Atmospheric Environment 264 (2021) 118633

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

# Modelling ultrafine particle number concentrations at address resolution in Denmark from 1979 to 2018 - Part 2: Local and street scale modelling and evaluation

Matthias Ketzel [a,c,*], Lise M. Frohn [a], Jesper H. Christensen [a], Jørgen Brandt [a,e], Andreas Massling [a], Christopher Andersen [a,f], Ulas Im [a], Steen Solvang Jensen [a], Jibran Khan [a,f], Ole-Kenneth Nielsen [a], Marlene S. Plejdrup [a], Astrid Manders [b], Hugo Denier van der Gon [b], Prashant Kumar [c], Ole Raaschou-Nielsen [d,a]

[a] Aarhus University, Department of Environmental Science, Roskilde, Denmark
[b] TNO, Netherlands Organization for Applied Research, Utrecht, the Netherlands
[c] Global Centre for Clean Air Research (GCARE), University of Surrey, Guildford, United Kingdom
[d] Danish Cancer Society Research Center, Copenhagen, Denmark
[e] iClimate – Interdisciplinary Centre for Climate Change, Aarhus University, Roskilde, Denmark
[f] Danish Big Data Centre for Environment and Health (BERTHA) at Aarhus University, Roskilde, Denmark

## HIGHLIGHTS

- Particle number concentrations (PNC) implemented in the Danish air quality modelling system DEHM/UBM/AirGIS.
- Outdoor concentrations are provided at the front door of all residential addresses in Denmark for 40 years (1979–2018)
- Spatial resolution of 1 km × 1 km taking all emission sectors in Denmark into account.
- Additionally at the street locations consider the local traffic contribution, all done in 1 hour time resolution.
- Comparing model with $PNC_{30\_250}$ measurementsgives Pearson correlation coefficients in the range 0.39–0.95.
- Model overestimated the observed concentrations, Normalized Mean Bias in the range 6%–190% compared to $PNC_{>10}$.

## ARTICLE INFO

*Keywords:*
Particle numer concentration
Local-scale air pollution modelling
Ultrafine particles

## ABSTRACT

Modelling of ambient particle number concentrations (PNC) has been implemented in the Danish air quality modelling system DEHM/UBM/AirGIS and evaluated with long-term measurements. We implemented particle dynamical processes in the regional scale model DEHM using the M7 aerosol dynamics module (presented in the accompanying article by Frohn et al., 2021), and we developed models for PNC at the local scale (UBM) and street scale (OSPM), in a first approximation without including the particle dynamics as presented in this article.

Outdoor concentration estimates are provided at the front door of all residential address locations in Denmark for the past 40 years (1979–2018) with a spatial resolution of 1 km × 1 km taking all emission sectors in Denmark into account and additionally at the street location, with significant traffic (>500 vehicles/day).

We evaluated our model with up to 18-year long measurement time series of particle number size distributions (PNSD) at Danish street, urban and rural background stations. Two particle size ranges were used for evaluation: $PNC_{>10}$ (count of particles with diameter larger than 10 nm) and $PNC_{30\_250}$ (diameter range 30–250 nm), in order to exclude nucleation events from the measurements and to obtain a consistent long-term measured time series.

When comparing our model estimates with $PNC_{30\_250}$ measurements, we obtain Pearson correlation coefficients ($R_p$) in the range 0.39–0.95 depending on station location (street, urban background, rural) and averaging time (hour, day, month, year). The highest correlations were found for yearly averages at a monitoring station located at a street with dense traffic ($R_p$ = 0.95) whereas shorter time averages and comparisons with

* Corresponding author. Aarhus University, Department of Environmental Science, Roskilde, Denmark.
*E-mail address:* mke@envs.au.dk (M. Ketzel).

https://doi.org/10.1016/j.atmosenv.2021.118633
Received 4 February 2021; Received in revised form 16 July 2021; Accepted 17 July 2021
Available online 21 July 2021

monitoring stations at urban and rural background locations provided lower correlations. The model performance for PNC in terms of correlation coefficients with respect to measurements is comparable to the performance for other pollutants such as $NO_X$, $PM_{2.5}$ and better than the performance for $PM_{10}$.

The model generally overestimated the observed concentrations, Normalized Mean Bias (NMB) was in the range 6%–190% compared to $PNC_{>10}$ and 90%–290% compared to $PNC_{30\_250}$. These relatively high NMBs are probably caused by uncertainties in the modelling process, especially the estimation of particle number emissions, which largely determine the ambient concentrations of PNC. Furthermore, uncertainties might as well originate from the complexity of modelling particle dynamical processes accurately and the great challenges in performing long-term PNC measurements.

The presented model can estimate PNC at all Danish addresses over the last 40 years with a 1-h time resolution. The data seem to provide a good indication of the relative differences in PNC at Danish addresses.

## 1. Introduction

Despite the large improvements over the last decades, air pollution is still considered a major environmental risk to human health causing annually more than 200 million years-of-life-lost due to exposure to $PM_{2.5}$ in the EU-27 (EC, 2021). For various air pollutants (AP) of gaseous or particulate matter form, an association has been found with a long list of health endpoints as e.g. various types of cancers, cardio-vascular diseases, asthma, psychological disorders and premature death.

The gaseous AP components (e.g. $NO_2$, CO, $O_3$), the particulate mass concentrations (in $\mu g/m^3$ units e.g. selecting particles with diameters less than 10 μm or 2.5 μm: $PM_{10}$, $PM_{2.5}$) and lately also chemical components of particle mass such as elemental carbon (EC) are frequently considered as causal pollutants. Their associations with health endpoints have been investigated in epidemiological studies using both long-term measurements, statistical models and dispersion models as exposure matrix. At present, only the concentration of particle mass in ambient air is regulated in air quality directives and widely measured in a monitoring context.

Particle number concentrations (PNC) are measured as number of particles per volume (in $\#/cm^3$ units) and is a more appropriate metric to measure smaller particles with very little mass, compared to measures such as $PM_{10}$ or $PM_{2.5}$. Ultrafine particles (UFP, particles with diameters $< 0.1$ μm) are a subset of PNC but often dominating the PNC since most of the ambient PNC are typically below 0.1 μm especially when observed close to source regions (Kumar et al., 2014). PNC and UFP attracted significant attention as another health-relevant component of AP since the late 1990s when the first studies showed an association of the smallest particles with health effects and the translocation of UFP into different organs of animals could be documented (Oberdörster et al., 2000; Peters and Wichmann, 2001; Stone et al., 2017; Oliveira et al., 2019) as well as in various recent studies, especially in studies of short-term effects of air pollution (Heal et al., 2012; Ohlwein et al., 2019).

Two scientific hypotheses exist concerning health effects and atmospheric particles: 1) that health effects might be stronger associated with particle mass (PM) dominated by particles in the diameter range of 0.1–2.5 μm and 2) that health effects might be stronger associated with particle number concentration (PNC), dominated in the ultrafine diameter range $< 0.1$ μm (UFP) (de Jesus et al., 2019). A literature review (HEI, 2013) concluded that "the evidence does not support a conclusion that exposures to UFP alone can account in substantial ways for the adverse effects that have been associated with other ambient pollutants, such as $PM_{2.5}$". From a realistic view of human exposure, UFP in most cases occur simultaneously with various other pollutants, which makes the direct assignment of such particles to health effects difficult. However, still very few epidemiological studies consider PNC/UFP as exposure matrix compared to the traditional gaseous AP and PM, due to a relatively much smaller number of long-term PNC/UFP measurements and difficulties in modelling PNC, in general, and at high resolution and over a long time period, specifically. The lack of PNC measurements is caused by the fact that these measurements are complex and that PNC is not regulated in environmental air quality standards and therefore not required in routine monitoring programs.

Despite this lack, some high quality measured PNC datasets exist and have been analysed (von Bismarck-Osten et al., 2013; de Jesus et al., 2019) and measurement databases including PNC are available (Birmili et al., 2009b, ACTRIS, EBAS.nilu.no, WMO-GAW).

Road traffic, other transport, various types of combustion but also secondary particle formation from gaseous precursors have been identified as dominating sources for PNC in urban and sub-urban areas (Ketzel et al., 2004, Kittelson et al. 2004; Kumar et al., 2011, Kumar et al. 2013; Keuken et al. 2015). The secondary particle formation (also called nucleation or new particle formation) is more important in remote and clean environments and is subject to a whole research field in itself with a strong relation to climate questions and is not in the primary scope of this paper (see as well note in chapter 2.1). The main area of interest for this work is PNC in relation to urban and sub-urban areas where the population density is highest and where road traffic and other stationary combustion are the predominant sources.

Several modelling approaches of PNC/UFP have been conducted, including process studies investigating the time scales of various aerosol transformation processes (Pohjola et al., 2003; Ketzel and Berkowicz, 2004; Zhang et al., 2004; Kumar et al., 2011), statistical modelling approaches (Hussein et al., 2006; Nikolova et al., 2014; von Bismarck-Osten et al., 2015) or deterministic modelling based on emission inventories and dispersion models of various scales and complexity (Gidhagen et al., 2004; Nikolova et al., 2014; Roldin et al., 2014; Kukkonen et al., 2016).

On an urban and local scale, PNC undergo transformation processes such as nucleation, condensational growth, evaporation, coagulation and deposition that may change concentrations fast. The importance of these processes need to be assessed in comparison to emission and dispersion and the processes might have to be considered in the modelling approach. On a regional to hemispheric scale, also cloud processing of particles has to be considered. The importance of these – often non-linear – transformation processes depend on the concentration of particles, their respective chemical composition, the presence of semi-volatile gasses and the dominating meteorological conditions. Using time scale analysis, the relative importance of these PNC specific processes has been compared to the emissions and dilution (Ketzel and Berkowicz, 2004; Kumar et al., 2011; Karl et al., 2016). In most urban environments and on smaller spatial scales outside the direct exhaust plume, emission and dilution can often be considered the most important processes (Gidhagen et al., 2005; Ketzel and Berkowicz, 2005; Wang et al., 2010b). Therefore, we do not take these transformation processes into account for the local scale in this first implementation (See section 2.4 for more details).

A few modelling approaches have been developed that are able to predict PNC including particle transformation processes in different domains and at different spatial resolutions: at street/local scale (Vignati et al., 1999; Gidhagen et al., 2004; Zhang et al., 2004; Nikolova et al., 2014), at urban scale (Ketzel and Berkowicz, 2005; Kukkonen et al., 2016) or even regional scale (Manders et al., 2017; Chen et al., 2020).

Kukkonen et al. (2016) presented a PNC modelling approach applied

for five European cities employing the LOTOS-EUROS model coupled with the M7 particle dynamics module for the regional/urban background scale while different models without particle dynamics were used for the street/local scale.

Both model results and observations of PNC and PM have shown that these pollutants show different spatial patterns and different temporal behaviour during atmospheric processing and the importance of specific emission sources can be very different between the two metrics. PNC is similar to $NO_X$ strongly affected by local (traffic) sources and shows strong concentration differences between kerbside and background locations, while PM is often dominated by long-range transport with smaller differences between kerbside and background locations (Ketzel et al., 2004). PNC and PM are only moderately correlated.

In Danish health studies, the use of air pollution models integrated into the Danish DEHM/UBM/AirGIS exposure modelling system (http://au.dk/AirGIS/; Jensen et al., 2001; Khan et al., 2019) has become a standard (Hvidtfeldt et al., 2019; Khan et al., 2019; Thacher et al., 2021). The presented work extends that modelling system to provide concentrations of PNC/UFP for the first time at Danish residential address level (N = 2.1 million) with high temporal (1 h) and spatial (front-door address location) resolution.

The whole model development, setup and evaluation are described in two accompanying papers. Part 1 is dealing with the setup of the PNC emission inventory and the particle dynamics in the regional/urban scale models, using DEHM and UBM as well as evaluation of these models with regional and urban scale measurements.

This paper is part 2 and describes the model setup at street scale level using the AirGIS system including the Operational Street Pollution Model (OSPM®) and the evaluation of this model with street scale measurements for almost two decades.

## 2. Modelling methods

### 2.1. Definitions, size ranges, naming conventions, scope of work

#### 2.1.1. Definitions – type of measurements – size ranges

As mentioned in Section 1, the most frequently used metric to measure particles in smaller size ranges is particle number concentration (PNC) that delivers number concentrations expressed in #/cm$^3$ units. Ultrafine particles (UFP) are often defined as particles below 0.1 μm in diameter assuming sphericity of the particles, and they are the dominating subset of PNC at urban atmospheric conditions as most particles in this environment are smaller than 0.1 μm in diameter.

The most common device for performing PNC/UFP measurements is a particle counter (e.g. a condensation particle counter – CPC). Such a device counts all particles above a certain diameter corresponding to the detection limit (e.g. 0.01 μm = 10 nm) and gives the total particle number concentration (e.g. $PNC_{total\ >10}$) as a result of the measurement (Mertes et al., 1995; Kumar et al., 2010). The more advanced type of measurement devices measures the particle number size distribution (PNSD), i.e. the concentration of particles in different size intervals over a total size range (e.g. 30 size bins in the range 6–700 nm). Measurements of PNSD allow for a post-processing, in order to calculate number concentrations in well-defined particle size ranges, e.g. number concentration of particles between 30 nm and 250 nm ($PNC_{30\text{-}250}$). Measurement devices that provide PNSD data are not trivial and require high-level resources with respect to maintenance, frequent calibrations and expert knowledge. Continuous measurements using such devices appear in the late 1990s and are still relatively scarce especially with higher spatial coverage for smaller regional areas (urban environment including the surrounding area) compared to CPCs or even PM monitors. PNC/UFP measurements are not mandatory in EU monitoring programmes, and no limit values or guidelines are defined so far. For this reason, they are not part of standard monitoring programmes, however, a few research-oriented PNC monitoring networks exist as, e.g. the German GUAN network (Birmili et al., 2016), the ACTRIS programme, the Danish Monitoring Programme (Ellermann et al., 2016) and a few European cities (Asmi et al., 2011; Hofman et al., 2016). The scarcity in measurements is also reflected in the lack of a standard for instrumentation, operation of instruments and processing of data with respect to PNC/UFP measurements, which typically guides the measurements of air pollution components in, e.g. the EMEP programme (Tørseth et al., 2012). Within the ACTRIS programme (Wiedensohler et al., 2012), such standards are being developed.

