from 2003 onwards (GFAS, provided by the Copernicus Atmospheric Modelling Service - CAMS). Natural emissions are based on the Global Emissions InitiAtive (GEIA; Frost et al., 2013). Finally, emissions from ships are based on the inventory from the EPITOME project (Geels et al., 2021), which is compiled from data from the Ship Traffic Emission Assessment Model (STEAM, Johansson et al., 2017). The ship emissions have global coverage up to 73°N for the year 2015 and are in DEHM modified with updated emission factors, depending on year, sulphur emission control area (SECA) and trends in shipping activities.

For the anthropogenic emissions, the SPREAD data are used for Denmark and the EMEP data are applied for the rest of the model domain within the EMEP area. Outside the EMEP area, the Eclipse v6b data are used for the rest of countries within the model domain.

The emitted components include nitrogen oxides (NO_X), CO, SO_2, ammonia (NH_3), methane (CH_4) and volatile organic compounds (VOCs) as well as particulate matter ($PM_{2.5}$ and particles with diameter below 10 µm; PM_{10}). The chemical mechanism is explicit (based on Strand & Hov, 1993) and includes eight classes describing particulate matter:

- Particles with diameter < 2.5 µm ($PM_{2.5}$)
- Particles with diameter < 10 µm (PM_{10})
- Total Suspended Particles (TSP)
- Fine fraction sea salt (diameter < 2.5µm)
- Coarse fraction sea salt (diameter > 2.5µm)
- Freshly emitted black carbon (BCf, diameter > 2.5µm)
- Aged black carbon (BCa, diameter > 2.5µm)
- Organic carbon (OC, diameter > 2.5µm)

All secondary inorganic aerosols (SIA) are included in the model, which furthermore calculates natural VOC emissions and secondary organic aerosols (SOA) based on a Volatility Basis Set approach (VBS; Zare et al., 2012; 2014; Bergstrøm et al., 2012). The model is routinely used in a number of applications ranging from operational air pollution forecasting to modelling of health impacts from air pollution and impacts of climate change on future air pollution levels (see e.g. Marécal et al., 2015; Im et al., 2019; Lansø et al., 2015; Hansen et al., 2015; Brandt et al., 2013; Brandt et al., 2012; Hedegaard et al., 2012). The model has been successfully applied in a number of model inter-comparisons and model ensemble studies (see e.g. Simpson et al., 2014; Solazzo et al., 2017).

The gaseous and particulate components are removed from the atmosphere by dry and wet deposition processes. In the DEHM model, the dry deposition is based on the commonly applied resistance method (see e.g. Simpson et al., 2012) where the dry deposition velocity is calculated for different land-use categories. For particulate matter, the dry deposition velocity is also dependent on the size and density of the particles. Wet deposition is accounted for in DEHM by in-cloud and below-cloud scavenging processes for both particulate and gaseous components.

Implementation of PNC/UFP in DEHM

The processes governing the physical transformation of PNC/UFP include nucleation, condensation, evaporation, coagulation, cloud processing and particle growth. The gaseous and particulate components that are already implemented in the model system DEHM/UBM/AirGIS, however, are primarily altered by chemical reactions and deposition processes, and it is therefore necessary to also address the physical transformation processes in the course of modelling of PNC/UFP. An overview of the contributing processes for different size ranges is schematically illustrated in Figure 2, which shows the particle number distribution (in red) as well as the particle mass distribution (in blue) as a function of particle diameter. Most important processes for particles smaller than 0.01 µm/10 nm in diameter ($PNC_{<10}$) is nucleation, condensation onto existing particles and coagulation of existing particles, which increases the size of these particles. Nucleation is especially important at rural sites away from local sources. For the particles with diameter from 10-1000 nm ($PNC_{10\text{-}1000}$) condensation onto the surface of primary particles and coagulation contribute to changing the solubility state of the particles and to increasing their size. Dry deposition is important for $PNC_{<10}$ due to the influence of Brownian diffusion, and important for coarse particles (diameter above ~10 µm) due to the influence of gravitational settling on these particles. With respect to dry deposition as a process, there is a global minimum for particulate matter around 1 µm in diameter. Wet deposition is important for all particles and especially for those that typically serve as cloud condensation nuclei with diameters > ~100 nm. The soluble fractions are subject to in-cloud scavenging and both soluble and insoluble particles are subject to below-cloud scavenging leading to wet deposition.

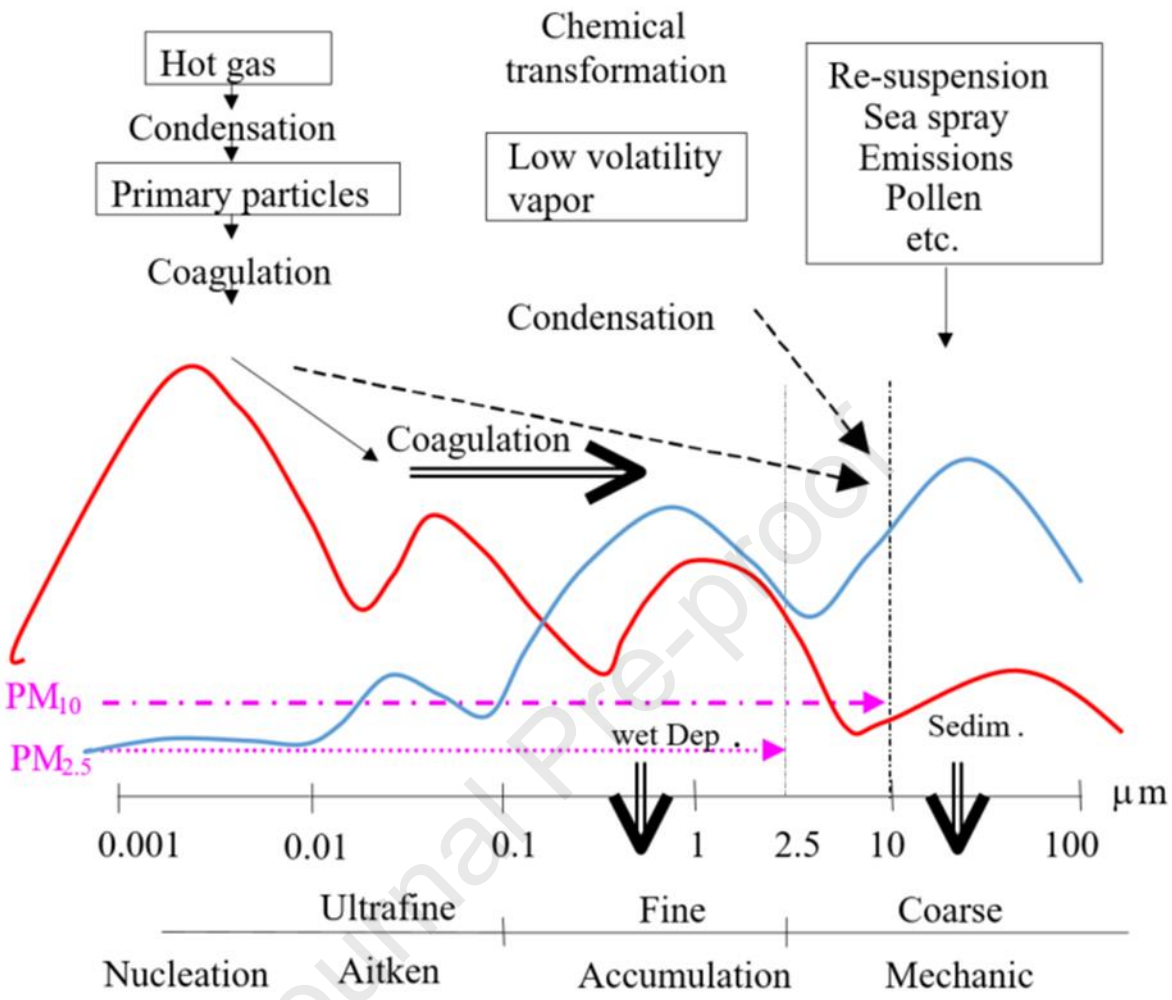

Figure 2. Schematic particle number size distribution (red) and particle mass distribution (blue) over the size range covering the four particle modes included in the M7 module. Text boxes describe the different processes that govern the changes between phases and size modes. Inspired by Whitby, 1976.

In the present study, the M7 particle dynamics module (Vignati et al., 2004) was chosen for implementation in the DEHM/UBM/AirGIS model system. The M7 module has been developed primarily for modelling of air pollution, and has been applied and further developed by other atmospheric modelling groups (Monahan et al., 2010, de Bruine et al., 2019). The most relevant example for the present study is the application in the EU-funded TRANSPHORM project, where PNC/UFP concentrations were modelled in five European cities (Kukkonen et al., 2016).

For this study, the M7 module has been adapted for implementation in the DEHM model. The overall functionality of the M7 module is depicted in Figure 3. The module includes four different aerosol modes as can be seen in the column in the right part of the figure: Nucleation, Aitken, Accumulation and Coarse mode. Furthermore, the processes nucleation, coagulation, condensation, and particle growth are described in the M7 module and indicated by solid and dotted lines and arrows in Figure 3.

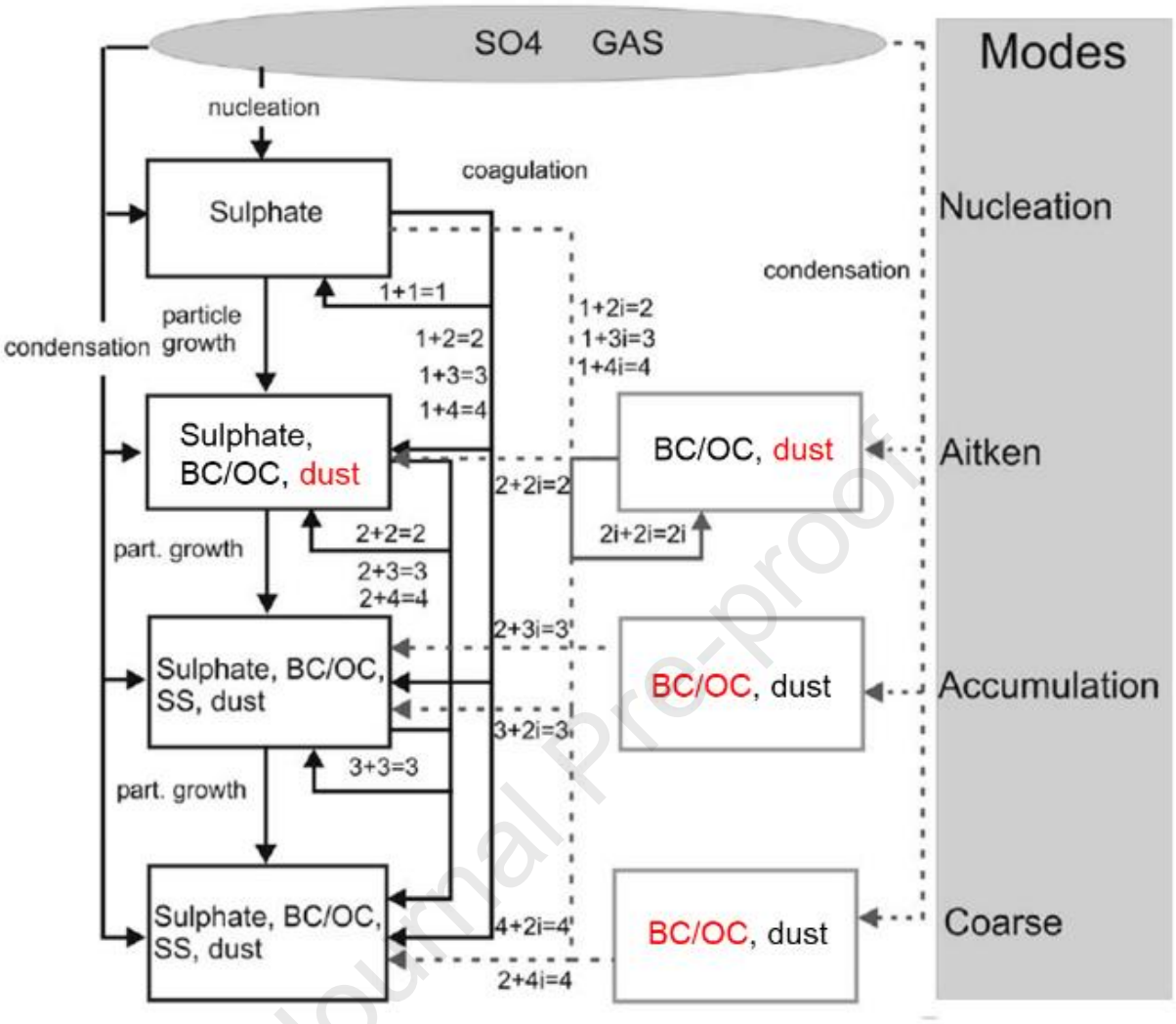

Figure 3. Schematic illustration of the modes and processes included in the M7 module. The seven rectangles indicate the seven modes, left mixed (soluble), right insoluble. In black the contents of the original version of M7, new components included in the version implemented in DEHM are marked in red. Solid lines correspond to processes influencing the mixed/soluble modes, and dashed lines correspond to the processes influencing the insoluble modes. Adopted from Vignati et al., 2004.