Since UFPs in this study are defined as particles below 100 nm in diameter, this corresponds to an upper size limit for $PNC_{<100}$. This upper size limit is crucial for the measurement of mass concentrations (corresponding to $PM_{0.1}$) to make a clear separation from particles larger than 100 nm in diameter containing a great portion of the total suspended PM under typical atmospheric conditions. For number concentration measurements the 100 nm upper size limit is less defining, since the number of particles above 100 nm is typically much lower than the number of particles below 100 nm in such locations when being close to the sources of particles. Analysis of time series of $PNC_{<100}$ with $PNC_{30\text{-}250}$ and $PNC_{total}$, reveals typically a very high correlation between these metrics, and the term PNC is therefore used in this study to cover all the various exact size range definitions. This is valid in environments where the contribution of nucleated particles is very small compared to all UFPs from different sources (own analysis of various Danish and European PNC data).

#### 2.1.2. Treatment of the smallest particles (nucleation mode) in the measurements and the model system

For PNC/UFP measurements, the lower particle diameter size limit or cut-off is often very crucial, as discussed in the text below. It is very difficult both to measure and model accurately the smallest particles, which are formed from gaseous precursors through nucleation (nucleation mode). In the first step of nucleation, such particles are also called clusters as this expression comes closer to the chemical state of the particles. Many measurement methods have a theoretical cut-off value or detection limit of around 3–10 nm. In addition, the nucleation mode particles have a very short lifetime and tend to deposit very fast (e.g. in the inlet device of the instrument or the instrument itself) and can self-coagulate at high concentrations, which makes their quantification close to emission sources difficult and uncertain. Moreover, instruments also tend to become less sensitive to the smallest particles over time when service and calibrations are not performed regularly with high accuracy. Data processing can account for the losses in instruments or inlets, but in reality, the exact accounting is debated, and different instrument operators apply these corrections differently.

With respect to the model system developed here, the implemented nucleation scheme in the M7 module describes the formation of sulphuric acid particles (with sizes primarily below 10 nm) based on the methodology of Vignati et al. (2004). Other types of nucleation processes occurring in the atmosphere are, however, not included in M7. Within the work presented here, we have not implemented any additional nucleation processes in M7 or evaluated the currently implemented nucleation scheme. Only a few groups within this scientific community have attempted to model nucleation events, and this has only been done for selected so-called super site measurement stations (Kulmala and Kerminen, 2008; Baranizadeh et al., 2017; Roldin et al., 2019). The attempt to model the number of particles accurately in the very small size fractions (below 10 nm in diameter) is therefore beyond the scope of this study. A further argument for this decision is based on the fact that we have to deal with a lower cut-off for PNC (around 10–40 nm) due to instrumental issues and data availability when comparing our model results with measurements. This is e.g. the case for the Danish measurements in the latest years, which had difficulties to measure particles below sizes of about 30–40 nm.

For the use in epidemiological studies, we need a robust and relatively simple approach for our developed modelling system to be able to estimate PNC/UFP for 40 years back in time and for the whole of

Denmark. Therefore, the preferred PNC/UFP definition here is the size range 30–250 nm, thereby limiting the influence of new particle formation events on the measurements and limiting the influence from instrumental challenges at low particle sizes. Similar size range limits were applied in previous studies (Asmi et al., 2011; Kukkonen et al., 2016). For the particle concentration data obtained within size ranges from the observed PNSD, the $PNC_{30\text{-}250}$ metric is derived as close as possible to this definition.

### 2.2. The DEHM/UBM/AirGIS modelling system

As basis for our PNC/UFP model development, we use the multi-scale and high-resolution Danish air pollution modelling system, called DEHM/UBM/AirGIS (http://au.dk/AirGIS/; Brandt et al., 2001; Ketzel et al., 2011; Khan et al., 2019). The system has been developed by the atmospheric modelling group at Aarhus University over the last three decades with focus on air pollutants important for human health and environment. DEHM/UBM/AirGIS output has served as estimate for human exposure to ambient air pollution in a large number of epidemiological studies (Jensen et al., 2009; Raaschou-Nielsen et al., 2011; Sørensen et al., 2012; Poulsen et al., 2016; Roswall et al., 2017; Hjortebjerg et al., 2018). The existing system covers a "chain of models" where the output of one model serves as input for the next. The system consists of a meteorological model and three air pollution models that operate on regional, urban/local and street scale, respectively. The meteorological model and two of the air pollution models including the implementation of PNC as new pollutant, are described in details in the accompanying paper (Frohn et al., 2021), and only a brief overview is given here. The main focus in this section is the description of the street scale model.

The model chain is driven by the meteorological Weather Research and Forecasting (WRF) Model. The model chain further consists of the Danish Eulerian Hemispheric Model (DEHM; Frohn et al., 2002; Brandt et al., 2012), a chemistry-transport model, which calculates emissions, atmospheric transport, chemical transformation and deposition of 110 chemical gaseous and particle phase species. The model includes four two-way nested geographical domains with different spatial resolutions in order to calculate intercontinental and regional transport and transformation of air pollution, while obtaining a relatively high resolution over Denmark.

The Urban Background Model (UBM; Brandt et al., 2001; Brandt et al., 2003) is a local scale model, and is applied to obtain air pollution concentrations over the whole of Denmark with a 1 km × 1 km resolution. The UBM model obtains boundary conditions from the DEHM model (at 5.6 km × 5.6 km resolution), at a location 25 km upstream from the receptor location in order to avoid double counting of emissions in both models. The UBM is a plume-in-grid model that calculates concentrations of selected chemical species, including the gasses $NO_x$, $NO_2$, CO and $O_3$ as well as the primarily emitted particles in size fractions of $PM_{2.5}$ and $PM_{10}$.

Finally, the AirGIS model system is applied, to calculate air pollution levels at the address level, in streets with more than 500 vehicles per day. The AirGIS system is coupled to the DEHM/UBM models, which provide background air pollution concentration levels and to the WRF model, which provides meteorological input in order to run the Operational Street Pollution Model (OSPM®) (Berkowicz, 2000). The AirGIS is built in a GIS-based system that contains all necessary information to run the OSPM model, e.g. traffic density (divided into different vehicle categories, i.e. cars, vans, trucks and buses and their corresponding emission factors), street and building configurations, and background concentrations for all streets in Denmark (see Khan et al., 2019 for details).

The OSPM is a widely used and validated street pollution model (Kakosimos et al., 2010; Ketzel et al., 2012) that calculates air pollution concentrations inside the street with hourly time resolution over a time period of 40+ years for all Danish addresses. OSPM is a so-called parameterised model that includes the main flow and dispersion processes inside a street canyon, in the form of a combination of a plume model for the direct contribution and a box model for the recirculating part of pollution inside the street canyon (Berkowicz, 2000). OSPM also considers the turbulence produced by the moving vehicles that contributes substantially to the dispersion process, especially at low wind speeds.

The AirGIS model system automatically calculates the input to the OSPM model, which then estimates the air pollution level at the front door at all relevant addresses in Denmark. The AirGIS model has been evaluated against both long-term air pollution measurements and short-term campaign data in Denmark (Ketzel et al., 2011; Jensen et al., 2017; Hvidtfeldt et al., 2018; Khan et al., 2019).

Calculations at residential addresses in streets with more than 500 vehicles per day were based on the full model chain DEHM/UBM/AirGIS that includes the OSPM as described above. For residential addresses at streets with less than 500 vehicles per day, the regional and urban background concentrations were calculated with the DEHM/UBM model system at a 1 km × 1 km resolution.

### 2.3. Emission inventories and vehicle emission factors

A crucial first step for implementing a new air pollution component (PNC) in the modelling system is the generation of an emission inventory with an appropriate spatial and temporal resolution.

A detailed description of the emission inventory used in the regional (DEHM model) and in the urban background (UBM model) part of our modelling system is given in the accompanying publication (Frohn et al., 2021), and these parts of the emission modelling setup are only described briefly in the following.

The emissions for DEHM for the different size modes of PNC, the Aitken mode (particle size range 10–100 nm) and the Accumulation mode (100–1000 nm), are based on different existing global and European inventories. Based on a European particle number emission inventory used for the LOTOS-EUROS model (Denier van der Gon et al., 2014; Manders et al., 2017) scaling methods were derived (Frohn et al., 2021), that were applied to the high-resolution Danish emission database (SPREAD model, Plejdrup et al., 2018) and the other global or European inventories implemented in DEHM. These emissions were extrapolated for the entire modelling period 1979 to 2018.

For the Urban Background Model (UBM), we implemented the sum of Aitken and Accumulation mode particles as emissions for the new PNC/UFP tracer based on the Danish emission database from the SPREAD model (1 km × 1 km resolution) and based on the above-mentioned scaling methods derived for the DEHM model and further described in Frohn et al. (2021).

For the street scale modelling, PNC/UFP emission factors have to be implemented in the OSPM. Emission factors for particle number emissions are very uncertain compared to the well investigated and documented emissions for traditional pollutants (NOx, CO, PM, etc.). Particle number emission factors (PNEF) depend – similar to traditional pollutants – on several parameters such as, e.g. vehicle speed, type and age of vehicles, and specific composition of the vehicle fleet. However, additional parameters relevant for PNEF are the ambient temperature, humidity and sulphur content of the fuel combined with an additional dimension, the particle size. Measurements of PNEF depend as well on the used instrumentation, their lower and upper cut-off size range and the dilution conditions applied for the raw exhaust under laboratory or ambient conditions. Many data for PNEF are reported for single years and specific stations (Kumar et al., 2011; Vouitsis et al., 2017), however, due to the before mentioned dependencies, the data situation is very complex, largely uncertain and in general valid and realistic emission factors are difficult to derive.

For the here described work, a relatively robust and consistent method was required that allows to estimate PNEF for a large variety of vehicle types. For this purpose, we implemented the PNEF reported in

the EMEP/EEA air pollutant emission inventory guidebook (EEA, 2019) and compared these PNEF with previous estimates for Danish conditions. The guidebook gives PNEF for urban, rural and highway driving conditions. Since the receptor locations that are relevant and used in this project are mostly located along streets with urban driving patterns, the **urban** emission factors are used in our model setup. The guidebook provides as PNC metric the total particle number (without mentioning a lower cut-off diameter) and the solid particle number (remaining after heat cycle to remove volatile compounds) divided into three different size bands (<50 nm, 50–100 nm, 100–1000 nm), of which the total particle number is used here.

Since the guidebook tables are not providing PNEF for all EURO classes, some expert judgment and extrapolation from nearest EURO classes had to be used. For other vehicle types, the emission factors in the guidebook had more sub-categories than resolved in OSPM, here a weighted interpolation was used to harmonize available existing data with the constraints of the OSPM model. Table 1 shows the complete set of PNEF applied in the OSPM model setup. Light-duty vehicles (LDV or vans) have been assigned the same emission factors as passenger cars.

The implemented basic set of PNEF (Table 1) is combined with the actual vehicle fleet composition with respect to EURO classes obtained from the Danish vehicle registration database for each of the years from 1979 to 2018. Therefore, the resulting PNEF will change from year to year as they are applied as input data in the OSPM model for further pollution concentration calculations.

Table 2 presents a comparison of various PNEF from the literature with a focus on previous Danish studies and the applied values from this study. Emission factors in Table 2 are reported separately for light-duty vehicles (LDV) and heavy-duty vehicles (HDV) or for the mixed vehicle fleet, and some important characteristics of the listed references are provided, e.g. year of the study, road type, vehicle speed and particle size range. There is a large diversity of reported PNEF and parameters, which is already obvious regarding this small selection. The PNEFs used for this study are given for the years 2001, 2008, 2010 and 2017 in order to show the development with time and provide data for the same years as in literature studies for comparison.

A large reduction of PNEF is observed between 2001 and 2017, which is in accordance to the large reduction in observed roadside PNC (Fig. 1, see Section 2.5) and caused by a continuous renewal of the vehicle fleet and introduction of exhaust gas after-treatment, while newer, less emitting vehicles are replacing older vehicles with higher emissions.

A direct comparison with previously estimated emission factors for 2001 in Denmark for a mixed fleet under urban conditions reveals substantially lower PNEF values in this study of 111 • $10^{12}$ #/(vehicle * km), compared to 280 • $10^{12}$ #/(vehicle * km) in previous work (Ketzel et al., 2003).

Also, a comparison for 2008 conditions points in the same direction with this study calculating PNEF of 18 and 724 • $10^{12}$ #/(vehicle * km) for LDV and HDV, respectively, that are much lower than previously estimated 80..100 • $10^{12}$ #/(vehicle * km) for LDV and 1750 … 2210 • $10^{12}$ #/(vehicle * km) for HDV (Wang et al., 2010a).

PNEFs used for the city of Helsinki for the year 2008 by (Kukkonen et al., 2016) are in the range of a factor 2 higher than estimated Danish values (Wang et al., 2010a) and a factor of 4–10 higher than the PNEFs in this study, showing the wide diversity in this parameter.

The comparison reveals that the PNEF used in this study are about a factor of 2–3 lower compared to previous Danish studies. This is in accordance with the fact that we here focus on a smaller particle size range of about 30 nm–250 nm while PNEFs obtained in previous studies typically cover all particles above 7 nm or 10 nm.

A next step in getting more accurate PNEF would be to change to size-resolved emission factors as, e.g. reported in (Wang et al., 2010a) and switching both in the UBM and OSPM models to a size-resolved representation of the PNC. This would require a substantially higher demand on input data, e.g. size-resolved emission factors for all the vehicle categories presented in Table 1. Such data are, however, very sparse, and so far, only a few data exist for specific years and locations. Working with size-resolved emission factors is therefore not feasible at the moment, especially for the present study that is covering a time span of 1979–2018, where a simple and robust method to estimate PNEF as described above, is more appropriate.

### 2.4. Dispersion and particle transformation modelling

PNC/UFP undergo transformation processes, which are different from those of gaseous species and other particulate metrics (e.g. $NO_x$, CO, $O_3$, $PM_{10}$, $PM_{2.5}$) that are already implemented in DEHM/UBM/AirGIS. PNC are altered by the transformation processes nucleation, condensation, evaporation, coagulation and cloud processing, and their assessment requires therefore a specific particle dynamics module. Based on a literature review of ultrafine particle modelling studies conducted in Europe, the M7 particle dynamics module (Vignati et al., 2004) was chosen for implementation into DEHM. The M7 module is developed mainly for air pollution modelling purposes and has been used by several groups in Europe and was recently applied in the EU TRANSPHORM project to model particle number concentrations in five European cities (Kukkonen et al., 2016). More details on the M7 implementation in DEHM and corresponding validation are given in the accompanying paper (Frohn et al., 2021).

In the UBM and OSPM models, the PNCs are treated as inert tracers, only emission and dispersion processes are included to date, and no particle transformation processes are applied. This approximation is based on the results of time scale analysis (Ketzel and Berkowicz, 2004; Zhang et al., 2004; Kumar et al., 2011) and modelling studies (Gidhagen et al., 2005; Karl et al., 2016; Wang et al., 2010b). These studies have concluded that at the street- and urban-scale, outside the direct vehicle plume and outside confined environments such as road tunnels, the emission and dispersion processes are by far the most important processes to be included in the modelling calculations. Processes such as deposition and coagulation might under certain conditions change PNCs in the range of 10–40%, however, taking as well the large uncertainties in the emission factors into account, we opted for a simple, less calculation time demanding and robust approach in this study and considered PNC as an inert tracer.

### 2.5. Measurements of PNC/UFP

For definitions of variables, type of instruments and size ranges, please consult section 2.1 at the beginning of this paper.

**Table 1**
Particle number emission factors (PNEF) in units $10^{12}$ #/(vehicle * km) as implemented in OSPM in this study applicable for urban driving situations.

| EURO class | PasCar Gasoline | PasCar Diesel | Trucks <7.5t | Trucks 7.5–12t | Trucks 12-32t | Trucks >32t | Urban bus | Coaches |
|---|---|---|---|---|---|---|---|---|
| **Conventional** | 0.9 | 404 | 319 | 678 | 1060 | 1500 | 688 | 823 |
| **EURO 1** | 9 | 404 | 319 | 678 | 1060 | 1500 | 688 | 823 |
| **EURO 2** | 9 | 212 | 319 | 678 | 1060 | 1500 | 688 | 823 |
| **EURO 3** | 1 | 80 | 319 | 678 | 1060 | 1500 | 688 | 823 |
| **EURO 4** | 0.07 | 9 | 2.7 | 5.8 | 9.1 | 15 | 6 | 7 |
| **EURO 5** | 0.07 | 9 | 8 | 16.9 | 26.4 | 45 | 17.3 | 21.5 |
| **EURO 6** | 0.07 | 0.07 | 8 | 16.9 | 26.4 | 45 | 17.3 | 21.5 |

**Table 2**
Comparison of PNEFs in units $10^{12}$ #/(vehicle * km) reported from field studies. LDV = Light-duty vehicles (cars and vans), HDV = Heavy-duty vehicles (trucks and buses).

| Reference | Road type | Year | Size range (nm) | Speed LDV/HDV (km $h^{-1}$) | PNEF LDV | PNEF HDV | PNEF MIX | Share of HDV in % | Ratio PNEF HDV/ LDV |
|---|---|---|---|---|---|---|---|---|---|
| Imhof et al. (2005) | Motorway | 2001 | >7 | 120/85 | 690 | 7300 | | | 11 |
| -"- | Highway | 2001 | >7 | 85/75 | 320 | 6900 | | | 22 |
| -"- | Urban road | 2001 | >7 | 0-50/0-50 | 80 | 5500 | | | 69 |
| Jones and Harrison (2006) | Urban road | 2002–2003 | 11–437 | 0-50/0-40 | 58 | 636 | | | 11 |
| Klose et al. (2009) | Urban road | 2005–2006 | 4–800 | 0-30/0-30 | 540 | 43000 | | | 80 |
| Birmili et al. (2009a) | Urban road | 2005 | 10–500 | 75–90 | 24 | 2960 | | | 123 |
| Ketzel et al. (2003) | Urban road | 2001 | 10–700 | 50 | | | 280 | 3.4 | |
| Wang et al. (2010a) | Motorway | 2008 | 10–700 | 110/90 | 81 | 1750 | 215 | 10.5 | 22 |
| -"- | Urban road | 2008 | 10–700 | 0-50/0-50 | 100 | 2210 | 187 | 4.5 | 22 |
| Kukkonen et al. (2016) | Urban road | 2008 | >10 | | 180 | 2700 | | | |
| This study | Urban road | 2001 | >7..20 | 50 | 50 | 931 | 111 | 3.4 | 36 |
| -"- | Urban road | 2008 | >7..20 | 50 | 18 | 724 | 63 | 3.4 | 40 |
| -"- | Urban road | 2010 | >7..20 | 50 | 15 | 277 | 38 | 3.4 | 18 |
| -"- | Urban road | 2017 | >7..20 | 50 | 6.4 | 70 | 12 | 3.4 | 11 |

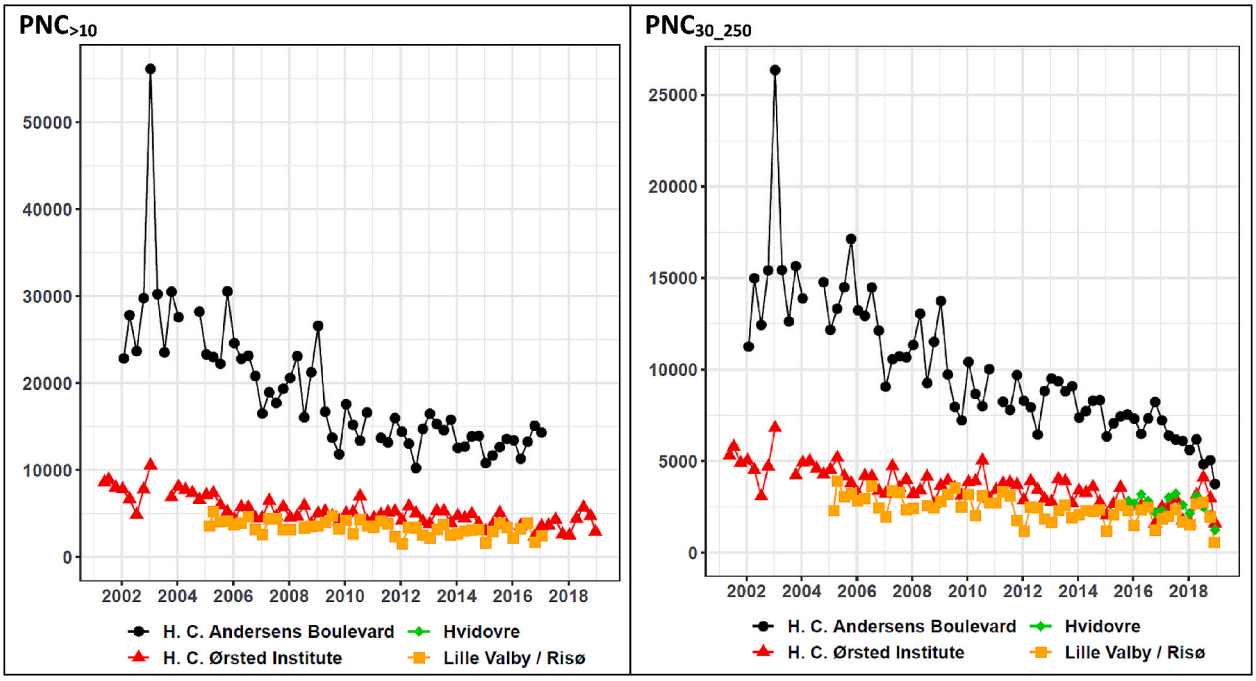


**Fig. 1.** Seasonal (3–monthly) averages at four Danish monitoring stations: the street station HCAB, the urban background station HCØ, the suburban station Hvidovre and the rural background station Lille Valby/Risø. Two types of instruments with different size ranges were used to measure PNSD at the four stations, DMPS and SMPS, see text for more details. $PNC_{>10}$ is only available for DMPS instruments. **Left Panel:** $PNC_{>10}$ (above 10 nm size, DMPS only). **Right Panel:** $PNC_{30\_250}$ (in the range from 30 nm to 250 nm).

In Denmark, the first routine measurements of particle number size distributions (PNSD) started in 2001, and today Denmark has some of the longest continuous PNSD data sets worldwide that consider a high spatial resolution of measurements in a limited area (larger city and surrounding region) and give a great base for model development and evaluation (Ellermann et al., 2016).

We employ this unique dataset of 18 years of continuous PNC/UFP measurements performed at three locations: a street station in the centre of Copenhagen (station: H.C. Andersens Boulevard = **HCAB**), an urban background station in Copenhagen, at the rooftop in 20 m height (station: H.C. Ørsted Institute = **HCØ**), and a rural background station about 30 km west of Copenhagen (station: **Lille Valby/Risø**) (Ellermann, 2016). The rural background station's measurement location was moved by a few km in 2010 from Lille Valby to Risø. The measured time series at both locations have been merged and considered as one long dataset representing a rural background, since there is only a minor difference

between the air pollution levels at these two places.

In addition, much shorter time series are available at another street (**Jagtvej**) in Copenhagen and at a suburban station (**Hvidovre**), which is located approximately six km southwest of the city centre of Copenhagen (Ellermann et al., 2016). We apply the observations at these stations to test and evaluate the developed PNC/UFP modelling approaches at all different model scales.

Fig. 1 shows the available time series at the four stations used in this work. The time series at Jagtvej was too short to calculate long term averages and was omitted in Fig. 1. Two types of instruments have been used to measure PNSD at the four stations: a DMPS (Differential Mobility Particle Sizer) with particle size range (10–700) nm and a SMPS (Scanning Mobility Particle Sizer) with particle size range (30–478) nm. At the suburban station of Hvidovre only the SMPS instrument has been used for all years. At two other stations (HCAB and Lille Valby/Risø), the DMPS was used until the end of 2016, and the SMPS measurements started in 2017, and at the urban background station (HCØ) the DMPS was used until the end of 2018, and the SMPS measurements started in 2019 (data not shown here). For the new SMPS instruments, 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 concentration will be somewhat underestimated.

Fig. 1 shows measured data both as $PNC_{>10}$ (only available for DMPS instruments) and as $PNC_{30\_250}$ (available for all instruments) in the respective size ranges $>10$ nm and (30–250) nm. Levels are about 50% lower for the limited size range $PNC_{30\_250}$. However, the relative shape of the curves looks very similar, as the ranking and the ratio between different stations are very similar. Also, the correlation coefficients between $PNC_{>10}$ and $PNC_{30\_250}$ are very high, 0.96 to 0.99 for the 3 stations, indicating the close relationship between the two PNC/UFP metrics, both used for evaluation with the modelled PNC.

### 2.6. Evaluation method, what and how to compare

This section summarizes the previous statements regarding PNC/UFP modelling and measurements with respect to metric/size ranges and justifies how the evaluation is performed in the following chapter.

#### 2.6.1. Model

Modelling the nucleation mode at rural level was beyond the scope of this work. Therefore, the four "middle" size modes from the M7 model, i.e. the Aitken and Accumulation modes in both insoluble and soluble form, are used as combined $PNC_{model}$ result from the DEHM output. At the rural level it is most appropriate to evaluate with $PNC_{30\_250}$, hereby excluding the nucleation mode particles from the measurements.

However, at urban and street level, particles smaller than 30 nm are as well locally produced and primarily emitted, e.g. from traffic. Due to the lack of size-resolved PNC emission factors, these locally produced particles are reflected in the local emissions both in UBM and OSPM models. In order to include those small traffic-related particles in the evaluation, it is more appropriate to compare with measurements of $PNC_{>10}$.

The largest uncertainties in the modelling part lies in the emission estimates at all model scales and in the omission of particle dynamics in the UBM and OSPM models.

#### 2.6.2. Measurements

Measuring PNC for particle sizes below 30 nm poses several instrumental challenges. For the latest years, the new SMPS instruments at the Danish stations were applied and only measurements of particles with diameters above 30 nm can be used (HCAB $\geq$ 2017, Lille Valby/Risø$\geq$2017, Hvidovre $\geq$ 2015). Using $PNC_{30\_250}$ for model evaluation will allow for longer time series by combining the old DMPS and the new SMPS data. Using $PNC_{>10}$ limits the available data availability, however, this metric is closer to the common definition of PNC/UFP.

Fortunately, the two metrics $PNC_{30\_250}$ and $PNC_{>10}$ are very closely correlated $R_P > 0.96$ for the here considered urban/near-city locations. This indicates that for practical application, the two metrics can be considered as a proxy for each other.

In absolute terms, $PNC_{30\_250}$ is about 50% of $PNC_{>10}$ indicating that about half of $PNC_{>10}$ can be found in the 10 nm–30 nm size range.

#### 2.6.3. Evaluation

As explained above, there are plenty of challenges connected with measuring and modelling PNC, especially for sizes below 30 nm and there are uncertainties and approximations involved in all the various steps. Since there are aspects both from the modelling and the measurement setup that are in favour of using either $PNC_{30\_250}$ or $PNC_{>10}$ for model evaluation, we consequently use both metrics in the evaluation analysis.

Evaluations are presented in graphical form as time series plots and scatter plots for both $PNC_{>10}$ and $PNC_{30\_250}$. As main validation metrics, the Pearson correlation coefficient ($R_P$) and the Normalized Mean Bias (NMB) are given in the text and in tables.

NMB is defined as $(Mod - Obs)/Obs * 100\%$, with *Mod* and *Obs* standing for the mean of model results and observations, respectively. NMB can range from $-100\%$ meaning the model predicts zero concentrations; over NMB $= 0$ meaning that the modelled average is equal to the observed average; to NMB $= 100\% \ldots 200\% \ldots 300\%$ etc. meaning that the modelled average is 2 … 3 … 4 … times the observed average.