The four black-lined boxes in the leftmost column of Figure 3 represent the components that contribute to the four modes in mixed/soluble state, whereas the three grey-lined boxes in the center column represent the components in insoluble state. As sulphate (SO_4^{2-}) only occurs in soluble state, and is the only component in the Nucleation mode in this module, there is no need for a box for insoluble Nucleation mode particles. The solid black arrows represent the processes that lead to particle growth (condensation and coagulation) taking place between the soluble components, e.g. condensation of sulphate in gas phase on Aitken mixed/soluble mode particles, which at some point transfers these particles by growth into the Accumulation mixed/soluble mode. The small numbers next to the arrows in the Figure indicate, which modes are combined in a process, and which mode the resulting particle ends up in. An “i” next to the number indicates insoluble mode. An example is the coagulation of two Nucleation mode particles to form a new Nucleation mode particle (1+1=1) or the coagulation of an

Aitken mode particle and a Coarse mode particle to form a Coarse mode particle (2+4=4). The dotted arrows represent the condensation and coagulation processes taking place in-between the insoluble components as well as the processes, where insoluble particles interact with soluble particles (and thereby automatically transfer to the soluble state). An example is the process of coagulation of one insoluble Aitken mode particle with one soluble Accumulation mode particle, resulting in a (larger) soluble Accumulation mode particle (3+2i=3). For the soluble modes, also water uptake is taken into account, which influences particle diameter.

In Table 1, the components included as number concentration variables in the DEHM model are presented, together with the size characteristics of the modes. All the components are also treated as separate mass concentration variables in DEHM, to retain mass closure in the model during processing. The included components originate from anthropogenic sources; sulfuric acid (H_2SO_4), BC, OC and mineral dust, as well as from natural sources; sea salt arising from wind-generated sea spray, which contributes to particulate matter in the atmosphere. H_2SO_4 particulates are present in all modes, whereas BC and OC only are present in the Aitken (soluble and insoluble), Accumulation (soluble and insoluble) and Coarse modes (soluble only, as it is the primary particles that are insoluble, and they are not emitted in the Coarse mode size range).

Table 1 Size range (diameter) and corresponding components included in the four modes of the M7 module implemented in DEHM. Blue indicates soluble components and red insoluble components. Adopted from Vignati et al., 2004.

Size range for particle diameter in nm	Mixed mode (soluble), i.e. either sea salt, pure H_2SO_4 or PM, coated with H_2SO_4	Insoluble, i.e. non coated PM
0-10	Nucleation (H_2SO_4)	
10-100	Aitken (H_2SO_4, BC, OC, dust)	Aitken (BC, OC, dust)
100-1000	Accumulation (H_2SO_4, BC, OC, sea salt, dust)	Accumulation (BC, OC, dust)
>1000	Coarse (H_2SO_4, BC, OC, sea salt, dust)	Coarse (dust)

The M7 module uses input emissions of SO_4^{2-}, BC, OC, sea salt, and dust in terms of particle numbers for the different modes, corresponding to the different size ranges where such emissions are expected. The process of nucleation is a very complex topic. Many different theories and "nucleation schemes" exist involving different types of precursors. The nucleation scheme used in M7 is relatively simple, just based on the concentration of H_2SO_4. However, nucleation is most important in the rural background with low population density and, therefore, further developing of the nucleation scheme implemented in the M7 module is outside of the scope of the present study.

In the original version of the M7 module, the contribution of dust in the Aitken mode was added to the particle number concentration of OC and the contribution of insoluble OC and BC in the Accumulation mode was added to the particle number concentration of dust, at the expense of the mass conservation for the particle mass concentrations. In the process of implementation of the M7 module in the DEHM model, four new components have, therefore, been added to DEHM and M7 (presented in red in Figure 3) to be able to transfer the emissions of mass tracers as realistically as possible to the size modes. The new components are dust in the soluble and insoluble Aitken mode, and BC and OC in the insoluble Accumulation mode. In this way, mass conservation is guaranteed for all components in the model system.

The dry deposition of PNC/UFP is calculated by combining the size distribution for each mode with size-dependent dry deposition velocities to obtain one weighted dry deposition velocity for the mass concentrations and corresponding particle number concentrations for each mode. When the DEHM model is run with the M7 module included, the chemical model is run simultaneously based on mass concentrations to ensure consistency.

Emissions of PNC/UFP

Emission inventories for particle number concentrations are extremely scarce. A global inventory was developed in the EUCAARI project (Kulmala et al., 2011). The research institution TNO in The Netherlands further developed an inventory covering Europe for the years 2005 and 2020 in the EU-TRANSPHORM project (Denier van der Gon et al., 2014), based on mass concentration emissions and mass to particle number conversion factors on activity level for each SNAP emission category (Selected Nomenclature for Air Pollutants) for the Aitken, Accumulation and Coarse mode particles. The Nucleation mode particles are generated in M7 from sulphuric acid in gas phase. This inventory has a spatial resolution of 7 km x 7 km covering Europe and has previously been applied as input data in connection with a study related to the M7 module (Kukkonen et al., 2016).

Emission inventories for Denmark are very detailed and disaggregated and they are the result of many years of work with the emission model SPREAD (Plejdrup and Gyldenkærne, 2011; Plejdrup et al., 2016; Plejdrup et al., 2018). In order to obtain the best results for Denmark, it is therefore important that the particle number emissions for Denmark fit well with the mass emissions equivalent from the Danish inventories. For this project, we therefore initially used an approach for calculating particle number emissions, by applying the SNAP based particle emission conversion factors from (Kukkonen et al., 2016) to the mass emissions from the Danish SPREAD emission inventory. As the ongoing project intends to study long-term exposure to PNC/UFP, a long time series is needed, and the current standard of the DEHM/UBM/AirGIS system is used to model particle number concentrations at exposure level for the time period 1979-2018. This requires emission input data for the same time span having in mind the short lifetime of aerosols compared to e.g. greenhouse gasses.

As a starting point, we compared the results of the above described approach with the TNO inventory for Denmark for 2005 and found large differences for important emission categories. The results based on SPREAD emissions display particle number emissions that for all SNAP categories are larger than the TNO emissions. For some SNAP categories the difference is very large and for these categories the SPREAD emissions are several orders of magnitude higher compared to the data from the TNO inventory as illustrated in Figure 4. A number of important emission categories stand out, e.g. SNAP1 (*combustion in the production and transformation of energy*), SNAP2 (*non-industrial combustion plants, e.g. domestic heating*), SNAP3 (*industrial combustion plants)* and SNAP5 (*extraction and distribution of fossil fuels and geothermal energy)*. Also, SNAP 9 (*waste treatment and disposal)* stands out with the emissions from the emission conversion factor approach based on SPREAD being much higher than the TNO emissions.

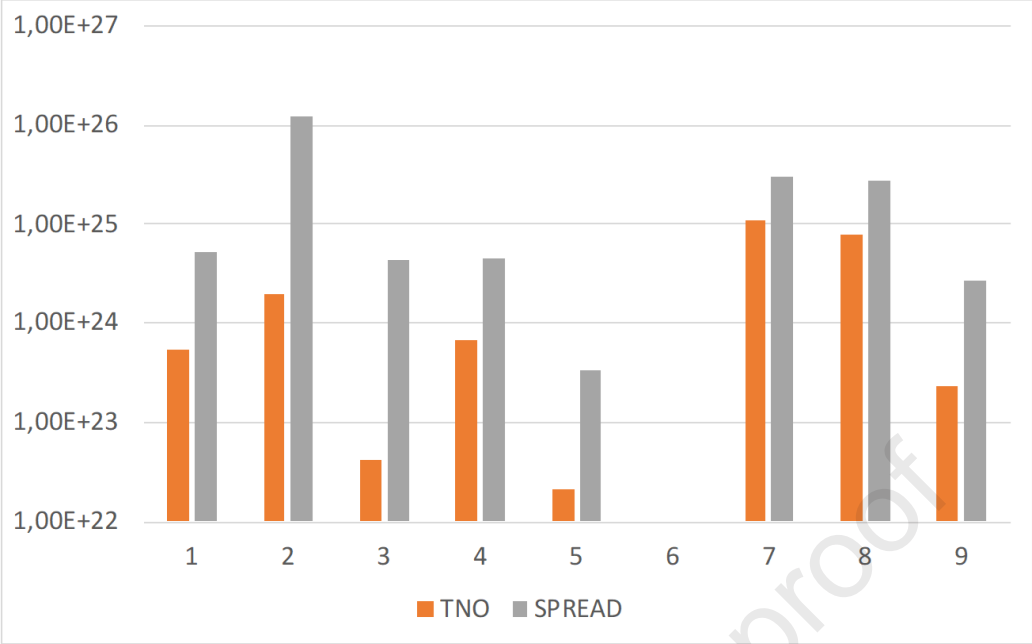

Figure 4. Particle number emissions for 2005 by emission sector (SNAP category) for Denmark calculated for the Aitken, Accumulation and Coarse mode particles. Numbers are calculated based on the SPREAD emission data and emission conversion factors from Kukkonen et al. (2016) (grey) and compared with the corresponding particle number emission inventory for Denmark in 2005 from TNO (orange). Note that the scale on the vertical axis is logarithmic.

To overcome these difficulties and develop a particle number based emission inventory, while still maintaining compliance with the detailed, high-resolution Danish mass based emission inventory (which is needed simultaneously in the model calculations), a scaling method has been developed. An advantage of this approach is that the scaling can also be applied for the other emission inventories needed by the hemispheric DEHM model. The method is based on statistical data extracted from the TNO emission inventory for the relation between particle number emission and particle mass emission for different SNAP categories and size classes (corresponding to the different modes in the M7 module). All emissions are centered in the Aitken, Accumulation and Coarse size modes and the conversion between DEHM particle mass emission (PM) and DEHM particle number emission (PN) for each grid cell is calculated for the three modes using the following equation (here demonstrated for the Aitken mode):

$$PN_{Aitken} = \sum_C \frac{TPN^C_{Aitken}}{TPM^C_{Aitken}} * \frac{TPM^C_{Aitken}}{TPM^C_{Aitken} + TPM^C_{Accumulation}} * PM^C$$

- *TPN/TPM refers to the particle number/mass concentration in the TNO 2005 database*
- *C refers to component C, i.e. BC, OC, dust, sea salt or sulphate*

In this way, the particle number emission of e.g. the Aitken insoluble mode particles in DEHM is calculated as a sum over all components contributing to the Aitken insoluble mode and then scaled with particle number per mass as well as with the Aitken insoluble mode mass per total mass in the original

emission database. Applying this approach in the calculation of particle number emissions, the spatial distribution of the Danish mass emissions, when converting the mass emission values to number emission values, is conserved.

Urban scale PNC/UFP model

The UBM Model (Brandt et al.; 2001a; 2001b; 2001c; 2003) is a Gaussian plume-in-grid receptor model, which in the present study is set up for a receptor net covering Denmark with a resolution of 1 km x 1 km. Dispersion in the UBM model is described as Gaussian plumes in the horizontal direction and linear dispersion from an initial dispersion height (set to 12 m) up to the mixing height in the vertical direction. The model calculates concentrations of particulate and gaseous chemical components contributing to $PM_{2.5}$, PM_{10}, NO_x, NO_2, CO, O_3 and SO_2. UBM includes photochemistry to account for the transformation of nitrogen-oxide (NO), NO_2 and O_3. For the rest of the components, the timescale of chemical transformation is assumed to be longer than the timescale of the transport and dispersion on the high resolution grid within the model. Background concentrations and boundary conditions for all components, in the present study now also including PNC/UFP, are obtained from the DEHM model (as the sum of all the Aitken and Accumulation mode particles, both soluble and insoluble). The background concentrations are added 25 km upwind from all the individual receptor points – in order to avoid double counting of local emissions. The UBM model has been validated in previous studies against all available measurements in Denmark (without PNC/UFP), showing good performance (see e.g. Brandt et al. 2003; Khan et al. 2019; Hvidtfeldt et al., 2019).