Most of the data pre- and post-processing, statistical analysis as well as the graphical presentation was carried out using the R-Studio software (version 3.x and 4.x, R core team, 2021) including a variety of user packages (e.g. openair, ggplot2, plotly, shiny, markdown).

## 3. Results and discussion

### 3.1. Modelled concentrations at monitoring stations Denmark

In the accompanying paper (Frohn et al., 2021) results from the regional model (DEHM) have been compared with regional and suburban background stations in Denmark and Europe and results from the urban model (UBM) have been compared with regional, suburban and urban background stations in Denmark. Here we will concentrate on the performance evaluation of the complete DEHM/UBM/OSPM modelling system for the Danish monitoring stations because the main intended area of model applications are health effect studies for Danish citizens. In order to show the progression in the modelling system and to illustrate the contributions from the different parts of the modelling chain, selected results from DEHM and UBM are shown in some figures and tables as well in this paper. Moreover, DEHM/UBM results are part of the final output from our air pollution modelling system at all locations away from busy streets and evaluations for those locations are given here for reasons of consistency and completeness.

In the following, evaluation results at four levels are reported, OSPM is only applied additionally to DEHM/UBM for street locations as indicated below:

| Levels | Models | Stations |
|---|---|---|
| Street | DEHM/UBM/OSPM | Jagtvej (very short data series only) and HCAB |
| Urban | DEHM/UBM | HCØ |
| Suburban | DEHM/UBM | Hvidovre (very short data series only) |
| Regional | DEHM/UBM | Lille Valby/Risø |

For the street station HCAB, Fig. 2 shows the time series of modelled and observed PNC concentrations for the nearly two decades, covering

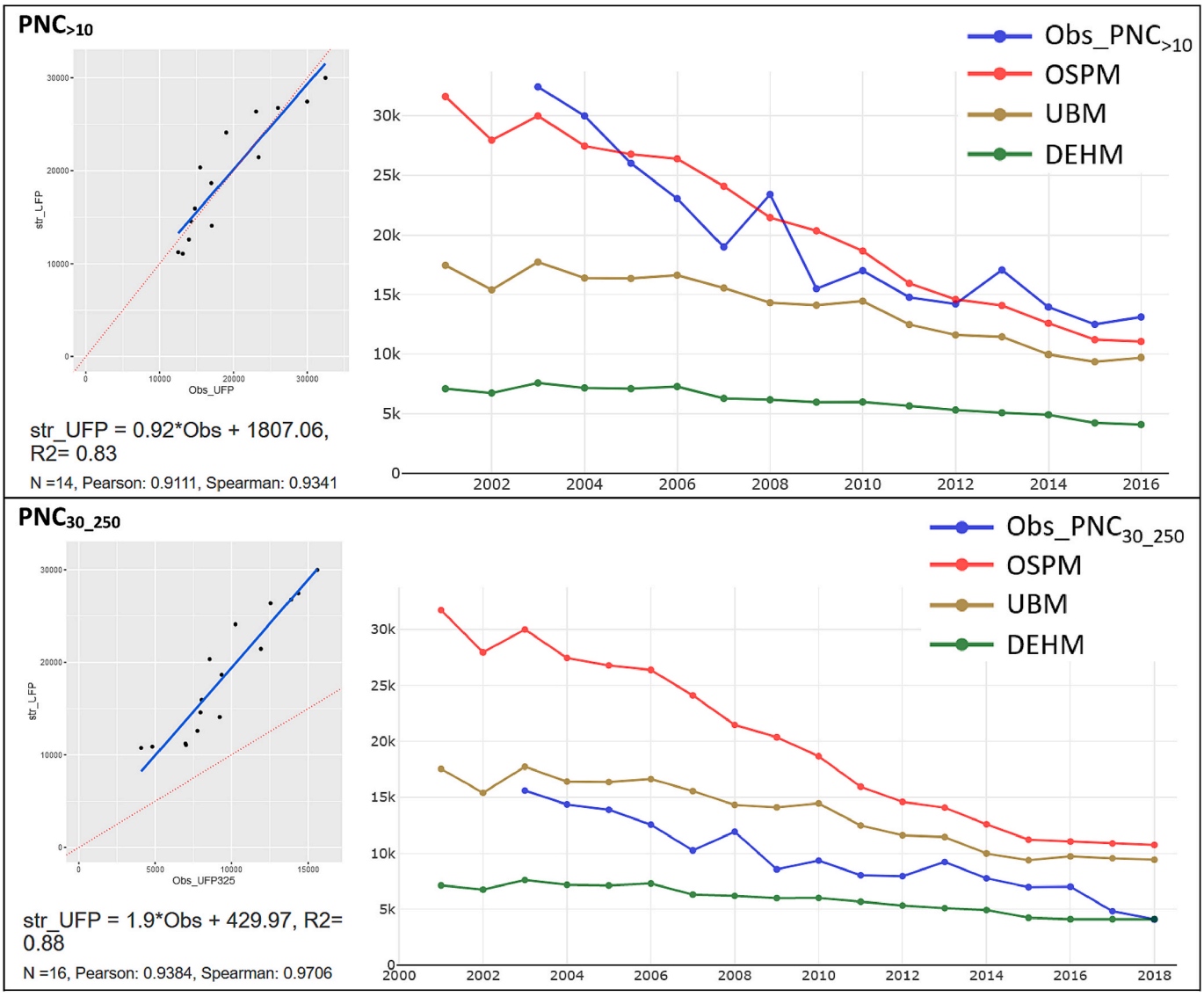


**Fig. 2.** Comparison of modelled and observed annual mean PNC (in #/cm$^3$) at HCAB. "Obs" = observations, "DEHM" = Regional model contribution, 25 km upstream input to UBM "UBM" = urban background model contribution added, "OSPM" = street model contribution added, final model estimate. **Right column:** Time series plots. The modelled contributions from **reg**ional, **urb**an background and **str**eet scale are shown separately. **Left column:** Scatter plot between modelled street concentrations and observed PNC. Modelled concentrations are compared with the two size ranges in the observed PNC: **Upper row:** Observed $PNC_{>10}$ (only until 2016) **Lower row:** Observed $PNC_{30\_250}$.

the time span with available observed PNC data. The contributions from the different parts of the modelling system from DEHM, UBM and finally OSPM are visible in progressive concentration levels.

A strong decreasing trend of PNC was estimated in the model (OSPM) between 2000 and 2018 from about 35,000 to 12,000 #/cm$^3$ at HCAB A similar decreasing trend was observed in PNC measurements in the corresponding time period. OSPM matches the measured levels well with very high $R_P$ of 0.91–0.94 and with NMB = 4% for observed $PNC_{>10}$ (upper row), and overestimates $PNC_{30\_250}$ with NMB = 94% (lower row).

Fig. 3 shows the aggregated average time variation of modelled and observed PNC (in #/cm$^3$) for hours of the day, days of the week and months of the year. Both the difference between weekdays and weekends as well as the seasonal variation of the particle number concentrations are well reproduced by the model for $PNC_{>10}$. As expected, lower PNCs are observed and modelled in the weekends, while the monthly variations show higher values in spring/fall and lower values in the summer. These basic trends are well reproduced by the OSPM model.

The levels of $PNC_{30\_250}$ are about 50% lower but show a similar time variation.

Table 3 provides the correlation matrix (Pearson) between modelled and observed PNC data at the HCAB location. The modelled PNC, including the street contribution (with OSPM, str_UFP) shows higher correlations with observations compared to similar values for UBM contribution only. This means adding OSPM to the modelling chain improves the prediction quality as the Pearson correlation coefficient increases by a value of about 0.1. Very similar correlation coefficients are observed between model (str_UFP) and $PNC_{30\_250}$ (0.62–0.94) compared to $PNC_{>10}$ (0.64–0.92). The correlation coefficient between $PNC_{>10}$ and $PNC_{30\_250}$ is very high (0.97–0.99) for this station. This indicates that the particles in size ranges 10–30 nm and 30–250 nm are having very similar time variation at the street scale, due to the same dominating source of traffic emissions with similar number size distributions.

Fig. 4 gives an example of the model performance at higher temporal resolution for daily averages at HCAB in a period in 2008. Correlation coefficients $R_P$ are between 0.72 and 0.74, i.e. in agreement with the values shown in Table 3 for the $R_P$ for daily averages for the complete

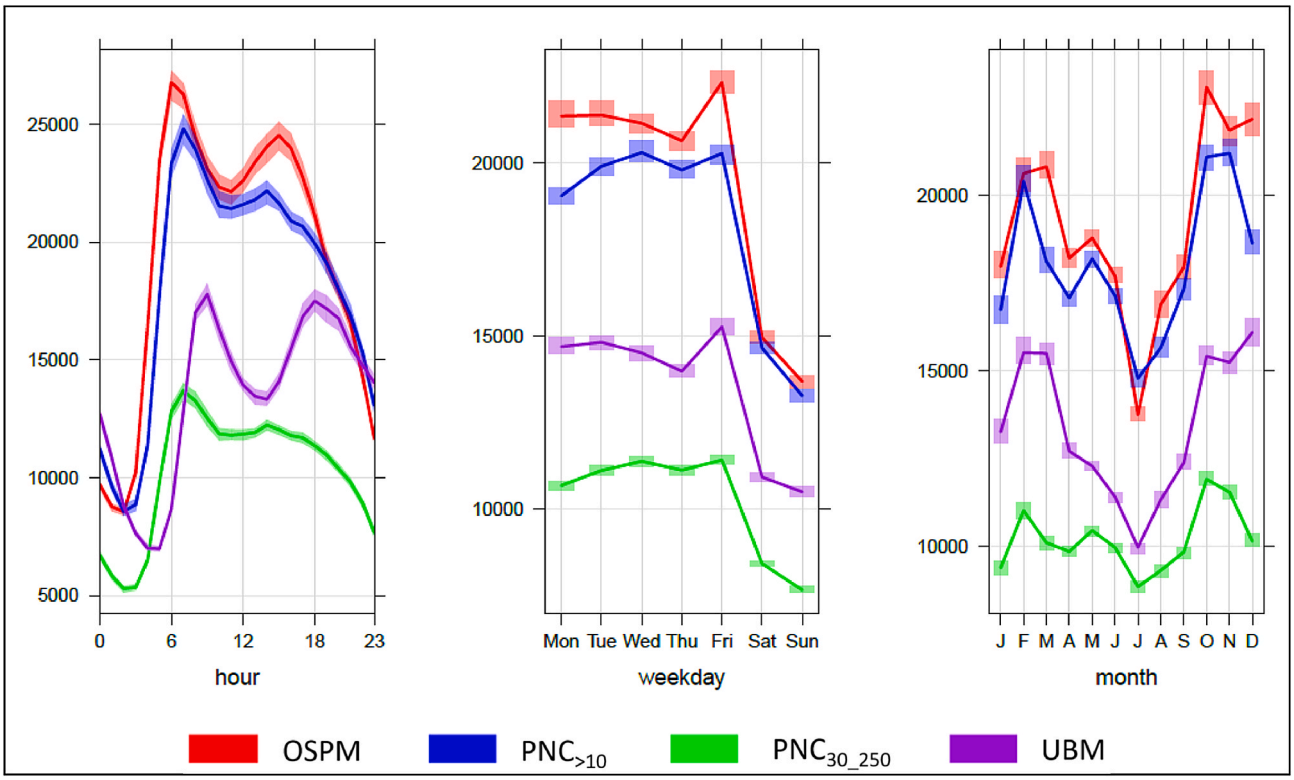


**Fig. 3.** Aggregated average time variation of modelled (OSPM, UBM) and observed $PNC_{>10}$ and $PNC_{30\_250}$ concentrations (in #/cm$^3$) at the street station HCAB for the years 2002–2016. Shaded area indicates the 95% confidence interval of the mean. Variations are shown for 1) hours of the day (both working days and weekends) 2) days of the week 3) month of the year.

**Table 3**
Correlation matrix (Pearson) between the modelled (UBM, str_UFP = OSPM) and observed ($PNC_{>10}$, $PNC_{30\_250}$) components at HCAB for the years 2002–2016. Data are given for hourly, daily, monthly and annual aggregation in time.

Hourly N = 75843

| | str_UFP | N_10_999 | N_30_250 |
|---|---|---|---|
| str_UFP | 1.00 | 0.64 | 0.62 |
| N_10_999 | 0.64 | 1.00 | 0.97 |
| N_30_250 | 0.62 | 0.97 | 1.00 |
| UBM | 0.66 | 0.49 | 0.50 |

Daily N = 3274

| | str_UFP | N_10_999 | N_30_250 |
|---|---|---|---|
| str_UFP | 1.00 | 0.71 | 0.71 |
| N_10_999 | 0.71 | 1.00 | 0.97 |
| N_30_250 | 0.71 | 0.97 | 1.00 |
| UBM | 0.95 | 0.63 | 0.64 |

Monthly N = 141

| | str_UFP | N_10_999 | N_30_250 |
|---|---|---|---|
| str_UFP | 1.00 | 0.82 | 0.82 |
| N_10_999 | 0.82 | 1.00 | 0.98 |
| N_30_250 | 0.82 | 0.98 | 1.00 |
| UBM | 0.94 | 0.73 | 0.72 |

Yearly N = 15

| | str_UFP | N_10_999 | N_30_250 |
|---|---|---|---|
| str_UFP | 1.00 | 0.92 | 0.94 |
| N_10_999 | 0.92 | 1.00 | 0.99 |
| N_30_250 | 0.94 | 0.99 | 1.00 |
| UBM | 0.98 | 0.86 | 0.89 |

dataset.

In order to illustrate the model performance for another street location, Jagtvej, Fig. 5 shows the comparison for the short period of available data in 2001–2004. Here weekly averages are presented, and Pearson correlation coefficients of 0.56 and 0.63 are obtained in the comparison with $PNC_{>10}$ and $PNC_{30\_250}$, respectively.

Table 4 summarizes the model performance at the three Danish stations with long data series available (Lille Valby/Risø, HCØ, HCAB). Correlation coefficients (Pearson) are given (as far as available) for hourly, daily, monthly and annual averaging periods. Besides $PNC_{30\_250}$, the pollutants $NO_X$, $PM_{2.5}$ and $PM_{10}$ are also analysed. PNC and $NO_X$ are pairwise synchronised before calculating the correlation coefficient, therefore the number of elements are the same between these two. However, small differences in values may appear in comparison to results in the accompanying paper where PNC was not synchronised with $NO_X$.

The correlations between model results and observations for $PNC_{30\_250}$ are in the range from moderate to very high (0.4–0.9). Correlation coefficients are especially high for the street level station HCAB (range 0.63–0.95) and are here comparable or even higher than those for the other three pollutants ($NO_X$ $PM_{2.5}$ and $PM_{10}$) independent on the averaging interval. For $PNC_{30\_250}$ at Lille Valby/Risø and HCØ we find a significant difference between the higher correlation coefficients for annual means (0.86, 0.87) compared to the moderate values for other averaging times (0.39–0.51). This might indicate that our new PNC model captures well the long-term trends at all stations, while variations on shorter time scales (e.g. seasonal variations) are not as well reproduced by the model for regional and urban background stations while showing very good results at street scale.

In summary, as indicated by the number of highlighted $R_P$ in Table 4, the model performance with respect to correlation in time for PNC is similar to those of $NO_X$ and $PM_{2.5}$. The PNC model performance was better than that for $PM_{10}$ at street level.

Table 5 gives the Normalized Mean Bias (NMB) for the same three stations as discussed above for the comparison of the modelled PNC with measured PNC both as $PNC_{>10}$ and $PNC_{30\_250}$. Modelled PNC at HCAB matches well (NMB = 6%) with observed $PNC_{>10}$ while the model overpredicts the level of $PNC_{>10}$ at HCØ and Lille Valby/Risø by a factor of

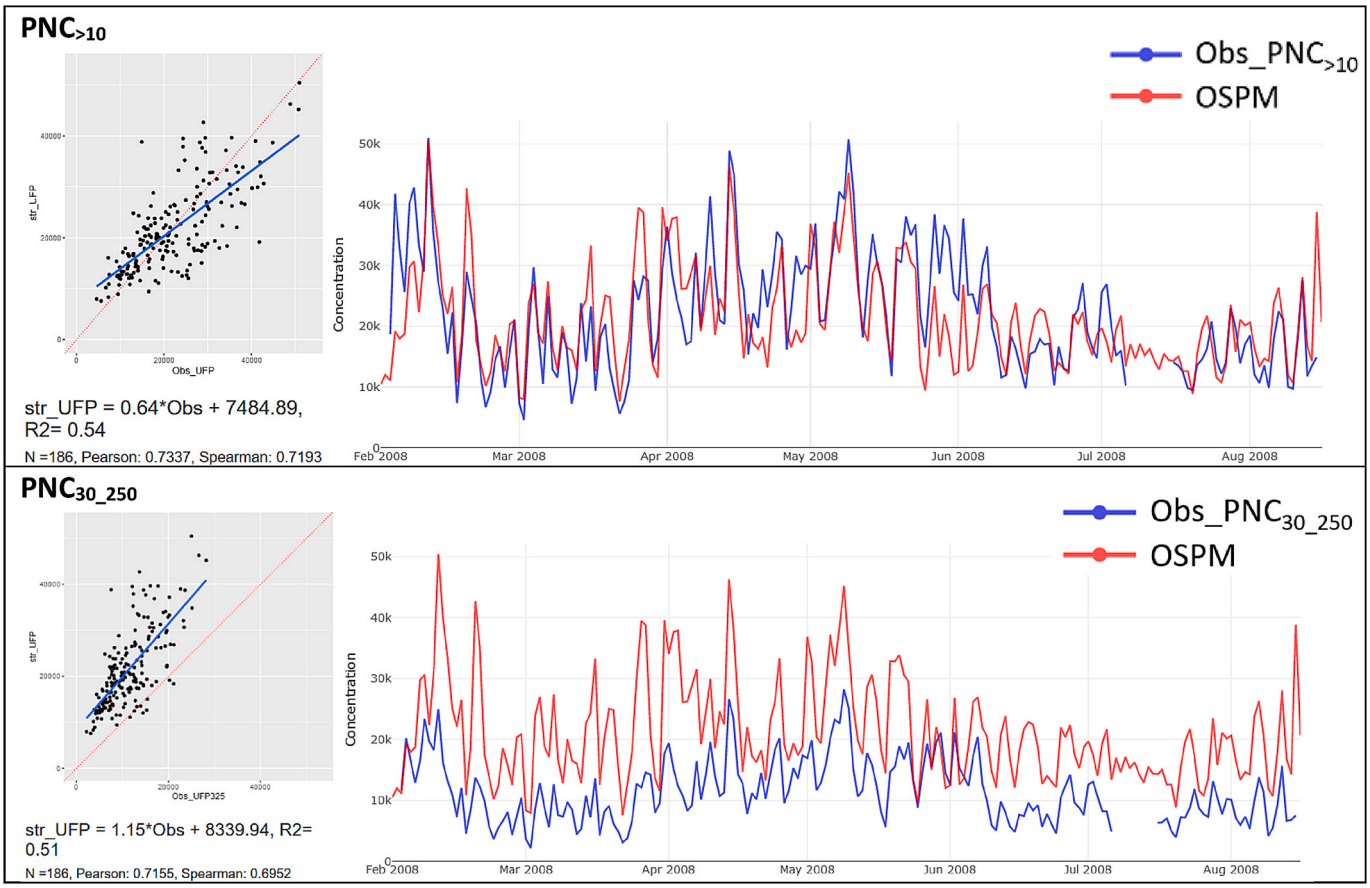


**Fig. 4.** Plots as in Fig. 2 as well for HCAB station, Here: **daily averages** for a 6-month period in 2008.

2.5–2.9 (NMB = 151%/193%). Shifting the comparison from $PNC_{>10}$ to $PNC_{30\_250}$ adds about 100% to the NMB at all stations increasing the overestimation by the model system, since the observed $PNC_{30\_250}$ is about 50% of $PNC_{>10}$.

These values of NMB are high compared to the model performances achieved for other pollutants, e.g. $NO_X$, $PM_{2.5}$, $PM_{10}$ (NMB range −25%–45%, not shown here). These high NMB values for the PNC model lead to the question about how well the PNC model is able to reproduce the concentration contrast observed in-between the different environments such as street, urban background and rural levels.

Fig. 6 (a-c, upper row) shows the modelled and observed annual mean concentrations at four Danish stations, observations for $PNC_{>10}$ and $PNC_{30\_250}$. In addition to the three stations discussed until now this comparison includes the sub-urban station Hvidovre in the outskirts of Copenhagen. For Hvidovre only a very short observed time series of four years is available, starting in 2015, and only for $PNC_{30\_250}$.

The modelled concentrations appear consistently in the same ranking when considering the PNC: HCAB highest, then HCØ, Hvidovre and Lille Valby/Risø with lowest PNC. For the measurements, the same ranking is observed with a few exceptions, probably due to a lower data coverage for specific stations in some years.

Fig. 6 (d-f, lower row) shows for the same type of stations the annual mean trends relative to the levels at HCØ, i.e. all concentrations were divided or normalized by the concentrations at HCØ in the actual year. The curves show herewith the ratio of concentrations relative to HCØ. As an example, modelled PNC at HCAB are for a long part of the time interval 1979–2009 about 1.5–2 times higher compared to HCØ (Fig. 6d). The observations at HCAB show about 3–4.5 times higher values than HCØ for $PNC_{>10}$ (Fig. 6e) while for $PNC_{30\_250}$ the ratios are in the range 2.5–3.5 (Fig. 6f). For both $PNC_{>10}$ and $PNC_{30\_250}$ the observed "concentration contrast" between HCAB and HCØ is larger compared to the modelled PNC ratios. Contrary to this, the ratio of PNC at Lille Valby/Risø compared to HCØ is in the range 0.6–0.8 in all cases, both for modelled PNC and observed $PNC_{>10}$ and $PNC_{30\_250}$.

This analysis might indicate that the PNC differences are more robustly reproduced by the PNC model system in the lower part of the concentration range (rural/urban) than in the upper part (street/urban background). Another possible reason for the larger HCAB/HCØ ratio in the measured PNC, compared to the modelled ratio, could be that HCØ concentrations are measured at the roof top level at 20 m height, and PNC might be reduced due to deposition and coagulation during the transport from ground level. Higher measured HCØ concentrations would lower the ratio and shift this ratio closer towards the modelled HCAB/HCØ ratio.

### 3.2. *Modelled concentrations at high spatial resolution for entire Denmark*

The developed PNC model system covers all Danish addresses 40 years back in time with hourly resolution. This data is intended to indicate the variation in PNC at home addresses across the entire Danish population to be used e.g. in epidemiological studies of health effects of air pollution. While the previous sections focussed on the presentation and evaluation of the temporal variation, we give in the following some examples for the modelled spatial distribution.

Fig. 7 shows the annual mean PNC for the year 2018 for Denmark on a 1 km × 1 km spatial resolution modelled with DEHM/UBM. General PNC are modelled in the range between 3,000 and 38,000 #/$cm^3$. High PNC concentrations are modelled in urban areas, near harbours and along shipping routes, along intense busy motorways, and near point sources such as power plants and industrial facilities. Low concentrations are predicted at the less populated west coast of Denmark and in

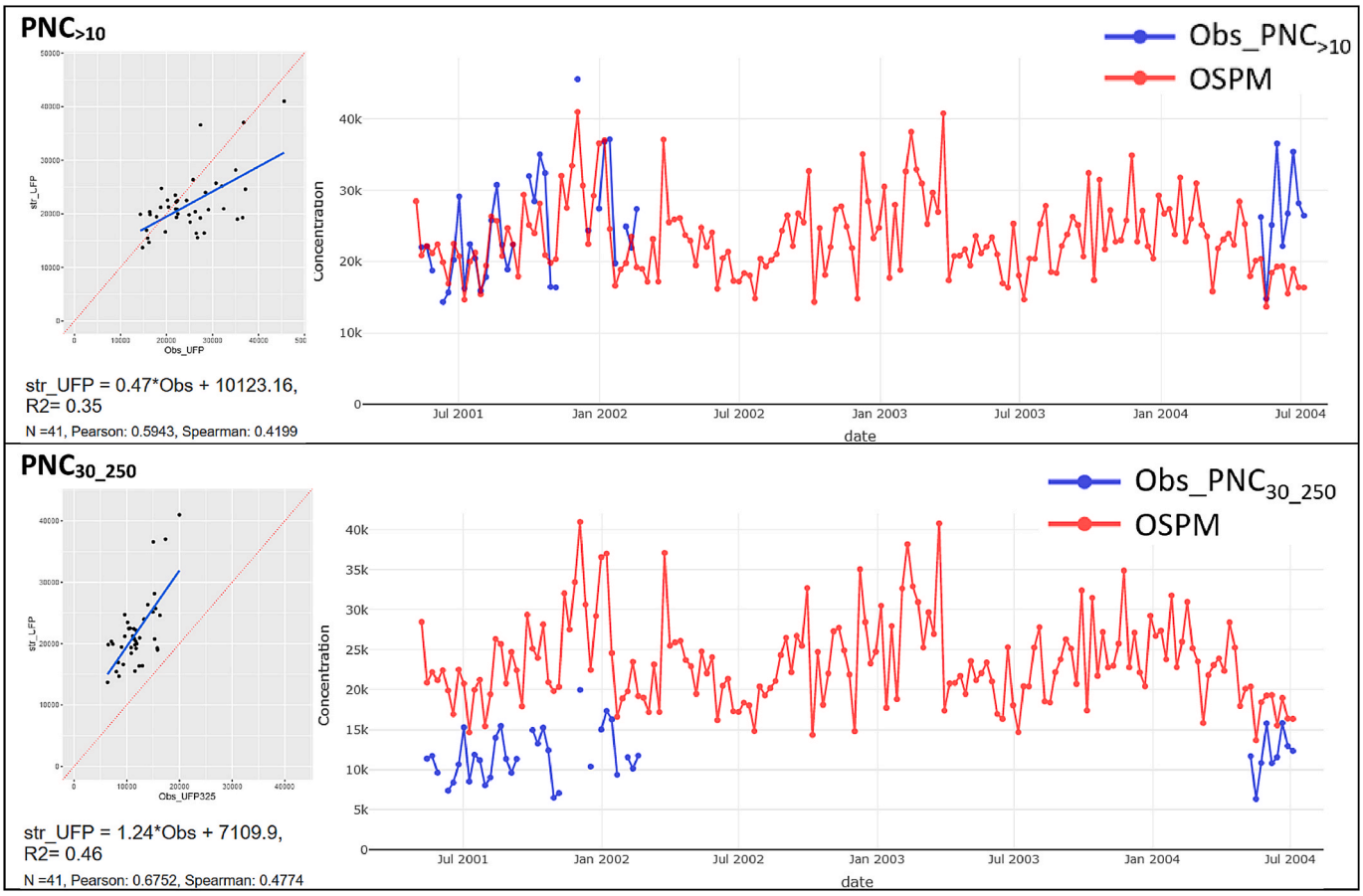


**Fig. 5.** Plots as in Fig. 2 now for **Jagtvej**, Here: **weekly** averages for a period in 2001–2004 with available PNC observations for Jagtvej.