The M7 module has not been directly implemented in the UBM model due to the limited high spatial resolution domain and the resulting small timescales in the UBM model, compared to the timescale of nucleation, condensation, coagulation and particle growth processes. Instead, the sum of the Aitken and Accumulation mode particle number concentrations (both soluble and insoluble) has been implemented as a tracer for PNC/UFP in UBM. The concentrations of this PNC/UFP tracer in UBM is based on the input of the corresponding regional scale particle number concentrations originating from DEHM (upwind contribution in each receptor point) and the SPREAD particle mass emissions converted to particle number emissions based on the scaling method described above. The physical processes taking into account the relation to the local scale concentrations of PNC/UFP are, therefore, primary emission, transport and dispersion. Deposition processes are not included in the UBM model.

Measurements available for model evaluation

For Denmark, measurements are available for particle number size distributions (PNSD) from 2002 for two locations (one urban and one street station), from 2005 for one location (rural), and for shorter time periods from two additional locations (one suburban and one street station).

For the latest years, new instruments and inlets at the Danish stations were applied and only measurements for particles with diameters larger than 30 nm in diameter are presented here. For these measurements, losses in the applied DMAs (Differential Mobility Analyzers) for particles < 30 nm were observed. In addition, also losses in the applied inlets are expected especially below 30 nm, but also above. That is why data were reported in Danish reports only for diameters above 41 nm. Here we present data for diameters > 30 nm to be consistent with earlier international studies despite the fact that the uncertainty for the particle number concentrations are high for the particles especially in the range below 41 nm and that the particle number concentrations will be somewhat underestimated. Additional PNC/UFP measurements for stations outside Denmark were retrieved from the EBAS atmospheric

composition database (http://ebas.nilu.no/), which is developed and operated by the Norwegian Institute for Air Research (NILU), (Tørseth et al. 2012). EBAS hosts data submitted by data originators in support of a number of national and international programs ranging from monitoring activities to research projects. Via a web interface it is possible to download both PNC and PNSD data from various locations mostly in Europe and a few places outside Europe. For the purpose of evaluating the performance of the DEHM model, PNC and PNSD measurements performed at three rural sites in Sweden, Germany, and Norway have been utilized. There is a large variety in the setup of mobility particle size spectrometers usually used for measuring PNSD in the submicrometer size range, and data are provided in different size ranges and intervals. In some preprocessing steps $PNC_{>10}$ and PNC_{30_250} have been derived based on the original data. For an in-depth discussion of the methods for extraction of observations for evaluation, see the accompanying paper (Ketzel et al., 2021).

Results

In this section, modelled concentrations of PNC/UFP on the regional and the local scale, the development in the total particle number emission and the average concentrations for Denmark as well as the results of the evaluation of the calculated particle number concentrations predicted by the DEHM and UBM models are presented. Most of the data pre- and post-processing, statistical analysis as well as graphical presentation was performed in R-Studio (version 3.x and 4x; R Core Team, 2021) using a variety of user packages (*openair*, *ggplot*, *plotly* and *shiny*).

Modelled concentrations on the regional (European) scale

The DEHM model was run for the time period 1979-2018. An example of modelled PNC/UFP concentrations from the DEHM model for 2016 is shown in Figure 5 for particles in Aitken mode, Accumulation mode and the sum of Aitken and Accumulation mode. In general, the spatial distribution of the number concentration of the Aitken mode particles across Europe follows the primary combustion sources, such as e.g. ship traffic, major roads and power production, and the Accumulation mode particles display less variability and a smaller contribution from e.g. ship traffic.

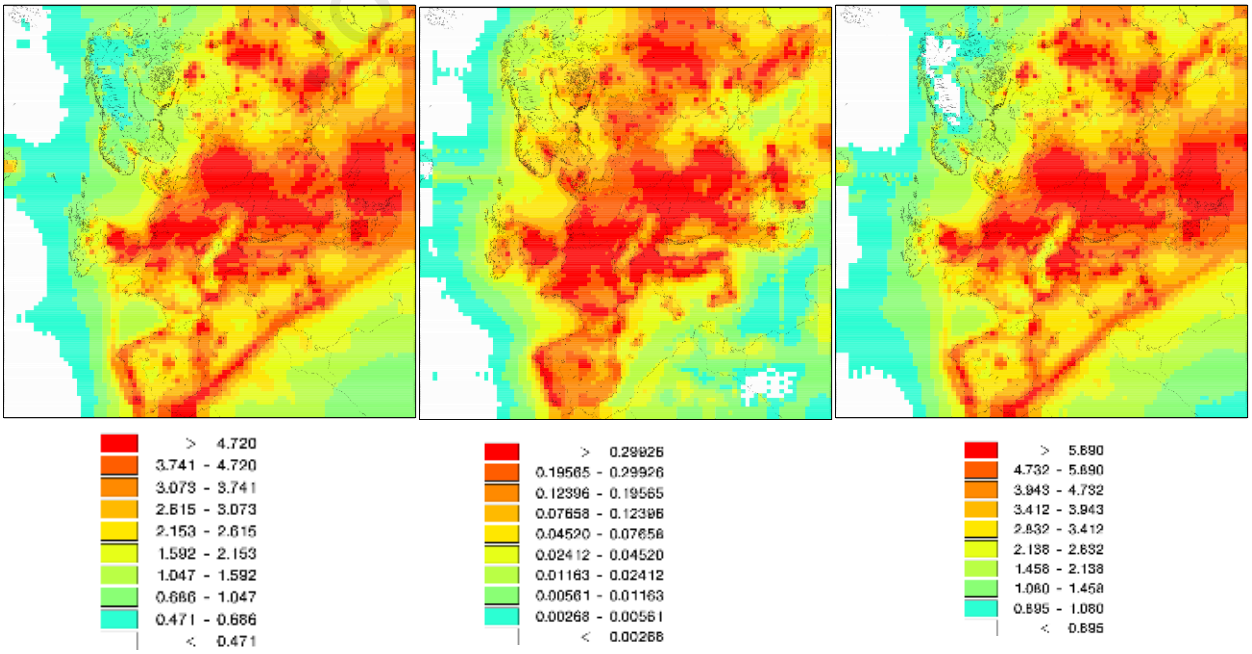

Figure 5. Examples of modelled annual mean PNC/UFP concentrations (10^3 PN/cm^3) calculated with the DEHM model for 2016 for the European domain (50 km x 50 km resolution). Left is the number concentration of particles in the size range 10 - 100 nm corresponding to Aitken mode particles, center is the number concentration of particles in the size range 100 - 1000 nm corresponding to Accumulation mode particles, and right is the sum in number concentration of Aitken and Accumulation mode particles.

In a first step, results from the regional model (DEHM) were compared with measurements from regional background stations in Denmark and Europe measuring submicrometer PNSD. Since the area of interest in this study is Denmark, four stations (rural background only) from Denmark and the neighboring countries Sweden, Norway and Germany were considered. Table 2 presents the Pearson correlation coefficients calculated for hourly, daily, monthly and annual mean concentrations as well as the normalised mean bias for the comparison between measurements of $PNC_{30\text{-}250}$ and model results for the sum of Aitken and Accumulation mode particles for all four stations. A discussion of the statistical parameters for the individual stations is addressed below. For all the plots of comparisons between measurements and model results also the observed number of particles with diameter above 10 nm are shown in the plots (denoted N_10_999). For all measurement stations this corresponds to particles in the size range from 10 nm to the upper limit of the instrument in operation at the station. In the text the expression $PNC_{>10}$ is used, as the equivalent to N_10_999.

Table 2. Values of the statistical parameters Normalised Mean Bias (*NMB*) and Pearson correlation coefficient (*r*) for the comparison of the modelled sum of Aitken and Accumulation mode particles with measurements of PNC/UFP in the size range 30 – 250 nm for the four rural background stations included in the evaluation of the DEHM model. In parentheses is the number of observations included in the calculation of the statistical value.

Station	***r* (hourly)**	***r* (daily)**	***r* (monthly)**	***r* annual**	***NMB (%)***
Lille Valby/Risø (DK)	0.49 (*81272*)	0.57 (*3545*)	0.38 (*153*)	0.86 (*14*)	161
Vavihill (SE)	0.44 (*88933*)	0.53 (*3890*)	0.46 (*166*)	0.65 (*17*)	119
Birkenes (NO)	0.28 (*21722*)	0.40 (*923*)	0.53 (*35*)	-0.05 (*5*)	138
Melpitz (D)	0.33 (*49459*)	0.44 (*2120*)	0.25 (*78*)	0.13 (*7*)	117

Denmark

Results for the rural background station Lille Valby/Risø are shown in Figure 6 and 7. The model results for the DEHM model (in red) are based on the sum of the Aitken and the Accumulation mode particles. They are within a factor of two with respect to the $PNC_{30\text{-}250}$ measurements (in green) with a general pattern of the DEHM model overestimating the PNC/UFP concentration, but following the trend of the measurements. The measured PNC/UFP concentrations show little variation over the day and week and a little more variation over the year with a maximum during the summer season at the regional background station Lille Valby/Risø (see Figure 7). The modelled data includes temporal emission profiles to account for the variation in particle mass and number emissions over the day, week and year e.g. for traffic, to account for rush hours and weekends, or for domestic heating, which relates to more widespread use of wood stoves in the winter season. These temporal variations are also evident in the time variation of the modelled particle number concentrations, which display a maximum during winter and a minimum during summer as well as two daily peaks in the morning and evening, the latter two peaks related to rush hour traffic. These rush hour peaks are not seen equally clearly in the

measurements, which can be due to the distance of respective precursor emission sources and following transport to the observation site, and the rapid processing of particles by coagulation, condensation and particle growth. The correlation coefficients (see Table 2) for the Lille Valby/Risø station are in the range 0.38-0.86 with lowest values for monthly mean concentrations and highest values for annual mean PNC/UFP.

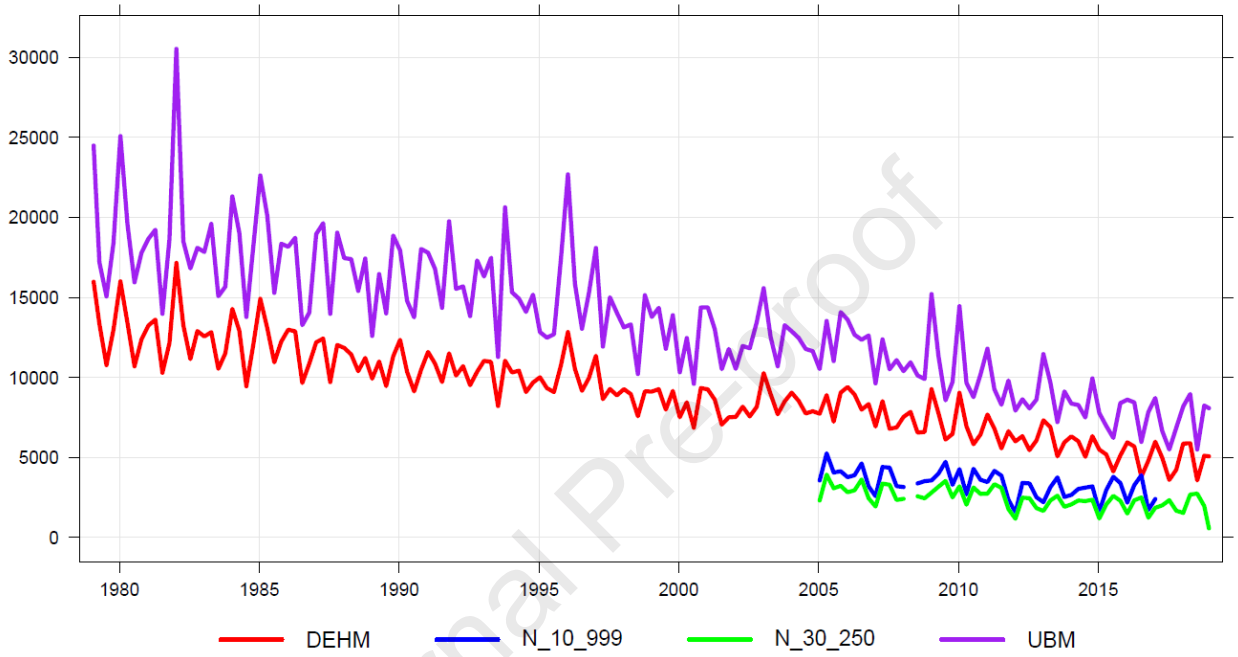

Figure 6. Seasonal average of PNC/UFP [#/cm^3] calculated with the DEHM and UBM models for the rural background site Lille Valby/Risø located in Denmark for the time period 1979-2018. Results are shown for regional scale and local scale modelled sum of Aitken and Accumulation mode particles (DEHM, red and UBM, purple). Furthermore are shown seasonal average of observations of PNC/UFP as number of particles with diameter above 10 nm (N_10_999, blue) and particle number concentration in the size range between 30 and 250 nm (N_30_250, green) for the time period 2005 – 2018 (only up until 2016 for N_10_999 due to the issues with the instrumentation described earlier).