**Table 4**
Correlation between model and measured concentrations at 3 Danish stations LVBY/Risø, HCØ and HCAB for the pollutants: $PNC_{30\_250}$, $NO_X$, $PM_{2.5}$ and $PM_{10}$. Correlation coefficients $R_P$ are given for different averaging times (hourly, daily, monthly and yearly). Only PM data obtained with LVS (Low Volume Sampler) instruments are included (reference method, considered most reliable). $R_P$ values > 0.7 are indicated bold and values > 0.5 are italics. R- Pearson correlation coefficient and N – number of elements in the correlation.

| Pollutant | Scale Station | $R_P$ hourly | $R_P$ daily | $R_P$ monthly | $R_P$ annual | N hourly | N daily | N monthly | N annual |
|---|---|---|---|---|---|---|---|---|---|
| $PNC_{30\_250}$ | Regional LVBY/Risø | 0.40 | *0.51* | 0.41 | **0.86** | 79450 | 3496 | 152 | 14 |
| $PNC_{30\_250}$ | Urban bg. HCØ | 0.39 | 0.48 | 0.43 | **0.87** | 84553 | 3712 | 171 | 18 |
| $PNC_{30\_250}$ | Street HCAB | *0.63* | **0.73** | **0.84** | **0.95** | 88586 | 3846 | 163 | 17 |
| $NO_X$ | Regional LVBY/Risø | *0.50* | *0.69* | **0.74** | **0.92** | 79450 | 3469 | 152 | 14 |
| $NO_X$ | Urban bg. HCØ | *0.59* | **0.75** | **0.86** | **0.94** | 84553 | 3712 | 171 | 18 |
| $NO_X$ | Street HCAB | *0.63* | *0.68* | *0.62* | *0.61* | 88586 | 3846 | 163 | 17 |
| $PM_{2.5}$ | Regional LVBY/Risø | – | **0.80** | **0.89** | **0.82** | – | 2230 | 76 | 7 |
| $PM_{2.5}$ | Urban bg. HCØ | – | **0.77** | **0.86** | **0.72** | – | 2217 | 77 | 7 |
| $PM_{2.5}$ | Street HCAB | – | **0.75** | **0.84** | **0.89** | – | 2164 | 75 | 6 |
| $PM_{10}$ | Regional LVBY/Risø | – | *0.69* | **0.73** | **0.90** | – | 1767 | 60 | 5 |
| $PM_{10}$ | Urban bg. HCØ | – | *0.62* | *0.64* | 0.42 | – | 1944 | 66 | 6 |
| $PM_{10}$ | Street HCAB | – | *0.62* | *0.54* | 0.22 | – | 2223 | 75 | 7 |

the northern parts of the country. The overall concentration map for Denmark looks realistic and shows similarities to the spatial distribution of other pollutants as e.g. $NO_X$.

Fig. 8 displays the PNC modelled with DEHM/UBM/AirGIS as the annual mean for 2018 at residential address locations in the centre of Copenhagen. The general PNC levels in Copenhagen result in values in the range of 6,000 to 12,000 #/$cm^3$. High concentrations are shown along very busy roads and here the OSPM model has been applied in addition to the DEHM/UBM. The predicted PNC along the same road with the same emissions might change according to the more or less dense building configuration along the street. This is a result of the influence of the height of the surrounding buildings and the width of the street on the flow and dispersion of air pollutants at the street location.

## 4. Summary, conclusions and outlook

We have presented the development of the Danish air quality modelling system DEHM/UBM/AirGIS towards modelling, for the first

**Table 5**
Normalized Mean Bias (NMB) in % for three Danish PNC monitoring stations. Compared are modelled PNC with measured $PNC_{>10}$ and $PNC_{30\_250}$ for the same time series as presented in Table 4.

| Scale | Station | NMB compared to $PNC_{>10}$ | NMB compared to $PNC_{30\_250}$ |
|---|---|---|---|
| Regional | Lille Valby/Risø | 193% | 292% |
| Urban background | HCØ | 151% | 259% |
| Street | HCAB | 6% | 89% |

time, particle number concentration (PNC). We implemented particle dynamical processes in the regional scale model DEHM using the M7 aerosol dynamics module (presented in the accompanying article by Frohn et al., 2021), and we implemented emission factors in the models for PNC at the local scale (UBM) and street scale (OSPM), for the latter two in a first approximation without a particle dynamics module as presented in the present article.

Here, we described the model development at local and street scale and evaluated the PNC model performance using long-term measurements at Danish monitoring stations at street, urban background and rural locations. The PNC models are intended to serve as proxy for the human exposure in epidemiological studies.

The very complex processes of particle formation, where nucleation mode particles are formed from gaseous precursors, are mostly observed and relevant at rural locations with very low population density. Therefore, modelling the nucleation mode at rural level was beyond the scope of this work. Consequently, model performance was evaluated using observed data for $PNC_{30\_250}$, hereby excluding the nucleation mode particles from regional nucleation events. Therefore, at rural monitoring stations, the model performance (correlation coefficient) is considerably better for $PNC_{30\_250}$ compared to $PNC_{>10}$ (Frohn et al., 2021), since our PNC model is not considering the regional nucleation events which matches best with measured $PNC_{30\_250}$ excluding nucleation mode particles as well.

However, at urban and street level, particles smaller than 30 nm are as well locally produced and primarily emitted, e.g. from traffic. These locally produced particles are included in the local emissions in the UBM and OSPM model. Therefore, we also evaluated the OSPM model for $PNC_{>10}$ for these stations. As confirmation that our model is considering these small particles, we observed only small differences in correlation coefficients between evaluations with $PNC_{30\_250}$ versus $PNC_{>10}$ at urban and street stations.

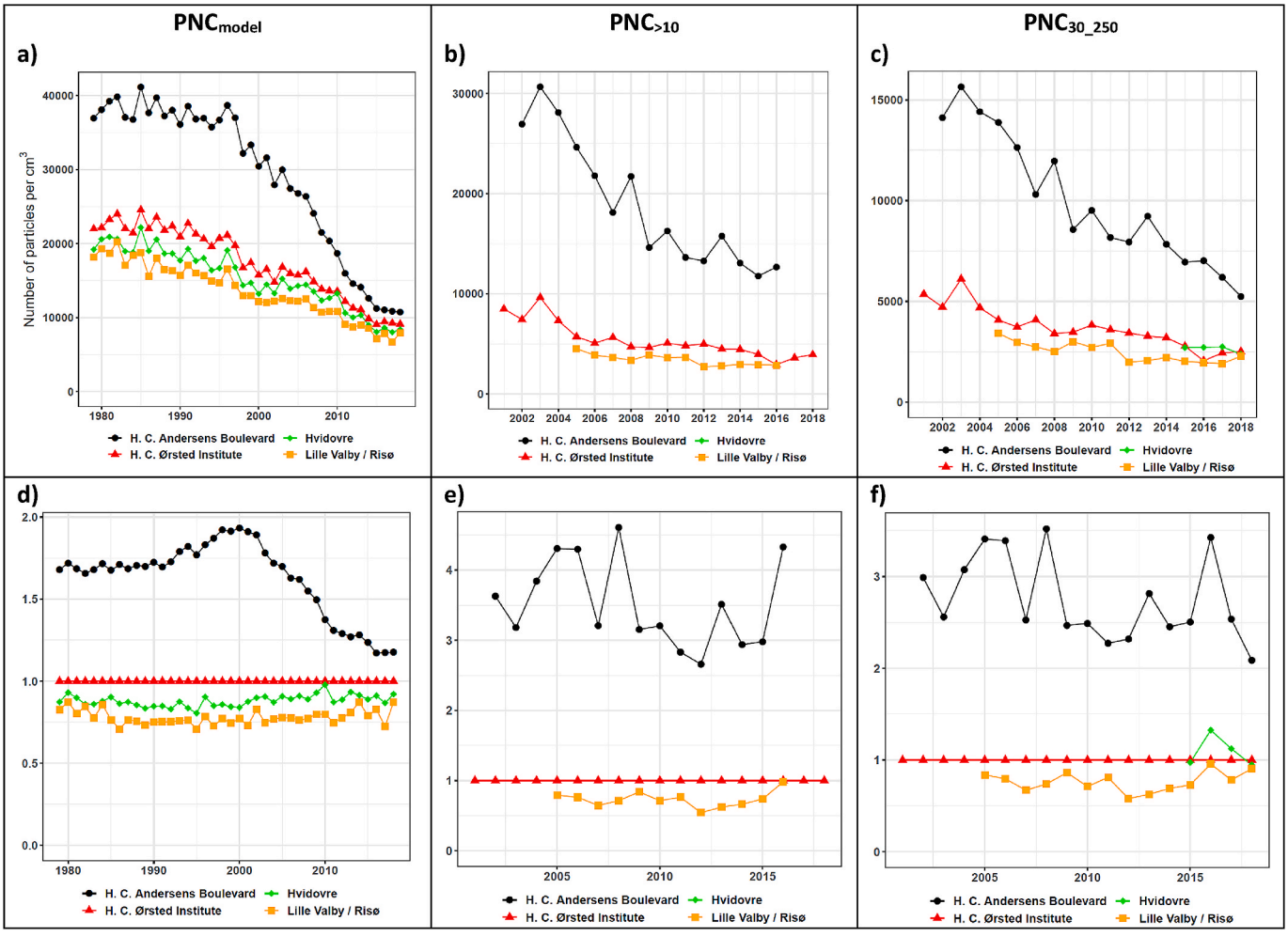


**Fig. 6.** Annual average concentrations at four Danish monitoring stations for 40 years 1979–2018. The plots illustrate the relative contrast between concentration levels at the various stations over time. Be aware of different ranges on both axes in-between the different plots. **Left column:** $PNC_{mod}$ OSPM/UBM Model results **Middle column:** Observed $PNC_{>10}$ **Right column:** Observed $PNC_{30\_250}$. **Upper Row:** Annual averages of PNC modelled or observed. **Lower Row:** PNC relative to HCØ. For each of the individual plots, the data from all stations are divided with the value at HCØ for the same year.

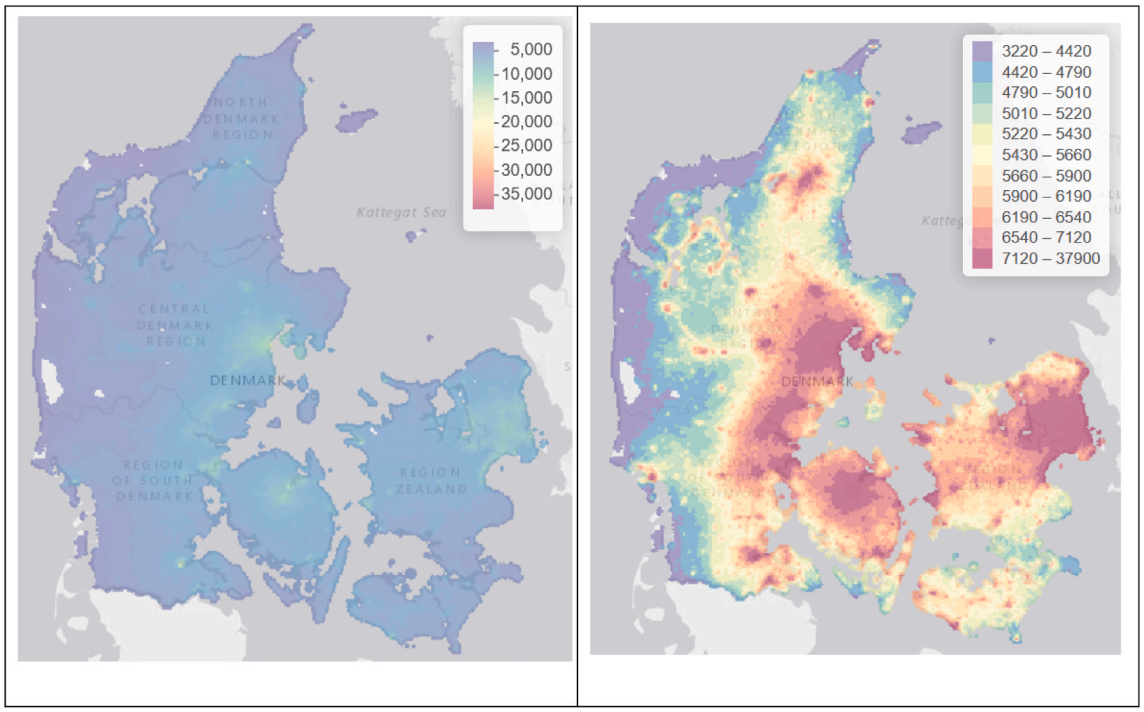


**Fig. 7.** PNC (#/cm$^3$) modelled with DEHM/UBM at 1 km × 1 km spatial resolution as annual average for 2018. Only areas with Danish residential addresses are displayed. Empty cells are unpopulated e.g. water or forest. The same data is displayed in both maps, just different colour scales:
**Left:** linear equidistant scale, **Right:** quantile scale, with very variable ranges in the colour classes, arranged so that each colour covers a similar total area in the map. Background: ©ESRI.WorldGrayCanvas. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

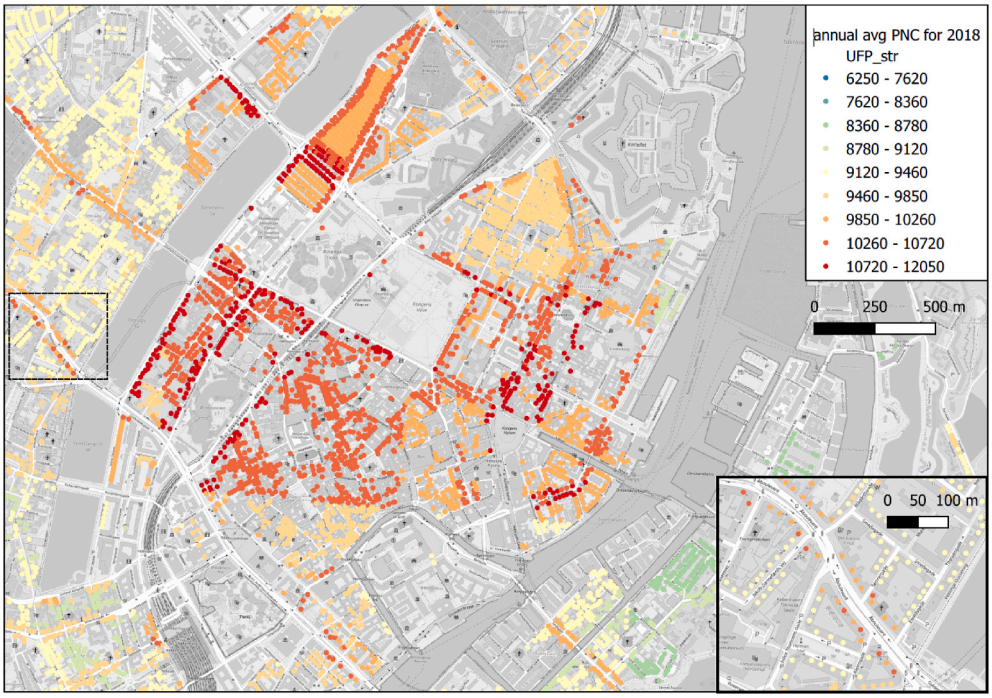


**Fig. 8.** PNC (#/cm$^3$) modelled with DEHM/UBM/AirGIS at address locations in central Copenhagen as an annual average for 2018. The smaller map is an enlarged detailed view of the marked dashed region in the big map showing the location of individual address points. Background: ©OSM-Open Street Map.