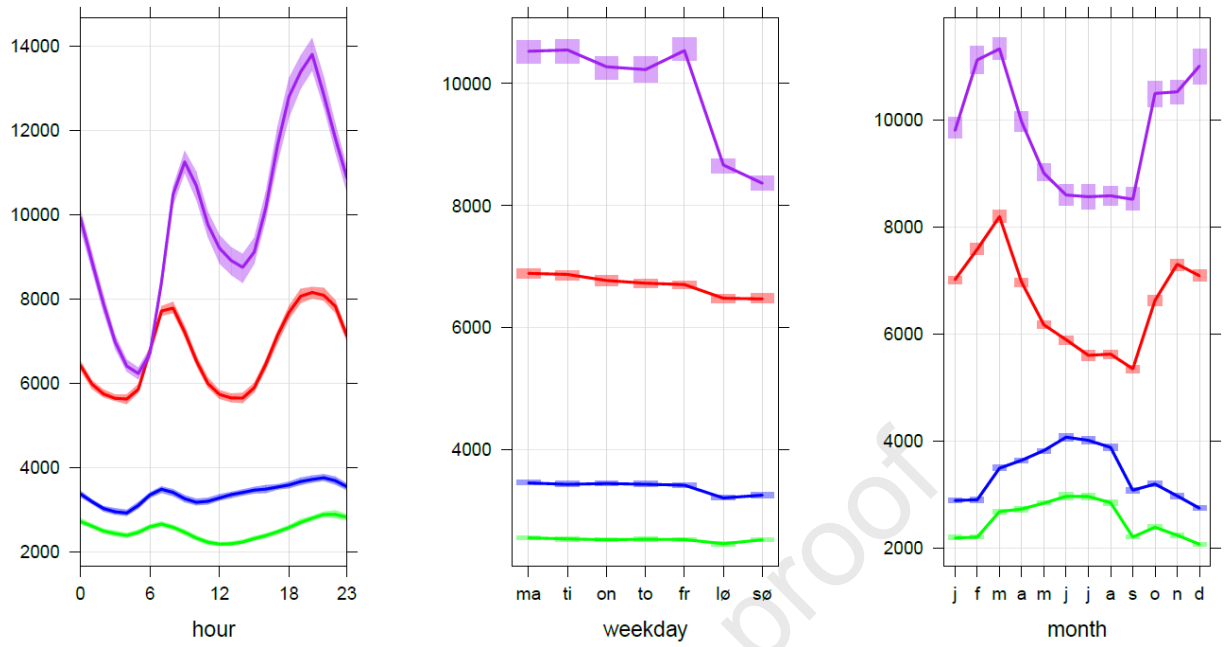

Figure 7. Time variation plot displaying PNC/UFP [#/cm^3] averaged over hour (left), weekday (center) and month (right) over the time period 1979-2018 for the rural background station Lille Valby/Risø located in Denmark. Model results for Aitken and Accumulation mode particle number concentrations using DEHM (red) and UBM (purple) for the time period 1979-2018. Blue line shows measured PNC/UFP with diameter above 10 nm and green line shows measured PNC/UFP in the size range between 30 and 250 nm for the time period 2005-2018 (only up until 2016 for N_10_999 due to the issues with the instrumentation described earlier).

Sweden

Results for the rural background station Vavihill are shown in Figure 8 and 9. The DEHM model overestimates the particle number concentrations with a factor of 1.5 – 2 when compared to the $PNC_{>10}$ observations. In general, there is a strong decreasing trend of the total PNC/UFP concentrations as estimated by the model from around 5000 particles per cm^3 to around 3000 particles per cm^3 (Aitken and Accumulation mode particles only) in the time period from 2002 to 2018, whereas the decreasing trend in the observed $PNC_{>10}$ concentrations is smaller, from around 3000 to around 2000 particles per cm^3. When comparing the modelled PNC/UFP for the sum of Aitken and Accumulation mode particles to measurements within the size range between 30 and 250 nm, the overestimation is increased to a factor of 2 - 3. In Figure 9, the average time variation of modelled and observed PNC/UFP concentrations for hours of the day, weekdays of the week and months of the year are illustrated for the Vavihill measurement site. It is seen that the measured $PNC_{>10}$ concentrations display the process of particle formation (nucleation) in events peaking during the middle of the day. The modelled sum of Aitken and Accumulation mode particles and the measured concentrations of particles between 30 and 250 nm do not include Nucleation mode particles, and hence there is no midday peak. The monthly observed $PNC_{>10}$ and $PNC_{30\text{-}250}$ number concentrations display higher concentrations in spring/summer and lower values in the wintertime as observed for the Danish stations. This pattern is reproduced in the model calculations to a limited extent. The correlation coefficients (see Table 2) for Vavihill for the comparison between the modelled sum of Aitken and Accumulation mode particles and the measured PNC/UFP

concentrations in the size range 30 – 250 nm, are in the range from 0.44 to 0.65, with the highest value for the annual mean concentrations and the lowest value for hourly mean concentrations.

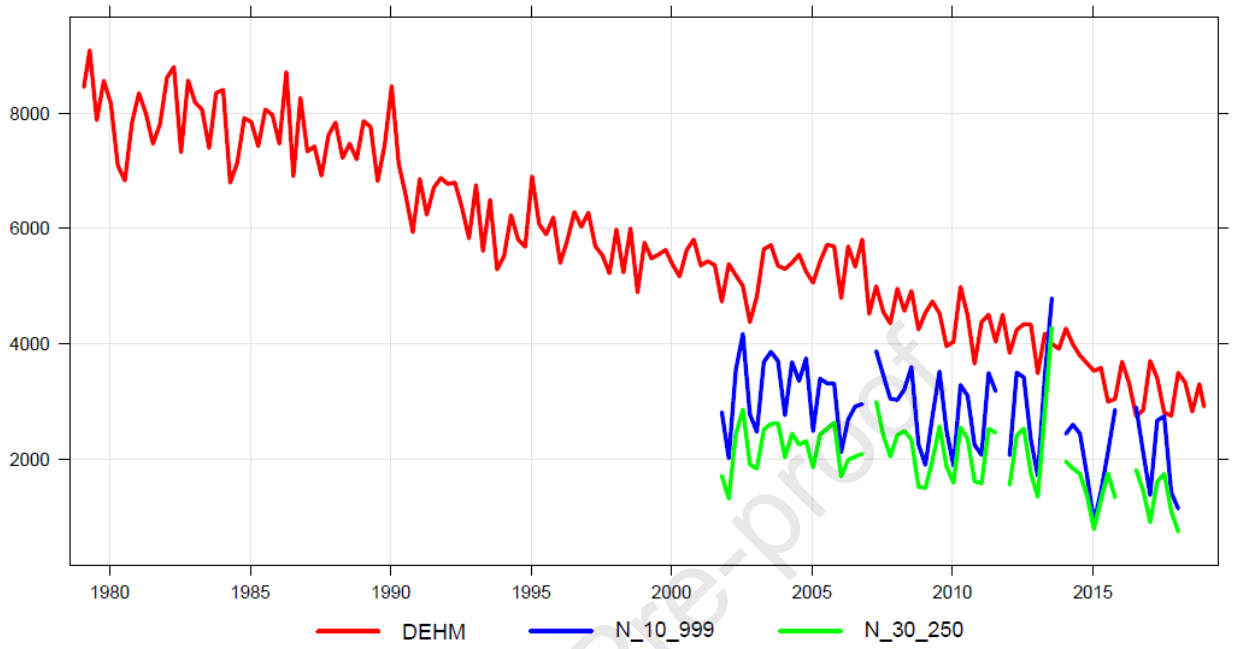

Figure 8. Seasonal average of PNC/UFP [#/cm^3] calculated with the DEHM model for the rural background site Vavihill located in Sweden for the time period 1979-2018. Results are shown for regional scale modelled sum of Aitken and Accumulation mode particles (DEHM, red). Furthermore are shown seasonal average of observations of PNC/UFP as number of particles with diameter above 10 nm (N_10_999, blue) and particle number concentration in the size range between 30 and 250 nm (N_30_250, green) for the time period 2002-2018.

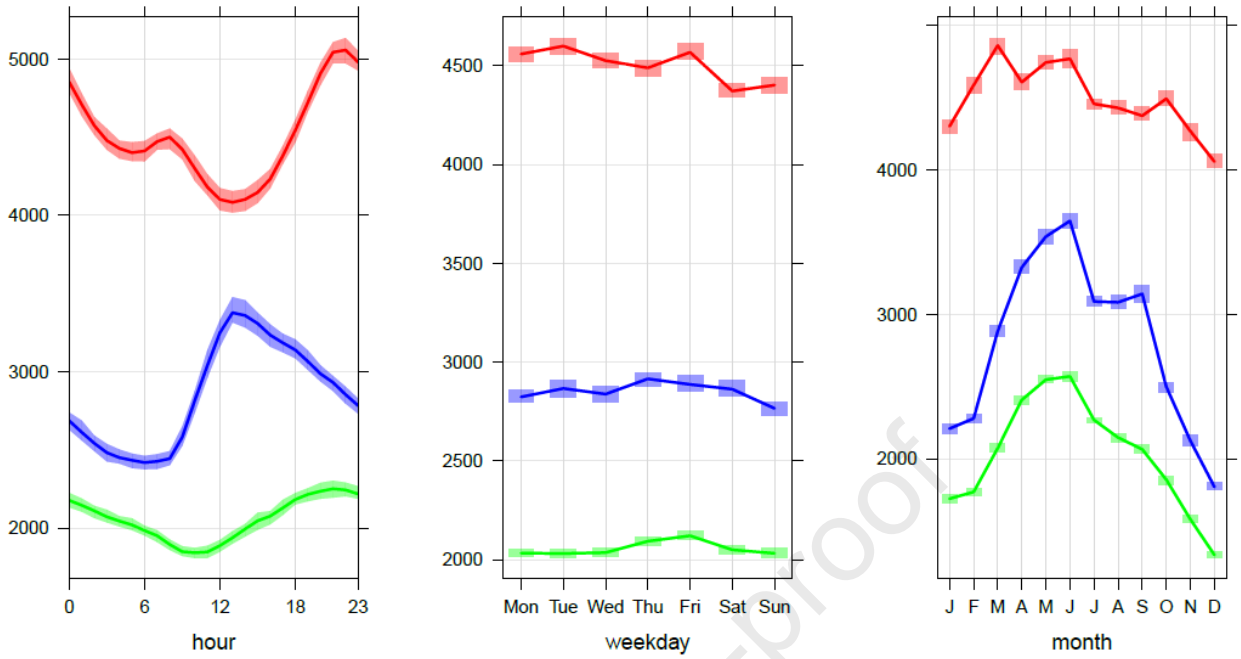

Figure 9. Time variation plot displaying PNC/UFP [#/cm^3] averaged over hour (left), weekday (center) and month (right) over the time period 1979-2018 for the rural background station Vavihill located in Sweden. Model results for Aitken and Accumulation mode particle number concentrations using DEHM (red). Blue line shows measured PNC/UFP with diameter above 10 nm and green line shows measured PNC/UFP in the size range between 30 and 250 nm for the time period 2002-2018.