The correlation coefficients between model results and observations for $PNC_{30\_250}$ are in the range from 0.39 to 0.95 for all stations (annual values). Correlation coefficients are especially high for the street level monitoring station (range 0.63–0.95) for all averaging periods (hourly, daily, monthly, annually). The model performance for PNC is similar to that of $NO_X$ and $PM_{2.5}$ with respect to correlation in time. The PNC model performance was better than that for $PM_{10}$ at street level.

In terms of reproducing the absolute PNC levels, the model performance is mixed. Modelled PNC at HCAB (street with dense traffic) matches well (NMB = 6%) with observed $PNC_{>10}$ while the model

overpredicts compared to measured $PNC_{>10}$ at HCØ (urban background) and Lille Valby/Risø (rural background) by a factor of 2.5–2.9 (NMB = 151%/193%). A possible reason for the model overprediction at the HCØ station could be that HCØ concentrations are measured at the roof top level in 20 m height and PNC might be reduced due to deposition and coagulation during the transport from ground level, while modelled PNC is more representative for ground ground-level PNC. Shifting the comparison from $PNC_{>10}$ to $PNC_{30\_250}$ adds about 100% to the NMB at all stations increasing the overestimation of the model, since the observed $PNC_{30\_250}$ is about 50% of $PNC_{>10}$.

This is the first implementation of PNC in our modelling system, and future developments could relate to: 1) improving the implementation of PNC size fractions in all emission inventories and all emission factors to work towards an implementation of size resolved PNC in the UBM and OSPM models, 2) improving the seasonal variation of PNC at background level by updating and improving seasonal variation of emissions, 3) reanalysis of the methods for handling the particle dynamics in the model and 4) validation of the model against many measurements with wide geographical variation. These improvements are expected to reduce the substantial positive bias of the modelled PNC.

In summary, we obtained high correlation coefficients between modelled and measured PNC, especially at street stations and for long-term averages. At monitoring stations at urban and rural background locations, the model over-predicts PNC. We believe that further work regarding the emission estimates and the modelling of the complex dynamical processes will improve the model results further.

## Author contributions

Matthias Ketzel, 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, 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.

Jibran Khan: Supervision of the AirGIS/OSPM model runs as well as pre- and post-processing of the modelled data, GIS inputs and databases (including street configuration inputs) for the OSPM®

Ulas Im, Steen S. Jensen, Ole-Kenneth Nielsen, Marlene S. Plejdrup: obtaining and processing Danish traffic data, GIS databases and 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 competing interest

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.

## Acknowledgements

The authors would like to thank Ghadi Alakrami and Thomas Schrøder Terneborn for their valuable help in the complex data management of air pollution model results and observed PNSD data.

We acknowledge the Danish Environmental Protection Agency (Miljøstyrelsen, mst.dk) for financial support to conduct PNSD measurements in Denmark.

Research described in this article was conducted under contract to the Health Effects Institute, United States, (HEI), an organization jointly funded by the United States Environmental Protection Agency (EPA) (Assistance Award No. R-82811201) and certain motor vehicle and engine manufacturers. The contents of this article do not necessarily reflect the views of HEI, or its sponsors, nor do they necessarily reflect the views and policies of the EPA or motor vehicle and engine manufacturers.

The Health Effects Institute, United States (HEI) funded the project "Health Effects of Air Pollution Components, Noise and Socioeconomic Status (HERMES)" with one of the aims to establish particle number/UFP as a new pollution component in all three Danish air pollution models that are used for assessment of concentrations at home addresses: the regional model, Danish Eulerian Hemispheric Model (DEHM), the Urban Background Model (UBM) and the Operational Street Pollution Model (OSPM®). Link: https://www.healtheffects.org/research/ongoing-research/HERMES-health-effects-air-pollution-components-noise-and-SES.

## References

ACTRIS - European center for aerosol calibration (ECAC) http://www.actris-ecac.eu/.

Asmi, A., Wiedensohler, A., Laj, P., Fjaeraa, A.M., Sellegri, K., Birmili, W., Weingartner, E., Baltensperger, U., Zdimal, V., Zikova, N., Putaud, J.P., Marinoni, A., Tunved, P., Hansson, H.C., Fiebig, M., Kivekas, N., Lihavainen, H., Asmi, E., Ulevicius, V., Aalto, P.P., Swietlicki, E., Kristensson, A., Mihalopoulos, N., Kalivitis, N., Kalapov, I., Kiss, G., de Leeuw, G., Henzing, B., Harrison, R.M., Beddows, D., O'Dowd, C., Jennings, S.G., Flentje, H., Weinhold, K., Meinhardt, F., Ries, L., Kulmala, M., 2011. Number size distributions and seasonality of submicron particles in Europe 2008-2009. Atmos. Chem. Phys. 11 (11), 5505–5538.

Baranizadeh, E., Nieminen, T., Yli-Juuti, T., Kulmala, M., Petaja, T., Leskinen, A., Komppula, M., Laaksonen, A., Lehtinen, K.E.J., 2017. Estimation of atmospheric particle formation rates through an analytical formula: validation and application in Hyytiala and Puijo, Finland. Atmos. Chem. Phys. 17 (21), 13361–13371.

Berkowicz, R., 2000. OSPM - a parameterised street pollution model. Environ. Monit. Assess. 65 (1–2), 323–331.

Birmili, W., Alaviippola, B., Hinneburg, D., Knoth, O., Tuch, T., Borken-Kleefeld, J., Schacht, A., 2009a. Dispersion of traffic-related exhaust particles near the Berlin urban motorway – estimation of fleet emission factors. Atmos. Chem. Phys. 9 (7), 2355–2374.

Birmili, W., Weinhold, K., Nordmann, S., Wiedensohler, A., Spindler, G., Mueller, K., Herrmann, H., Gnauk, T., Pitz, M., Cyrys, J., Flentje, H., Nickel, C., Kuhlbusch, T.A. J., Loschau, G., Haase, D., Meinhardt, F., Schwerin, A., Ries, L., Wirtz, K., 2009b. Atmospheric aerosol measurements in the German ultrafine aerosol network (GUAN). Gefahrst. Reinhalt. Luft 69 (4), 137–145.

Birmili, W., Weinhold, K., Rasch, F., Sonntag, A., Sun, J., Merkel, M., Wiedensohler, A., Bastian, S., Schladitz, A., Loschau, G., Cyrys, J., Pitz, M., Gu, J.W., Kusch, T., Flentje, H., Quass, U., Kaminski, H., Kuhlbusch, T.A.J., Meinhardt, F., Schwerin, A., Bath, O., Ries, L., Wirtz, K., Fiebig, M., 2016. Long-term observations of tropospheric particle number size distributions and equivalent black carbon mass concentrations in the German Ultrafine Aerosol Network (GUAN). Earth Syst. Sci. Data 8 (2), 355–382.

Brandt, J., Christensen, J.H., Frohn, L.M., Berkowicz, R., 2001. Operational air pollution forecast from regional scale to urban street scale. Part 1: system description. Phys. Chem. Earth 26 (10), 781–786.

Brandt, J., Christensen, J.H., Frohn, L.M., Berkowicz, R., 2003. Air pollution forecasting from regional to urban street scale - implementation and validation for two cities in Denmark. Phys. Chem. Earth 28 (8), 335–344.

Brandt, J., Silver, J.D., Frohn, L.M., Geels, C., Gross, A., Hansen, A.B., Hansen, K.M., Hedegaard, G.B., Skjoth, C.A., Villadsen, H., Zare, A., Christensen, J.H., 2012. An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport of air pollution. Atmos. Environ. 53, 156–176.

Chen, X., Yu, F., Yang, W., Sun, Y., Chen, H., Du, W., Zhao, J., Wei, Y., Wei, L., Du, H., Wang, Z., Wu, Q., Li, J., An, J., Wang, Z., 2020. Global-regional nested simulation of particle number concentration by combing microphysical processes with an evolving organic aerosol module. Atmos. Chem. Phys. Discuss. 2020, 1–48.

de Jesus, A.L., Rahman, M.M., Mazaheri, M., Thompson, H., Knibbs, L.D., Jeong, C., Evans, G., Nei, W., Ding, A.J., Qiao, L.P., Li, L., Portin, H., Niemi, J.V., Timonen, H., Luoma, K., Petaja, T., Kulmala, M., Kowalski, M., Peters, A., Cyrys, J., Ferrero, L., Manigrasso, M., Avino, P., Buonano, G., Reche, C., Querol, X., Beddows, D., Harrison, R.M., Sowlat, M.H., Sioutas, C., Morawska, L., 2019. Ultrafine particles and PM2.5 in the air of cities around the world: are they representative of each other? Environ. Int. 129, 118–135.

Denier van der Gon, H.A.C., et al., 2014. European Particle Number Emissions for 2005, 2020 and 2030, 9th Int. Conf. On Air Quality. Garmisch-Partenkirchen.

EEA, 2019. EMEP/EEA Air Pollutant Emission Inventory Guidebook. Technical Guidance to Prepare National Emission Inventories. EEA Report No 13/2019. https://www.eea.europa.eu/publications/emep-eea-guidebook-2019.

EC, 2021. "The Second Clean Air Outlook" , Report from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the

Committee of the Regions, European Commission, Brussels, 8.1.2021; COM(2021) 3 Final, EUR-Lex - 52021DC0003 - EN - EUR-Lex (europa.Eu). FIN. https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=COM:2021:3.

Ellermann, T., Nygaard, J., Nøjgaard, J.K., Nordstrøm, C., Brandt, J., Christensen, J., Ketzel, M., Massling, A., Jensen, S.S., 2016. The Danish Air Quality Monitoring Programme. Annual Summary for 2015. ." Scientific Report from DCE – Danish Centre for Environment and Energy No. 201. http://dce2.au.dk/pub/SR201.pdf.

Frohn, L.M., Christensen, J.H., Brandt, J., 2002. Development of a high-resolution nested air pollution model - the numerical approach. J. Comput. Phys. 179 (1), 68–94.

Frohn, L.M., Ketzel, M., Christensen, J.H., Brandt, J., Im, U., Massling, A., Andersen, C., Plejdrup, M.S., Nielsen, O.-K., Manders, A., Denier van der Gon, H., Raaschou-Nielsen, O., 2021. Modelling ultrafine particle number concentrations at address resolution in Denmark from 1979 to 2018 – Part 1: regional and urban scale modelling and evaluation. Atmospheric Environment 264, 118631.

Gidhagen, L., Johansson, C., Langner, J., Foltescu, V.L., 2005. Urban scale modeling of particle number concentration in Stockholm. Atmos. Environ. 39 (9), 1711–1725.

Gidhagen, L., Johansson, C., Langner, J., Olivares, G., 2004. Simulation of NOx and ultrafine particles in a street canyon in Stockholm, Sweden. Atmos. Environ. 38 (14), 2029–2044.

Heal, M.R., Kumar, P., Harrison, R.M., 2012. Particles, air quality, policy and health. Chem. Soc. Rev. 41 (19), 6606–6630.

HEI, 2013. HEI Review Panel on Ultrafine Particles: Understanding the Health Effects of Ambient Ultrafine Particles. HEI Perspectives 3. Health Effects Institute, Boston, MA. https://www.healtheffects.org/publication/understanding-health-effects-ambient-ultrafine-particles.

Hjortebjerg, D., Andersen, A.M.N., Ketzel, M., Raaschou-Nielsen, O., Sorensen, M., 2018. Exposure to traffic noise and air pollution and risk for febrile seizure: a cohort study. Scand. J. Work. Environ. Health 44 (5), 539–546.

Hofman, J., Staelens, J., Cordell, R., Stroobants, C., Zikova, N., Hama, S.M.L., Wyche, K. P., Kos, G.P.A., Van der Zee, S., Smallbone, K.L., Weijers, E.P., Monks, P.S., Roekens, E., 2016. Ultrafine particles in four European urban environments: results from a new continuous long-term monitoring network. Atmos. Environ. 136, 68–81.

Hussein, T., Karppinen, A., Kukkonen, J., Harkonen, J., Aalto, P.P., Hameri, K., Kerminen, V.M., Kulmala, M., 2006. Meteorological dependence of size-fractionated number concentrations of urban aerosol particles. Atmos. Environ. 40 (8), 1427–1440.

Hvidtfeldt, U.A., Ketzel, M., Sørensen, M., Hertel, O., Khan, J., Brandt, J., Raaschou-Nielsen, O., 2018. Evaluation of the Danish AirGIS air pollution modeling system against measured concentrations of PM2.5, PM10, and black carbon. Environ. Epidemiol. **Latest Articles**.

Hvidtfeldt, U.A., Sorensen, M., Geels, C., Ketzel, M., Khan, J., Tjonneland, A., Overvad, K., Brandt, J., Raaschou-Nielsen, O., 2019. Long-term residential exposure to PM2.5, PM10, black carbon, NO2, and ozone and mortality in a Danish cohort. Environ. Int. 123, 265–272.

Imhof, D., Weingartner, E., Ordonez, C., Gehrigt, R., Hill, N., Buchmann, B., Baltensperger, U., 2005. Real-world emission factors of fine and ultrafine aerosol particles for different traffic situations in Switzerland. Environ. Sci. Technol. 39 (21), 8341–8350.

Jensen, S.S., Berkowicz, R., Hansen, H.S., Hertel, O., 2001. A Danish decision-support GIS tool for management of urban air quality and human exposures. Transport. Res. Transport Environ. 6 (4), 229–241.

Jensen, S.S., Ketzel, M., Becker, T., Christensen, J., Brandt, J., Plejdrup, M., Winther, M., Nielsen, O.K., Hertel, O., Ellermann, T., 2017. High resolution multi-scale air quality modelling for all streets in Denmark. Transport. Res. Transport Environ. 52, 322–339.

Jensen, S.S., Larson, T., Deepti, K.C., Kaufman, J.D., 2009. Modeling traffic air pollution in street canyons in New York City for intra-urban exposure assessment in the US Multi-Ethnic Study of atherosclerosis and air pollution. Atmos. Environ. 43 (30), 4544–4556.

Jones, A.M., Harrison, R.M., 2006. Estimation of the emission factors of particle number and mass fractions from traffic at a site where mean vehicle speeds vary over short distances. Atmos. Environ. 40 (37), 7125–7137.

Kakosimos, K.E., Hertel, O., Ketzel, M., Berkowicz, R., 2010. Operational Street Pollution Model (OSPM) - a review of performed application and validation studies, and future prospects. Environ. Chem. 7 (6), 485–503.

Karl, M., Kukkonen, J., Keuken, M.P., Lutzenkirchen, S., Pirjola, L., Hussein, T., 2016. Modeling and measurements of urban aerosol processes on the neighborhood scale in Rotterdam, Oslo and Helsinki. Atmos. Chem. Phys. 16 (8), 4817–4835.

Ketzel, M., Berkowicz, R., 2004. Modelling the fate of ultrafine particles from exhaust pipe to rural background: an analysis of time scales for dilution, coagulation and deposition. Atmos. Environ. 38 (17), 2639–2652.

Ketzel, M., Berkowicz, R., 2005. Multi-plume aerosol dynamics and transport model for urban scale particle pollution. Atmos. Environ. 39 (19), 3407–3420.

Ketzel, M., Berkowicz, R., Hvidberg, M., Jensen, S.S., Raaschou-Nielsen, O., 2011. Evaluation of AirGIS: a GIS-based air pollution and human exposure modelling system. Int. J. Environ. Pollut. 47 (1–4), 226–238.

Ketzel, M., Jensen, S.S., Brandt, J., Ellermann, T., Olesen, H.R., Berkowicz, R., Hertel, O., 2012. Evaluation of the street pollution model OSPM for measurements at 12 streets stations using a newly developed and freely available evaluation tool. J. Civ. Environ. Eng. 1.

Ketzel, M., Wåhlin, P., Berkowicz, R., Palmgren, F., 2003. Particle and trace gas emission factors under urban driving conditions in Copenhagen based on street and roof-level observations. Atmos. Environ. 37, 2735–2749.

Ketzel, M., Wåhlin, P., Kristensson, A., Swietlicki, E., Berkowicz, R., Nielsen, O.J., Palmgren, F., 2004. Particle size distribution and particle mass measurements at urban, near city and rural level in the Copenhagen area and Southern Sweden. Atmos. Chem. Phys. 4 (1), 281–292.

Khan, J., Kakosimos, K., Raaschou-Nielsen, O., Brandt, J., Jensen, S.S., Ellermann, T., Ketzel, M., 2019. Development and performance evaluation of new AirGIS - a GIS based air pollution and human exposure modelling system. Atmos. Environ. 198, 102–121.

Klose, S., Birmili, W., Voigtländer, J., Tuch, T., Wehner, B., Wiedensohler, A., Ketzel, M., 2009. Particle number emissions of motor traffic derived from street canyon measurements in a Central European city. Atmos. Chem. Phys. Discuss. 3763–3809. **2009**.

Kukkonen, J., Karl, M., Keuken, M.P., van der Gon, H.A.C.D., Denby, B.R., Singh, V., Douros, J., Manders, A., Samaras, Z., Moussiopoulos, N., Jonkers, S., Aarnio, M., Karppinen, A., Kangas, L., Lutzenkirchen, S., Petaja, T., Vouitsis, I., Sokhi, R.S., 2016. Modelling the dispersion of particle numbers in five European cities. Geosci. Model Dev. (GMD) 9 (2), 451–478.

Kulmala, M., Kerminen, V.M., 2008. On the formation and growth of atmospheric nanoparticles. Atmos. Res. 90 (2–4), 132–150.

Kumar, P., Ketzel, M., Vardoulakis, S., Pirjola, L., Britter, R., 2011. Dynamics and dispersion modelling of nanoparticles from road traffic in the urban atmospheric environment-A review. J. Aerosol Sci. 42 (9), 580–603.

Kumar, P., Morawska, L., Birmili, W., Paasonen, P., Hu, M., Kulmala, M., Harrison, R.M., Norford, L., Britter, R., 2014. Ultrafine particles in cities. Environ. Int. 66, 1–10.

Kumar, P., Robins, A., Vardoulakis, S., Britter, R., 2010. A review of the characteristics of nanoparticles in the urban atmosphere and the prospects for developing regulatory controls. Atmos. Environ. 44 (39), 5035–5052.

Manders, A.M.M., Builtjes, P.J.H., Curier, L., van der Gon, H.A.C.D., Hendriks, C., Jonkers, S., Kranenburg, R., Kuenen, J.J.P., Segers, A.J., Timmermans, R.M.A., Visschedijk, A.J.H., Kruit, R.J.W., van Pul, W.A.J., Sauter, F.J., van der Swaluw, E., Swart, D.P.J., Douros, J., Eskes, H., van Meijgaard, E., van Ulft, B., van Velthoven, P., Banzhaf, S., Mues, A.C., Stern, R., Fu, G.L., Lu, S., Heemink, A., van Velzen, N., Schaap, M., 2017. Curriculum vitae of the LOTOS-EUROS (v2.0) chemistry transport model. Geosci. Model Dev. (GMD) 10 (11), 4145–4173.

Mertes, S., Schroder, F., Wiedensohler, A., 1995. The particle-detection efficiency curve of the Tsi-3010 Cpc as a function of the temperature difference between saturator and condenser. Aerosol. Sci. Technol. 23 (2), 257–261.

Nikolova, I., Janssen, S., Vos, P., Berghmans, P., 2014. Modelling the mixing of size resolved traffic induced and background ultrafine particles from an urban street canyon to adjacent backyards. Aerosol Air Qual. Res. 14 (1), 145–155.

Oberdörster, G., Finkelstein, J.N., Johnston, C., Gelein, R., Cox, C., Baggs, R., Elder, A.C., 2000. Acute Pulmonary Effects of Ultrafine Particles in Rats and Mice. Research Report. Health Effects Institute, pp. 5–74, 96.

Ohlwein, S., Kappeler, R., Joss, M.K., Kunzli, N., Hoffmann, B., 2019. Health effects of ultrafine particles: a systematic literature review update of epidemiological evidence. Int. J. Publ. Health 64 (4), 547–559.

Oliveira, J.R.D.E., Base, L.H., de Abreu, L.C., Ferreira, C., Ferreira, C., Morawska, L., 2019. Ultrafine particles and children's health: literature review. Paediatr. Respir. Rev. 32, 73–81.

Peters, A., Wichmann, H.E., 2001. Epidemiological evidence on the health effects of ultrafine particles. Epidemiology 12 (4), 544.

Plejdrup, M.S., Nielsen, O.-K., Gyldenkærne, S., Bruun, H.G., 2018. Spatial High-Resolution Distribution of Emissions to Air – SPREAD 2.0. Aarhus University, DCE – Danish Centre for Environment and Energy, p. 186. Scientific Report from DCE – Danish Centre for Environment and Energy No. 131. http://dce2.au.dk/pub/TR131.pdf.

Pohjola, M., Pirjola, L., Kukkonen, J., Kulmala, M., 2003. Modelling of the influence of aerosol processes for the dispersion of vehicular exhaust plumes in street environment. Atmos. Environ. 37 (3), 339–351.

Poulsen, A.H., Sorensen, M., Andersen, Z.J., Ketzel, M., Raaschou-Nielsen, O., 2016. Air pollution from traffic and risk for brain tumors: a nationwide study in Denmark. Canc. Causes Contr. 27 (4), 473–480.

R core team, 2021. R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria. URL: https://www.R-project.org/.

Roldin, P., Ehn, M., Kurten, T., Olenius, T., Rissanen, M.P., Sarnela, N., Elm, J., Rantala, P., Hao, L.Q., Hyttinen, N., Heikkinen, L., Worsnop, D.R., Pichelstorfer, L., Xavier, C., Clusius, P., Ostrom, E., Petaja, T., Kulmala, M., Vehkamaki, H., Virtanen, A., Riipinen, I., Boy, M., 2019. The role of highly oxygenated organic molecules in the Boreal aerosol-cloud-climate system. Nat. Commun. 10.

Roldin, P., Eriksson, A.C., Nordin, E.Z., Hermansson, E., Mogensen, D., Rusanen, A., Boy, M., Swietlicki, E., Svenningsson, B., Zelenyuk, A., Pagels, J., 2014. Modelling non-equilibrium secondary organic aerosol formation and evaporation with the aerosol dynamics, gas- and particle-phase chemistry kinetic multilayer model ADCHAM. Atmos. Chem. Phys. 14 (15), 7953–7993.

Roswall, N., Raaschou-Nielsen, O., Ketzel, M., Overvad, K., Halkjaer, J., Sorensen, M., 2017. Modeled traffic noise at the residence and colorectal cancer incidence: a cohort study. Canc. Causes Contr. 28 (7), 745–753.

Raaschou-Nielsen, O., Andersen, Z.J., Hvidberg, M., Jensen, S.S., Ketzel, M., Sørensen, M., Hansen, J., Loft, S., Overvad, K., Tjønneland, A., 2011. Air pollution from traffic and cancer incidence: a Danish cohort study. Environ. Health 10 (67). https://doi.org/10.1186/1476-069X-10-67.

Stone, V., Miller, M.R., Clift, M.J.D., Elder, A., Mills, N.L., Moller, P., Schins, R.P.F., Vogel, U., Kreyling, W.G., Jensen, K.A., Kuhlbusch, T.A.J., Schwarze, P.E., Hoet, P., Pietroiusti, A., De Vizcaya-Ruiz, A., Baeza-Squiban, A., Teixeira, J.P., Tran, C.L., Cassee, F.R., 2017. Nanomaterials versus ambient ultrafine particles: an opportunity to exchange toxicology knowledge. Environ. Health Perspect. 125 (10).

Sørensen, M., Hoffmann, B., Hvidberg, M., Ketzel, M., Jensen, S.S., Andersen, Z.J., Tjønneland, A., Overvad, K., Raaschou-Nielsen, O., 2012. Long-term exposure to traffic-related air pollution associated with blood pressure and self-reported hypertension in a Danish cohort. Environ. Health Perspect. 120 (3), 418–424.

Thacher, J.D., Roswall, N., Damm, P., Hvidtfeldt, U.A., Poulsen, A.H., Raaschou-Nielsen, O., Ketzel, M., Jensen, S.S., Frohn, L.M., Valencia, V.H., Munzel, T., Sorensen, M., 2021. Transportation noise and gestational diabetes mellitus: a nationwide cohort study from Denmark. Int. J. Hyg Environ. Health 231, 9.

Tørseth, K., Aas, W., Breivik, K., Fjæraa, A.M., Fiebig, M., Hjellbrekke, A.G., Lund Myhre, C., Solberg, S., Yttri, K.E., 2012. Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972-2009. Atmos. Chem. Phys. 12 (12), 5447–5481.

Vignati, E., Berkowicz, R., Palmgren, F., Lyck, E., Hummelshoj, P., 1999. Transformation of size distributions of emitted particles in streets. Sci. Total Environ. 235 (1–3), 37–49.

Vignati, E., Wilson, J., Stier, P., 2004. M7: an efficient size-resolved aerosol microphysics module for large-scale aerosol transport models. J. Geophys. Res.-Atmospheres 109 (D22).

von Bismarck-Osten, C., Birmili, W., Ketzel, M., Massling, A., Petaja, T., Weber, S., 2013. Characterization of parameters influencing the spatio-temporal variability of urban particle number size distributions in four European cities. Atmos. Environ. 77, 415–429.

von Bismarck-Osten, C., Birmili, W., Ketzel, M., Weber, S., 2015. Statistical modelling of aerosol particle number size distributions in urban and rural environments - a multi-site study. Urban Clim. 11, 51–66.

Vouitsis, I., Ntziachristos, L., Samaras, C., Samaras, Z., 2017. Particulate mass and number emission factors for road vehicles based on literature data and relevant gap filling methods. Atmos. Environ. 168, 75–89.

Wang, F., Ketzel, M., Ellermann, T., Wahlin, P., Jensen, S.S., Fang, D., Massling, A., 2010a. Particle number, particle mass and NOx emission factors at a highway and an urban street in Copenhagen. Atmos. Chem. Phys. 10 (6), 2745–2764.

Wang, F., Roldin, P., Massling, A., Kristensson, A., Swietlicki, E., Fang, D., Ketzel, M., 2010b. Aerosol dynamics in the Copenhagen urban plume during regional transport. Atmos. Chem. Phys. Discuss. 8553–8594. **2010**.

Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjaraa, A.M., Asmi, E., Sellegri, K., Depuy, R., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J.A., Swietlicki, E., Williams, P., Roldin, P., Quincey, P., Huglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Gruning, C., Faloon, K., Beddows, D., Harrison, R.M., Monahan, C., Jennings, S.G., O'Dowd, C.D., Marinoni, A., Horn, H. G., Keck, L., Jiang, J., Scheckman, J., McMurry, P.H., Deng, Z., Zhao, C.S., Moerman, M., Henzing, B., de Leeuw, G., Loschau, G., Bastian, S., 2012. Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions. Atmos. Meas. Tech. 5 (3), 657–685.

WMO-GAW. World calibration center for aerosol physics (WCCAP). http://www.wmo-gaw-wcc-aerosol-physics.org/.

Zhang, K.M., Wexler, A.S., Zhu, Y.F., Hinds, W.C., Sioutas, C., 2004. Evolution of particle number distribution near roadways. Part II: the 'Road-to-Ambient' process. Atmos. Environ. 38 (38), 6655–6665.