Norway

Results for the rural background station Birkenes are shown in Figure 10 and 11. There are quite few and somewhat scattered measurements available for this station, and the large variation of the measured concentrations of $PNC_{>10}$ and $PNC_{30\text{-}250}$ is only to a limited extent captured by the model. In general the DEHM model overestimates the measured concentrations with a factor of two to four, however for the years 2006 and 2014 the level of the model results of Aitken and Accumulation mode particles correspond to the levels of the measured $PNC_{30\text{-}250}$ concentrations. The model results display a continuously decreasing trend with large interannual variability, however, the measurements do not show this decreasing trend, possibly due to the limited amount of measurements. The time variation plots in Figure 12 display some relatively small variations over the day and in the week, and the month-to-month variability appears with highest measured concentrations in February and lowest in December. No measurements were available for the month of January and only very few measurements were available for the month of February, hence the comparison with model results for this part of the pattern is difficult. However, it appears that the expected summer time high (due to nucleation events) is represented by the model to some extent.

The correlation coefficients (see Table 2) for Birkenes are in the range from -0.05 to 0.53, with the lowest value for annual mean concentrations (based on only five yearly mean values) and the highest value for monthly mean concentrations.

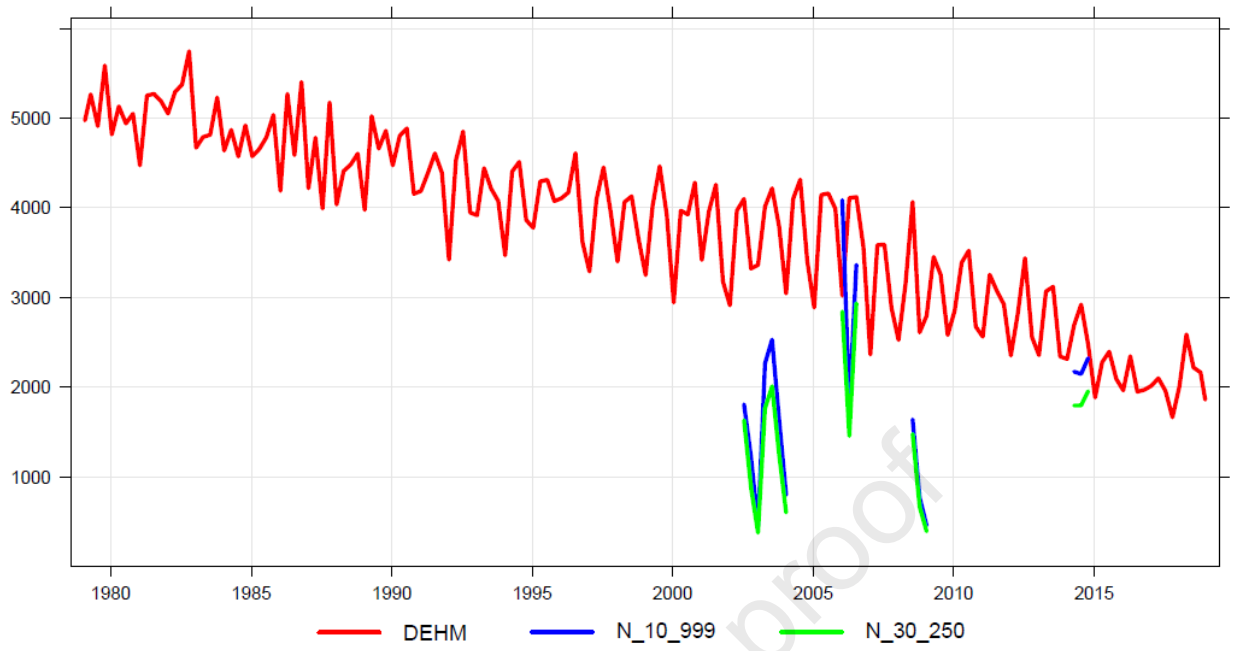

Figure 10. Seasonal average PNC/UFP [#/cm^3] calculated with the DEHM model for the rural background site Birkenes located in Norway for the time period 1979-2018. Results are shown for regional scale modelled sum of Aitken and Accumulation mode particles (DEHM, red). Furthermore are shown observations of PNC/UFP as number of particles with diameter above 10 nm (N_10_999, blue) and particle number concentration in the size range between 30 and 250 nm (N_30_250, green) for the time period 2002-2015.

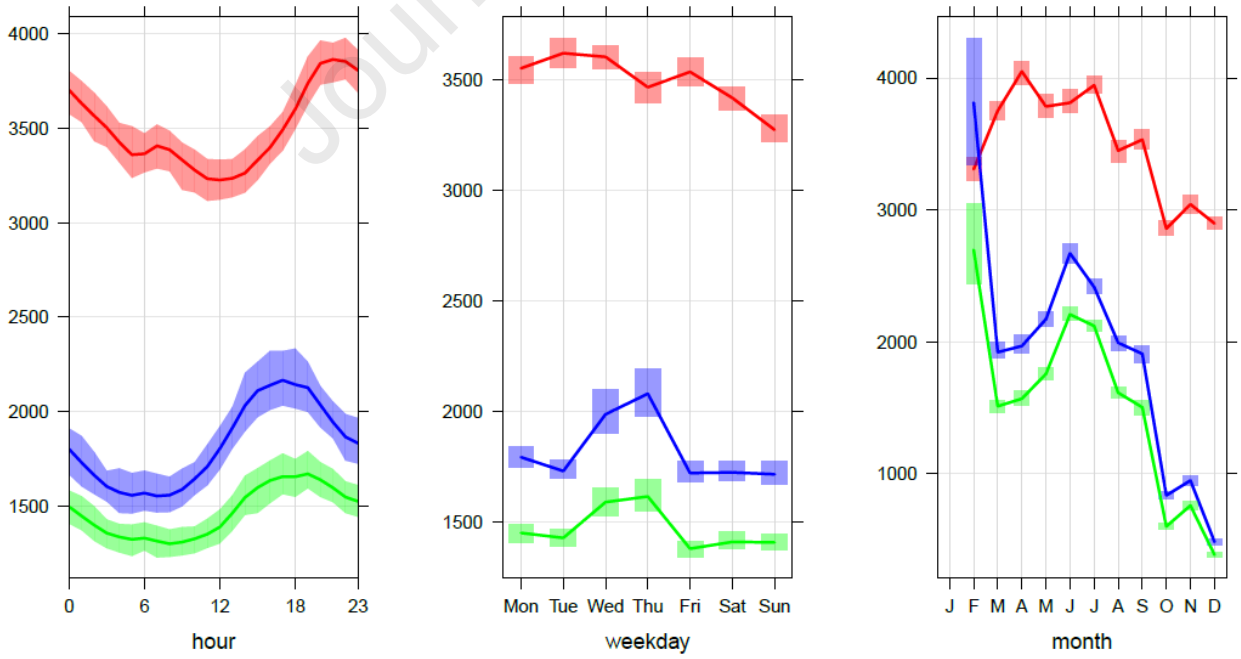

Figure 11. Time variation plot displaying PNC/UFP [#/cm^3] averaged over hour (left), weekday (center) and month (right) over the time period 1979-2018 for the rural background station Birkenes located in

Norway. Model results for Aitken and Accumulation mode particle number concentrations using DEHM (red). Blue line shows measured PNC/UFP with diameter above 10 nm and green line shows measured PNC/UFP in the size range between 30 and 250 nm for the time period 2002-2015.

Germany

Results for the rural background station Melpitz are shown in Figure 12 and 13. For the time period where measurements are available for comparison, DEHM overestimates the PNC_{30-250} concentrations when compared to the modelled sum of Aitken and Accumulation mode particles with a factor of two to three. Figure 13 left, shows a midday peak in measured $PNC_{>10}$ concentrations, corresponding to the highest nucleation values during midday, whereas the modelled sum of Aitken and Accumulation mode particles and the measured PNC_{30-250} concentrations do not display this feature. The weekday variation is relatively small in both modelled and measured values, consistent with a rural background site. The variation of the model results over the year shows an opposite trend compared to the measured values, with modelled values high in winter and low in summer, whereas the measurements (of both $PNC_{>10}$ and PNC_{30-250}) are highest during the summer period and lowest during the winter, due to nucleation events in spring and summer. This is also reflected in the correlation coefficients (see Table 2), which are in the range from 0.13 to 0.44 with the lowest values for the annual mean and monthly mean concentrations and somewhat higher values for daily and hourly mean PNC/UFP.

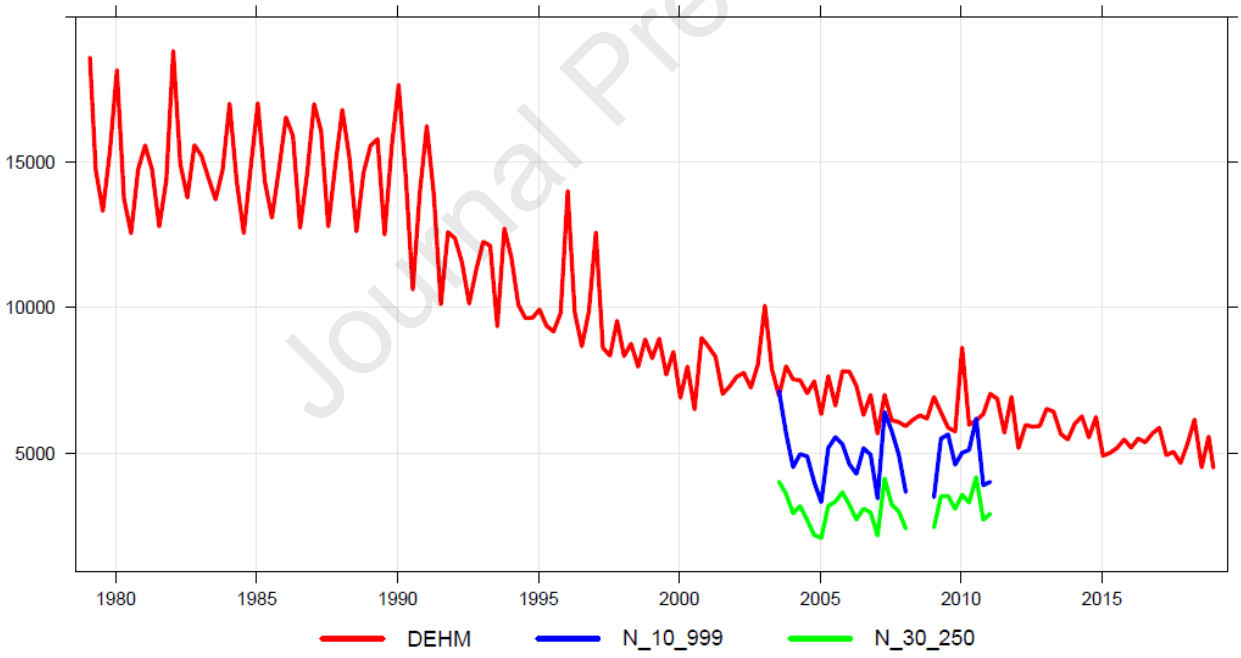

Figure 12. PNC/UFP [#/cm^3] calculated with the DEHM model for the rural background site Melpitz located in Germany for the time period 1979-2018. Results are shown for regional scale modelled sum of Aitken and Accumulation mode particles (DEHM, red). Furthermore are shown observations of PNC/UFP as number of particles with diameter above 10 nm (N_10_999, blue) and particle number concentration in the size range between 30 and 250 nm (N_30_250, green) for the time period 2003-2011.

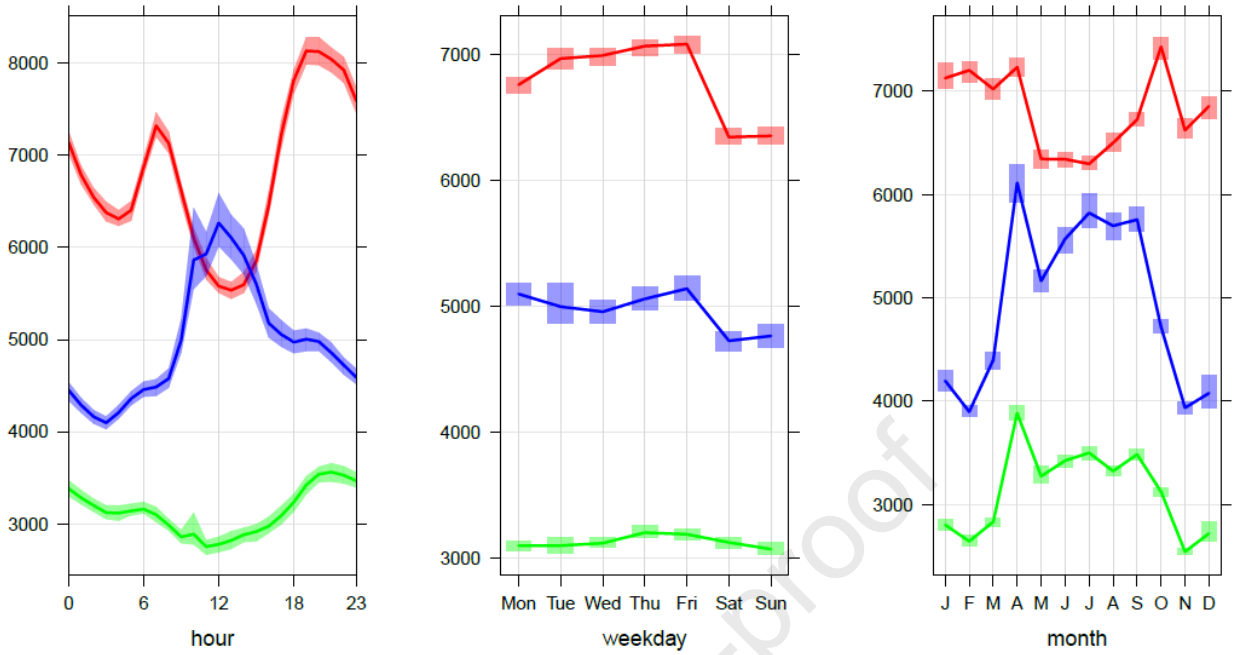

Figure 13. Time variation plot displaying PNC/UFP [#/cm^3] averaged over hour (left), weekday (center) and month (right) over the time period 1979-2018 for the rural background station Melpitz located in Germany. Model results for Aitken and Accumulation mode particle number concentrations using DEHM (red). Blue line shows measured PNC/UFP with diameter above 10 nm and green line shows measured PNC/UFP in the size range between 30 and 250 nm for the time period 2003-2011.

Modelled concentrations on the urban background scale

The development in the total emissions per year of PNC/UFP, BC and SO_2 for Denmark as applied in the UBM model is presented in Figure 14. All three components display a reduction over the time period 1990-2018, and the PNC/UFP decreasing trend in these emissions follows the emissions of BC. Furthermore, the sharp decreases in SO_2 emissions following the introduction of the sulphur emission control areas (SECA) in 2010 and 2015 are also seen in the PNC/UFP emissions due to the influence of sulphur emissions forming particulate sulphate.

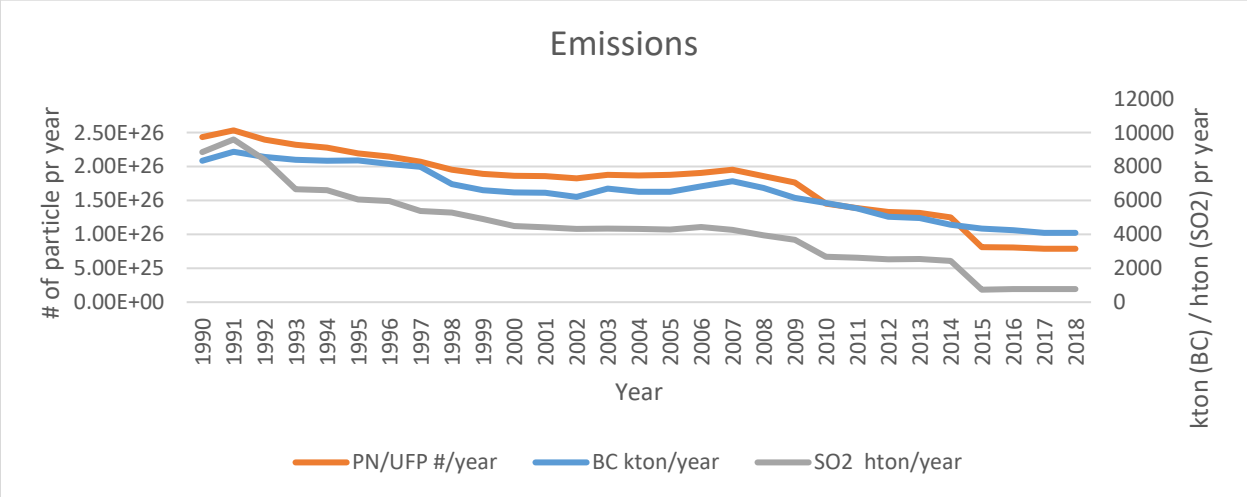

Figure 14. Danish emissions of PN/UFP (sum of Aitken mode and Accumulation mode particles, orange), BC (blue) and SO_2 (grey) for the time period 1990-2018. Note that PN/UFP corresponds to the left vertical axis, whereas BC and SO_2 corresponds to the right axis. SO_2 emissions have been scaled to fit the axis interval, hence different units are displayed.

Modelled annual mean concentrations of PNC/UFP as the sum of Aitken and Accumulation mode particles for the years 1990 and 2018 using UBM are presented in Figure 15. The spatial distribution of particle number concentrations for 1990 reflects the international ship traffic intensity combined with common particle sources in the larger cities. Also, major roads identifying vehicular traffic are visible. In 2018, the pattern has changed and the level of PNC/UFP concentrations has decreased to approximately one-third of the concentration level in 1990. The distribution of particle number concentration in 2018 primarily presents the larger cities as dominant emission sources for PNC/UFP and roads across Denmark, with a significantly smaller contribution from ship traffic.

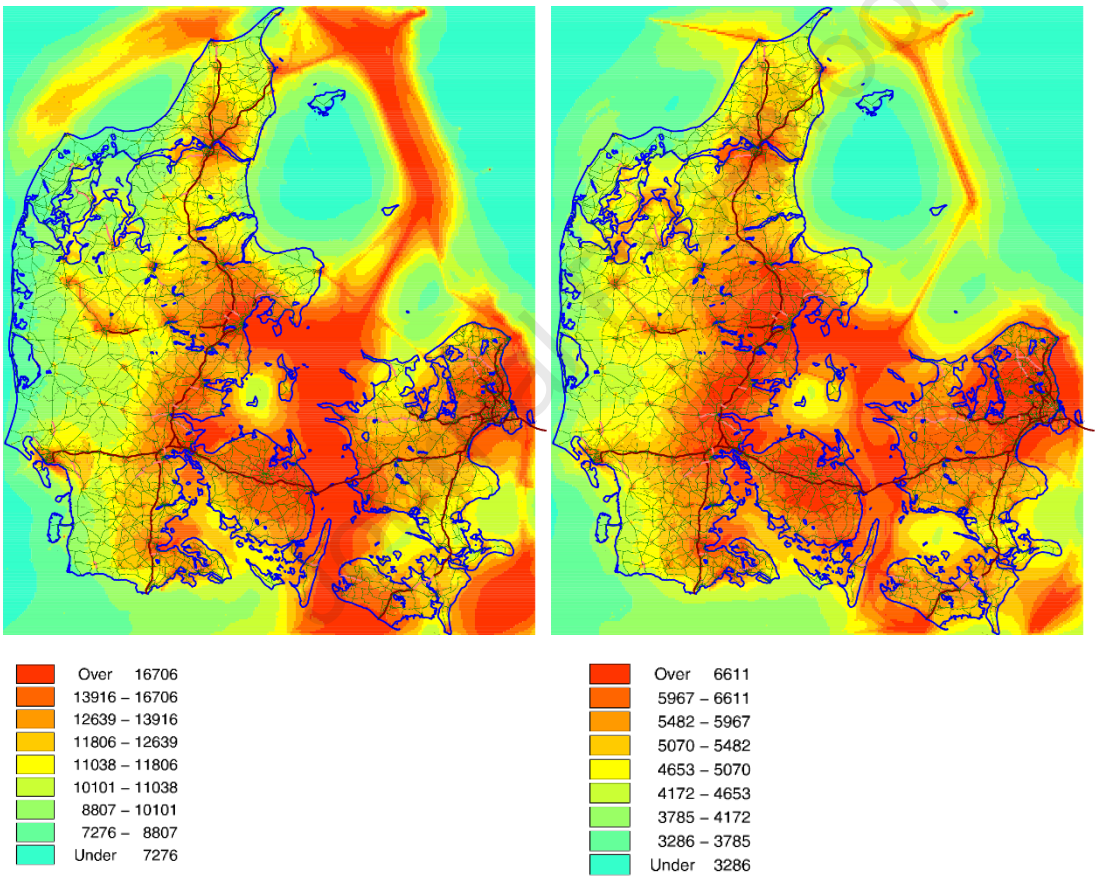

Figure 15. Modelled annual mean PNC/UFP concentration (sum of Aitken and Accumulation mode particles) calculated with the UBM model as number of particles per cm^3. Left 1990 and right 2018. Note that the legend is different in the two plots, due to the large changes in concentration levels.

The modelled annual mean concentration of PNC/UFP as the sum of Aitken and Accumulation mode particles as well as the annual mean concentration of SO_2 and $PM_{2.5}$ averaged over the entire UBM model domain for the time period 1990-2018 are shown in Figure 16. The decrease observed in the Danish emissions in Figure 14 is reflected in the trend for the concentration levels estimated by UBM.

The variations and therewith patterns in PNC/UFP concentrations follow those of both $PM_{2.5}$ (which includes BC) and SO_2 in Figure 14 and the SECA implementation stands out for both SO_2 and PNC/UFP with a drop in 2015 in the estimated concentration levels.

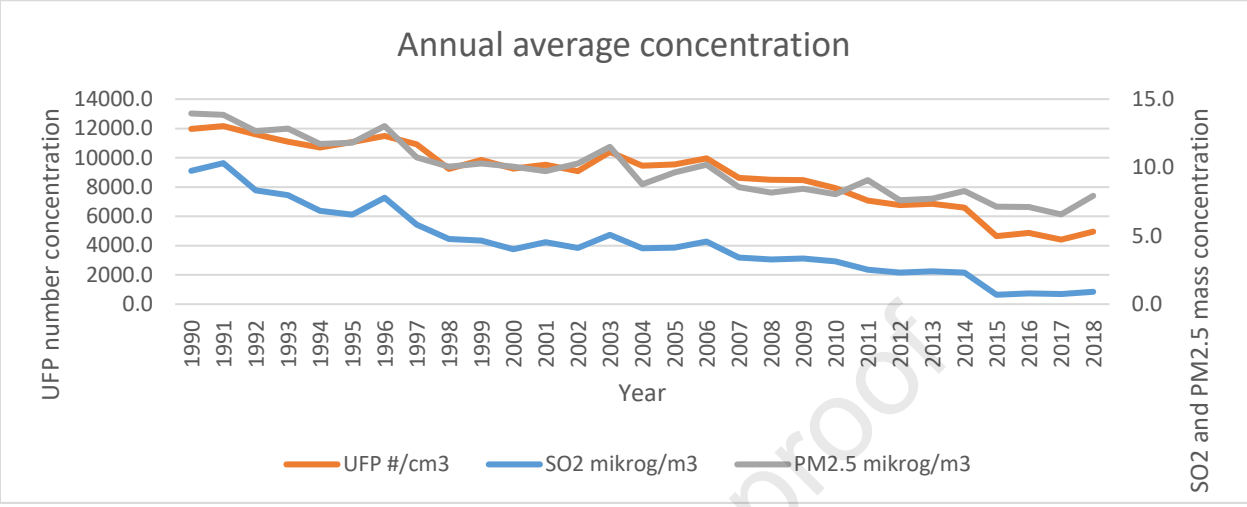

Figure 16. Modelled annual mean PNC/UFP (sum of Aitken and Accumulation mode particles, orange), SO_2 (blue) and total $PM_{2.5}$ (grey) concentrations averaged over the entire UBM domain for the time period 1990 - 2018. Left vertical axis corresponds to PNC/UFP number concentrations and right vertical axis corresponds to SO_2 and $PM_{2.5}$ mass concentrations.

In the second step of the evaluation, results originating from the local scale model (UBM) are compared with measurements from the urban background station HCØ, located at roof-top level in central Copenhagen in Denmark, the regional background station Lille Valby/Risø located some 30 km west of Copenhagen, and the suburban background station Hvidovre, located in the outskirts approximately 5-7 km west of Copenhagen. Figures 17 and 18 show the modelled particle number concentrations of PNC/UFP from UBM and the measurements of PNC/UFP for all particles with diameters larger than 10 nm (N_10_999) and corresponding to the size interval 30 – 250 mn (N_30_250) for HCØ. The corresponding results for Lille Valby/Risø are presented in Figures 6 and 7, and the results for Hvidovre are presented in Figures 19 and 20. For all stations, it can be concluded that the UBM model overestimates the concentrations of PNC/UFP with a factor of app. two to three, which is not surprising, as the background concentrations for UBM are taken from DEHM, which already overestimates these concentrations at the rural background stations with a factor of 1.5-4. The model also includes all particles in Aitken mode, and this most likely additionally contributes to the overestimation, as some of the Aitken mode particles in the model will have a diameter smaller than 30 nm.

The model tends to exaggerate the amplitude of the diurnal and weekly variations at all three sites, even though the timing of the variations is captured relatively well. These variations are in the model dominated by road traffic sources. The annual variation is seen to be anti-correlated with respect to the measurements for all three sites, where the model results predict the highest concentrations during winter and the observations show the highest concentrations during spring/summer. In the model, the high concentrations during winter are due to emissions from wood stoves, which is a major source for primary particles in Denmark, but this is not seen in the measurements. The maximum during spring/summer in the measurements could indicate that wood burning contributes less to the PNC/UFP

formation than anticipated, and that particle formation from VOC emissions from vegetation could be an important process, however, not presently included in the models.

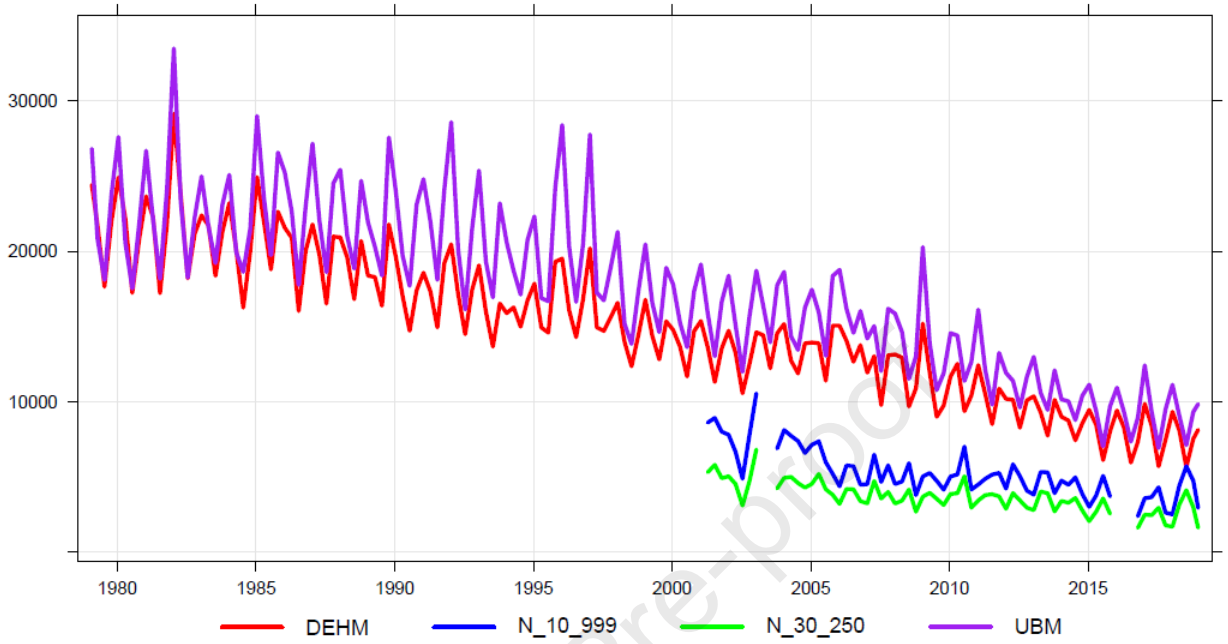

Figure 17. PNC/UFP [#/cm^3] calculated with the DEHM and UBM models for the urban background site HCØ located in Copenhagen in Denmark for the time period 1979-2018. Results are shown for regional scale and local scale modelled sum of Aitken and Accumulation mode particles (DEHM, red and UBM, purple). Furthermore are shown observations of PNC/UFP as number of particles with a diameter above 10 nm (N_10_999, blue) and particle number in the size range between 30 and 250 nm (N_30_250, green) for the time period 2001 - 2018.

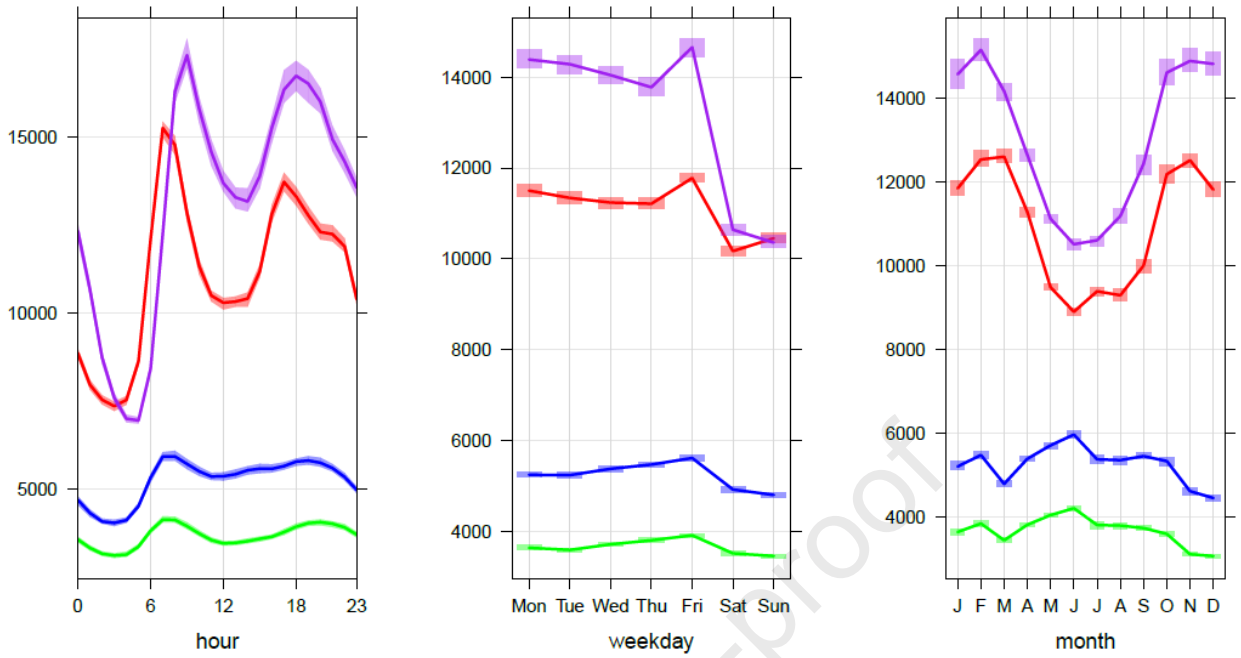

Figure 18. Time variation plot displaying PNC/UFP [#/cm^3] averaged over hour (left), weekday (center) and month (right) over the time period 1979-2018 for the urban background station HCØ located in Denmark. Model results for Aitken and Accumulation mode particle number concentrations using DEHM (red) and UBM (purple) for the time period 1979-2018. Blue line shows measured PNC/UFP with a diameter above 10 nm and green line shows measured PNC/UFP in the size range between 30 and 250 nm for the time period 2001-2018.

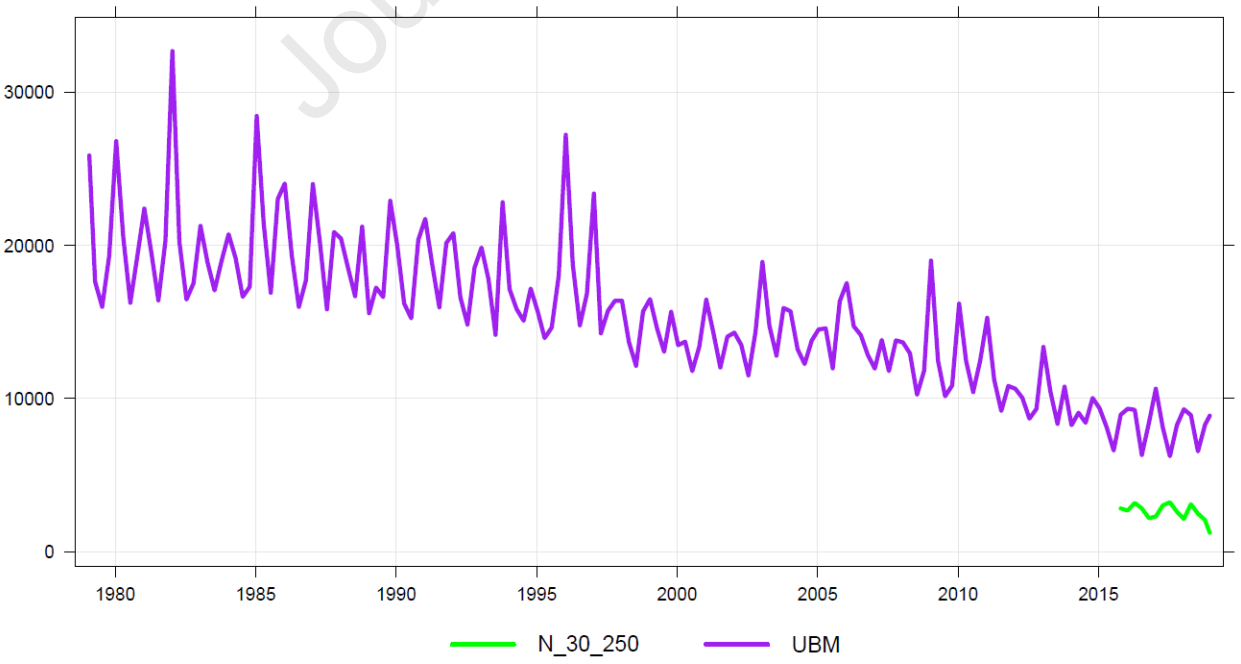

Figure 19. PNC/UFP [#/cm^3] calculated with the UBM model for the suburban background site Hvidovre

located in suburban Copenhagen in Denmark for the time period 1979-2018. Results are shown for the local scale modelled sum of Aitken and Accumulation mode particles (UBM, purple). Furthermore are shown observations of PNC/UFP as number of particles in the size range between 30 and 250 nm (N_30_250, green) for the time period 2016 - 2018. Note, no available measurements of $PNC_{>10}$ at this station, due to the issues with the instrumentation described earlier.

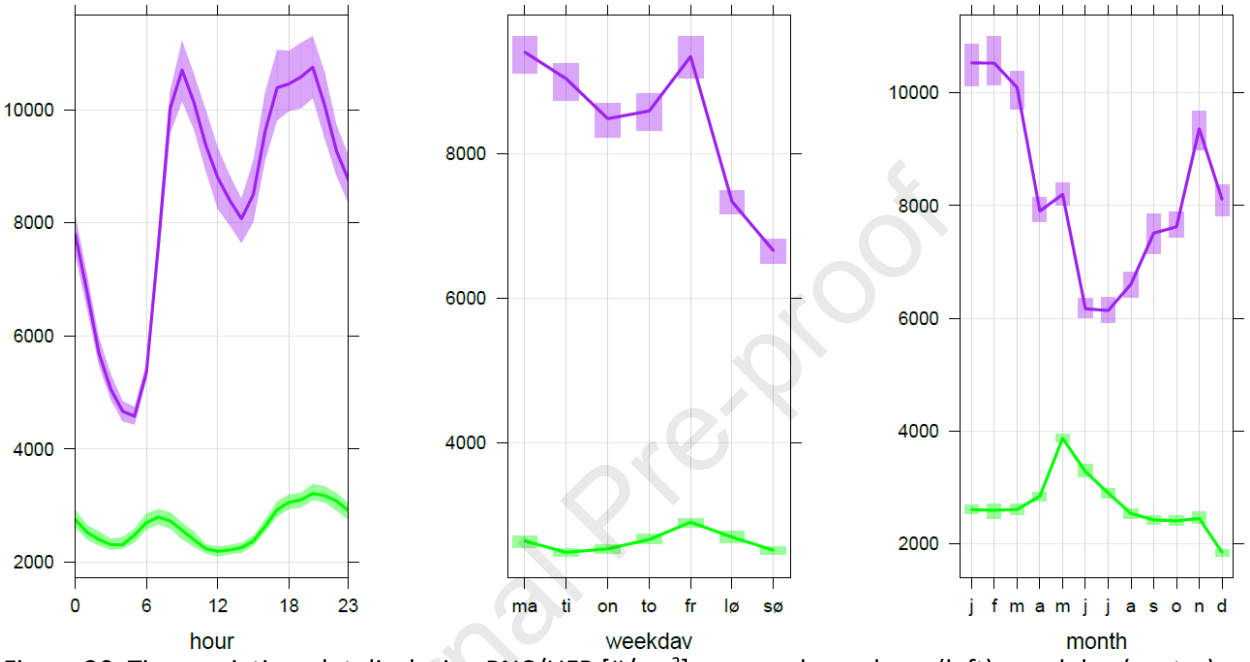

Figure 20. Time variation plot displaying PNC/UFP [#/cm^3] averaged over hour (left), weekday (center) and month (right) over the time period 1979-2018 for the suburban background station Hvidovre located in Denmark. Model results for Aitken and Accumulation mode particle number concentrations using UBM (purple) for the time period 1979-2018. Green line shows measured PNC/UFP in the size range between 30 and 250 nm for the time period 2016-2018.

Table 3 presents the Pearson correlation coefficients calculated for hourly, daily, monthly and annual mean concentrations for the comparison between measured particle number concentrations in the size range 30 – 250 nm and modelled results for the sum of Aitken and Accumulation mode particles at the local scale. Correlation coefficients for HCØ for hourly, daily and monthly mean values are in the range 0.39-0.47 (best for daily mean values), whereas the correlation coefficient for annual mean values is 0.87, indicating that e.g. the hourly and monthly temporal variation profiles for emissions are not adequately describing the observed variations. In contrast, the year-to-year variation in emissions, which follows the variation in $PM_{2.5}$ mass emissions, represents quite well the variation. The correlation coefficients for Lille Valby/Risø are similarly in the range from 0.40-0.86, with the highest value for annual averages, whereas the correlation coefficients for Hvidovre are 0.39 for hourly, 0.49 for daily, but -0.09 for monthly values and not available for annual values due to a limited number of measurements.

Table 3. Values of the statistical parameters Normalised Mean Bias (*NMB*) and Pearson correlation coefficient (*r*) for the comparison of the modelled sum of Aitken and Accumulation mode particles with measurements in the size range 30 – 250 nm for the three local background stations included in the

evaluation of the UBM model. In parentheses is the number of observations included in the calculation of the statistical value.

Station	***r* (hourly)**	***r* (daily)**	***r* (monthly)**	***r* annual**	***NMB (%)***
Lille Valby/Risø (rural)	0.40 (*81272*)	0.52 (*3545*)	0.41 (*153*)	0.86 (*14*)	285
Hvidovre (suburban)	0.39 (*21447*)	0.49 (*925*)	-0.09 (*38*)	NA*	218
HCØ (urban)	0.39 (*87138*)	0.47 (*3787*)	0.43 (*171*)	0.87 (*18*)	260

* number of annual mean values too few for statistical calculation (<5)

Discussion

In order to implement the M7 particle module in the regional chemistry-transport model DEHM and the urban background model UBM, emission data for particle number concentrations are necessary as input data. For this study, we initially intended to apply the same methodology for the emission calculations as was done in Kukkonen et al (2016), based on the emission dataset prepared by TNO (Denier van der Gon et al., 2014). Unfortunately, the resulting particle number emission data for 2005 from this approach did not compare well with the corresponding data for Denmark from the TNO particle number emission database. There are several possible explanations for this. Firstly, the TNO emission database only contains the mass and number of particles with diameters < 250 nm, as opposed to the SPREAD emission database, where all particles with a diameter below 2.5 µm are included. This fact alone gives the potential for large discrepancies, when the same factors for mass emission to number emission conversion are applied in the two cases. Furthermore technical developments since 2005 may have contributed to changes in the particle number emissions from traffic, which is one of the most significant sources.

Instead, we applied the derived statistical correspondence between mass and particle number emissions to scale the Danish national mass emissions and in this way we calculated emissions of particle numbers, in the effort to obtain compliance with both the national emission inventory and the TNO methodology. The problem with this method is that the TNO particle number emission inventory was based on particle number emission estimates per source type/sector, and larger particles were not taken into account as they contribute little to particle number concentration. Consistency with the Danish mass-based inventory from the SPREAD model for $PM_{2.5}$ cannot be expected, since the larger particles that contribute most to the mass, are neglected in the particle number emission inventory. When we in this study scale the $PM_{2.5}$ emissions to obtain particle number emissions, we, therefore, automatically overestimate the particle number. Information on size distribution is not available in the SPREAD emission inventory, which is the reason for this choice of method.

The original emission data set, from which we have extracted statistics, is derived for 2005. When applying the models, we use the statistical information to convert mass emissions, for which inventories are available from 1979-2018 for the present study. The year-to-year variation of the particle number emissions, will in this approach follow the year-to-year variation in the mass emissions of $PM_{2.5}$. For the results from the UBM model, where the particulate components in the model are treated as tracers, a quite good performance is achieved when examining correlations of annual mean values.

In the paper by Kukkonen et al. (2016) many discrepancies with observations were found, and attributed to the neglection of ammonium nitrate and SOA, which contribute to increasing the particle size during

ageing (but not so much to the increase of the particle number). In the present study, biogenic SOA could be important for the summer discrepancies in the comparison between models and measurements. Also Fountoukis et al. (2012) reported a discrepancy in particle size representation, using a different approach.

Another reason for discrepancies is the nucleation of particles that is only based on H_2SO_4 in the current work presented here. In reality, also NH_3, organic compounds of biogenic and anthropogenic origin and ambient NOx concentrations play a role (Dunne et al, 2016). Nucleation events strongly contribute to the large variability of concentration levels of PNC/UFP, and are dependent on the local environment in terms of atmospheric composition and meteorology.

Measurements of PNC/UFP with high spatial resolution in selected areas (larger cities and the surrounding region) are still relatively scarce, and the lower level cut-off diameter for the instruments are a source of uncertainty and can be variable, thereby giving rise to an underestimation of the particle number concentrations measured, especially for particles with a diameter smaller than 10 nm. The DEHM and UBM models overestimate the PNC/UFP concentrations of the sum of particles in Aitken and Accumulation mode, in general with a factor of two to three, when comparing with measured values of $PNC_{30\text{-}250}$. The main reasons for the overestimations are likely to be the lower cut-off of the measurements excluding some Aitken mode particles (included in the model), the application of a particle mass emission inventory together with statistics derived from an older emission inventory and the resulting risk of inadequate distribution of particle numbers between the different aerosol dynamical modes. The scarcity in measurements is an issue to address in the evaluation of the models, as e.g. the variability of the PNC/UFP concentrations across Denmark is difficult to evaluate with only three stations some 30 km apart.

With focus on the aim of the present study – to deliver address-level concentration data for the Danish population – it is relevant to evaluate the performance of the DEHM model in providing the input of PNC/UFP data to the UBM model, and the performance of the UBM model in providing that input to the OSPM model, which is described and applied in the accompanying Part 2 paper (Ketzel et al., 2021). As annual mean measured values are available at only one regional background station in Denmark, it is necessary to rely on the assumption that the distribution of particle number concentration in space resembles to a large extent that of the particle mass concentration. Similarly for the UBM model, with only three local background stations providing measurements for this study for evaluation, of which all are located within less than 30 km of each other, it is needed to assure that the distribution of the particle number concentrations across Denmark will resemble that of particle mass concentrations.

A lack of the PNC/UFP formation from emissions of VOCs and the related SOA formation in the particle dynamics module applied in the chemistry-transport model, could give some explanation of the discrepancy between modelled and measured monthly values – i.e. the annual variation. The anticipated production of PNC/UFP from VOCs will be highest during spring and summer time due to photochemical processes, and nearly zero during winter time. This is a feature that needs further development in the future work with the emission inventory, and the PNC/UFP modelling approach.

Conclusions

The M7 particle dynamics module was implemented in the regional scale air pollution model DEHM and the resulting modelled PNC/UFP concentrations for the sum of Aitken and Accumulation mode particles

were compared to measurements of particle number concentrations in the size range 30 – 250 nm obtained at four European stations. Based on calculated background concentrations of PNC/UFP, the UBM model has similarly produced local scale model results for the sum of Aitken and Accumulation mode particles, which have been compared to measurements at three stations in Denmark. The deterministic modelling of particle number concentration has been vitiated by the lack of consistency between emission inventories, and the evaluation of the models is challenged by the lack of consistent long-term measurement data.

The performance evaluation of the DEHM and UBM models shows that both models overestimate the level of the particle number concentrations at all stations with a factor of 1.5-4 (DEHM) and 2-3 (UBM), however, the results for the correlation coefficients for Danish measurement stations are 0.86 for DEHM and in the range 0.86-0.87 for UBM, for annual mean particle number concentrations. These are reasonable results, taking into account the difficulties encountered with respect to the setup of the emission inventories, and the challenges experienced with respect to measurement methodology. The distribution of particle number concentrations across Denmark of the Aitken and Accumulation mode particles features expected patterns with respect to emission distribution, and transport and transformation processes.

We conclude that the DEHM and UBM model results describing annual values of ambient PNC/UFP concentrations show some capability of reproducing observed patterns, when comparing the results of the models with available measurements, but that there is also room for improvement, especially with respect to the derivation and implementation of emission data and the treatment of volatile organic compounds based on natural emissions during summer time. Next steps are related to the most important current sources of uncertainty. Besides modelling the physical and chemical processes influencing the particle number size distribution, the overall challenge of modelling PNC/UFP lies within the emission inventory, where more understanding and thus improved estimates need to be acquired. The sources to very fine scale particles are in this study assumed to be correlated with emissions of primary $PM_{2.5}$ (dust, BC and OC) and sulphate emissions. The annual variation in observations indicate that PNC/UFP is also formed from emissions of VOCs and the related SOA formation, and here the particle chemistry needs to be accounted for in the respective models. Measurements of the chemical speciation of submicrometer particles would also be important to understand the sources to this fine aerosol fraction.

Author contributions

Matthias Ketzel, Jesper Christensen, Jørgen Brandt and Lise M. Frohn: Study conceptualization and design.

Ole Raaschou-Nielsen: Principal Investigator of the HERMES project, which the PNC modelling is part of.

Jesper Christensen, Lise M. Frohn, Jørgen Brandt and Matthias Ketzel: Model development and model calculations.

Matthias Ketzel, Lise M. Frohn and Christopher Andersen: Statistical analysis and evaluation of model results with measurements, graphical presentations and manuscript writing.

Astrid Manders and Hugo Denier van der Gon: advising and support on the implementation of the M7 model and the PNC emission database.

Andreas Massling: providing, processing and analysing PNSD measurements in Denmark.

Ulas Im, Ole-Kenneth Nielsen, Marlene S. Plejdrup: derivation and evaluation of spatially distributed emission data.

All authors contributed to the interpretation of the results. All authors have read and revised the manuscript for the important intellectual content, and approved the final draft of the manuscript.

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Credit Author Statement regarding the manuscript:

"Modelling ultrafine particle number concentrations at address resolution in Denmark from 1979-2018 – Part 1: Regional and urban scale modelling and evaluation"

by Lise Marie Frohn, Matthias Ketzel, Jesper Heile Christensen, Jørgen Brandt, Ulas Im, Andreas Massling, Christopher Andersen, Marlene Schmidt Plejdrup, Ole-Kenneth Nielsen, Hugo Denier van der Gon, Astrid Manders, and Ole Raaschou-Nielsen

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Matthias Ketzel, Jesper Christensen, Jørgen Brandt and Lise M. Frohn: Study conceptualization and design.

Ole Raaschou-Nielsen: Principal Investigator of the HERMES project, which the PNC modelling is part of.

Jesper Christensen, Lise M. Frohn, Jørgen Brandt and Matthias Ketzel: Model development and model calculations.

Matthias Ketzel, Lise M. Frohn and Christopher Andersen: Statistical analysis and evaluation of model results with measurements, graphical presentations and manuscript writing.

Astrid Manders and Hugo Denier van der Gon: advising and support on the implementation of the M7 model and the PNC emission database.

Andreas Massling: providing, processing and analysing PNSD measurements in Denmark.

Ulas Im, Ole-Kenneth Nielsen, Marlene S. Plejdrup: derivation and evaluation of spatially distributed emission data.

All authors contributed to the interpretation of the results. All authors have read and revised the manuscript for the important intellectual content, and approved the final draft of the manuscript.

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: