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# Modelling the dispersion of particle numbers in five European cities

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## Abstract

We present an overview of the modelling of particle number concentrations (PNC's) in five major European cities, namely Helsinki, Oslo, London, Rotterdam and Athens, in 2008. Novel emission inventories of particle numbers have been compiled both on urban and European scales. We use atmospheric dispersion modelling for PNC's in the five target cities and on a European scale, and evaluate the predicted results against available measured concentrations. The concentrations of PN in the selected cities were mostly influenced by the emissions originated from local vehicular traffic; however, in some of the cities, also harbour and airport activities were significant. The highest values of the predicted PNC's were higher in the megacities, London and Athens, and also in Rotterdam, whereas these were lower in Helsinki and Oslo. It was numerically evaluated that the influence of coagulation and dry deposition on the predicted PNC's was substantial for urban background in Oslo. The predicted and measured annual average PNC's in four cities agreed within approximately $\leq 36\,\%$ (measured as fractional biases), except for one traffic station in London. The indexes of agreement (IA) for the comparisons of hourly measured and predicted time-series in Oslo and Helsinki ranged from 0.75 to 0.79.

## 1 Introduction

Airborne particulate matter (PM) affects human health and climate (e.g. Smith et al., 2009). While a large base of scientific information exists on particle mass, especially for $PM_{10}$ and $PM_{2.5}$, there are substantially less studies on particle number (PN) and in particular on modelling dispersion of PN's in urban areas (e.g., Kumar et al., 2013). This may be attributed to (i) scarcity of reliable information on emissions, (ii) the greater complexity of physical and chemical atmospheric processes and (iii) lack of monitoring data of PN. The majority of urban particles – in terms of number concentration – are ultrafine particles (UFP), i.e., particles with diameter ($D_p$) < 100 nm, originating mainly





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from traffic-related emission (e.g., Morawska et al., 1998). The rapid transformation processes of PN after emissions in ambient air, such as condensation/evaporation, coagulation, dry deposition and dilution pose challenges for dispersion modelling, especially on an urban scale (e.g. Ketzel et al., 2004; Kittelson et al., 2004; Kumar et al., 2011, von Bismarck-Osten et al., 2013). In addition, PN measurement techniques are also more complex and resource-consuming, compared with the measurements of particulate mass fractions.

Although attention on the health effects of particulate matter has been focused on particle mass fractions, a number of studies are indicating that UFP's may have specific health effects. UFP's are poorly filtered in the human respiratory tract after inhalation, and such particles are able to penetrate the epithelial cells of the lungs and accumulate in lymph nodes (Nel et al., 2006). Epidemiological and toxicological studies show a strong correlation between exposure to ultrafine particles and various health endpoints, such as cardiovascular hospital admission (short-term exposure), mortality (long-term exposure) and neurological effects (Oberdörster et al., 2004; Delfino et al., 2005; Atkinson et al., 2010; Franck et al., 2011; Daher et al., 2013; Loane et al., 2013).

There is a severe lack of representative sets of urban measurements of particle number concentrations (PNC's) that could be used in epidemiological studies, when compared to particle mass. Similarly, the scientific literature is scarce on predicting the dispersion of PN in urban environments. However, it is not currently feasible to set up continuous measurements of nanoparticles at numerous urban locations. It is therefore necessary to develop and evaluate dispersion modelling systems capable of reliably predicting PNC's.

Combustion is a direct source of UFP's, and secondary particle formation may occur via atmospheric reactions and condensation of semi-volatile components produced in photochemical reactions (Kulmala et al., 2013, 2014; Kumar et al., 2014). Combustion of carbon-based fuels for power generation, heating and transport are important sources for PN emissions (Shi et al., 2001; Koppmann et al., 2005; Obaidullah et al., 2005; Kittelson et al., 2006; Maricq, 2007; Buzea et al., 2007; Abbasi et al., 2013;





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Kumar at al., 2013; Pant and Harrison, 2013). In most European cities, road traffic emissions of PN are the most important source for exposure of the population, due to the near-ground emissions and the vicinity of road traffic to populated areas.

The importance of aerosol processes has been analyzed via aerosol process time scales by Zhang and Wexler (2004) and Ketzel and Berkowicz (2004), and with aerosol dynamics model simulations by Jacobson and Seinfeld (2004). Pohjola et al. (2003) simulated the transformation and dilution of particulate matter on distance scale of less than 100 m from a road in an urban area. As expected, dilution was found to be the most important process affecting the PNC's; however, condensation of an insoluble organic vapour was also found to be important, if its concentration exceeds a certain threshold value. Ketzel and Berkowicz (2004) evaluated that the influence of dry deposition would be irrelevant on an urban time scale. Kerminen et al. (2007) evaluated that coagulation, condensation and evaporation could be important in conditions, where dilution with cleaner background air is restricted.

Small-scale combustion may also be a prominent source of PNC's in winter (Glasius et al., 2008). Elevated levels of PN have also been found in specific areas, such as, near harbors, refineries and in particular near airports (González and Rodríguez, 2013; Westerdahl et al., 2008; Zhu et al., 2011; Keuken et al., 2012; Hsu et al., 2014). Whereas most of the state-of-the-art chemical transport models include treatments for aerosol size distributions and microphysics (Kukkonen et al., 2012), such treatments are substantially less commonly included in urban scale models. There are currently very few models, which are especially designed to predict particle number concentrations by taking into account particle dynamics. Kumar et al. (2012) presented a review on the importance of aerosol transformation processes at various urban scales and environments.

A first European size-resolved anthropogenic PN emission inventory was compiled in the framework of the EU-funded EUCAARI project (Denier van der Gon et al., 2010). Consolidated emission factor data bases (e.g., COPERT, PARTICULATES and TRANSPHORM) have recently become available to establish PN emission inventories





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in Europe; these have been reviewed by Kumar et al. (2014). According to the inventory by Paasonen et al. (2013), for the 28 EU countries in 2010, road transport contributed over 60 % of the total PN emissions, non-road transport (including partly also shipping) 19 % and domestic combustion 13 %.

The first stage between the point of emission (vehicle tailpipe) and the kerbside is characterized by strong turbulence generated by the moving vehicles. According to Zhang and Wexler (2004), the initial stages of dilution within a few first seconds would be accompanied with nucleation. On-road measurements by Rönkkö et al. (2007) demonstrated that the nucleation mode was already present after 0.7 s residence time in the atmosphere. However, the modelling of nucleation will require detailed information about the environmental conditions very near the tailpipe (e.g., temperature gradient, and chemical composition and concentrations of volatile nucleating vapours). Nucleation mode particles grow rapidly by condensation of high-molecular weight low-volatile hydrocarbons from the unburned lubrication oil and sulphur compounds (Kittelson et al., 2006).

In the second stage between the street and a few hundred meters away from the street, atmospheric turbulence, induced by wind and atmospheric instability, is the main cause for dilution of particle concentrations. In this stage, condensation/evaporation and dilution become the major mechanisms in altering the particle size distribution, while coagulation and deposition play minor roles (Zhang et al., 2004). In the third stage, between street canyon/street neighborhood and the urban background, the number size distribution is altered by multiple processes, such as dilution with cleaner air, entrainment of polluted air, condensation of vapors, oxidative ageing, and coagulation of particles (e.g., Wehner et al., 2002).

Asmi et al. (2011) examined aerosol number size distribution data from 24 European field monitoring sites in 2008 and 2009. The data was collected from the stations at the EUSAAR (European Supersites for Atmospheric Aerosol Research) and GUAN networks (German Ultrafine Aerosol Network), and represented mainly regional background or remote locations. They categorized the aerosol to several types: central Eu-





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ropean aerosol, Nordic aerosol, mountain sites and southern and western European regions, and analyzed the seasonal characteristics and patterns of the various size modes.

Pohjola et al. (2007) conducted a field measurement campaign near a major road in an urban area in Helsinki in February 2003. Measured PNC data at various distances from the road was compared with dispersion and aerosol process model predictions. A similar measurement campaign was conducted downwind of a motorway in Rotterdam (Keuken et al., 2012). Size-resolved PNC measurements were compared with dispersion modelling and an aerosol process model (Karl et al., 2011). Both these studies concluded that dilution was shown to be the most important process.

Gidhagen et al. (2005) implemented a three-dimensional dispersion model in Stockholm and presented the spatial distribution of number concentrations over the whole city. Typical number concentrations in the urban background of Stockholm were 10 000 $cm^{-3}$, and approximately three times higher close to a major highway and seven times higher within a densely trafficked street canyon. Coagulation was found to contribute to losses of PNC's of only a few percent as compared to inert particles, while including dry deposition resulted in PNC losses of up to 25 % in certain locations. However, removal of PN's due to coagulation and deposition was more significant during peak episodes.

This study is part of the European Union funded research project TRANSPHORM (Transport related Air Pollution and Health impacts – Integrated Methodologies for Assessing Particulate Matter). This project was one of the very few international projects, where dispersion models have been developed and applied to predict spatially and temporally resolved concentrations of PN for exposure and health applications (ww.transphorm.eu). The cities Helsinki, Oslo, Rotterdam, London and Athens were involved to test the methodologies developed within the TRANSPHORM project at an urban scale. These cities were selected in order to include at least one major urban agglomeration from the following regions: (i) the Nordic countries (Helsinki and





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Oslo), (ii) the central and north-western Europe (Rotterdam and London) and (iii) the Mediterranean region (Athens).

For PN, hardly any health studies are available, and currently only a concentration-response function based on expert judgment is available; more specifically for a 0.3 % increase in all-cause mortality per $10^3$ particles per $cm^3$ (Hoek et al., 2010). Von Klot et al. (2005) underlined similar effects for hospital re-admissions of a susceptible population, in cases, for which the aerosol number increased $10^3$ particles per $cm^3$ or aerosol mass by 10 $\mu g\,m^{-3}$. However, in view of the potential health effects for exposure to PNC's, there is a need to combine epidemiological data and PNC's with a high spatial resolution.

The aim of this article is to present an overview of the modelling of PNC's on an urban scale in five major European cities, presented in Fig. 1: Helsinki, Oslo, Rotterdam, London and Athens. The target cities represent megacities, such as London (population of approximately 8.3 million) and Athens (we address here Greater Athens, 3.5 million), and other major cities, such as Helsinki Metropolitan Area, Oslo and Rotterdam (populations of 1.0, 0.6 and 0.6 million, respectively). For simplicity, we refer to Helsinki Metropolitan Area simply as "Helsinki" in the following. The primary year used in the computations is 2008. The modelling of PNC's for these cities has been presented in the present article for the first time. The previous literature also does not contain any compilations of PNC modelling for several cities.

We address emission inventories and emission modelling of PN, dispersion modelling of PNC's, numerical results on the annual average spatial distributions in the target cities and evaluation of the predicted results against measured PNC's. The main scientific goals were (i) to undertake a comparative analysis of the capability of models to predict PNC's in several European cities, (ii) to examine spatial characteristics of PN in the selected cities, (iii) in at least one of these cities, quantitatively evaluate the contributions of various source categories on the concentrations, and (iv) to highlight areas of improvements in modelling PN for health based studies.





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## 2 Modelling methods

In this section the computational methods are presented, which were used for the evaluation of PNC's in the five target cities. We address both the methods for the evaluation of emissions, and the atmospheric dispersion modelling systems. For practical reasons, it was not possible to completely harmonize the computations, by using only one modelling system for all the cities. All of the urban emission and dispersion modelling systems were therefore locally or nationally developed ones; these were different for each city. However, the regional background concentrations for all the urban scale modelling systems were computed with the same model, the LOTOS-EUROS chemical transport model (Schaap et al., 2008). We have therefore also briefly discussed a new European-scale emission inventory used as input for the above mentioned regional scale chemical transport model.

### 2.1 Overview of the PNC computations in the target cities

For readability, selected summary information has been presented in Table 1 on the urban scale computations. The more detailed information will be presented in the following sections.

The TRANSPHORM project emission database was used on an urban scale in three of the target cities. Two urban modelling systems applied a meteorological preprocessing model, two others other meteorological models, and one modelling system applied directly measured data. All the models included the emissions from vehicular traffic, and some of the models included also the emissions from major and/or small-scale stationary sources and other sources. The shipping emissions were explicitly included in the computations of Oslo and Athens, and the importance of primary shipping emissions was separately evaluated for Helsinki (Soares et al., 2014). However, in case of Rotterdam and London, the local scale shipping emissions were not taken into account.





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Regional background concentrations were derived from the LOTOS-EUROS model computations for all target cities; however, the detailed method on how these values were used as input for the urban scale models varied from city to city. The aerosol transformation processes were taken into account in the LOTOS-EUROS computations. Measured PNC data was available in four of the cities, in three of these for a complete year; however, only at one or two measurement stations for each city.

## 2.2 Emission inventories

We describe in this section both a new European scale emission inventory and the urban emission inventories in the five target cities.

### 2.2.1 European-scale emission inventory

A new emission inventory was compiled for the EU-wide transport activities, supplemented by non-transport activities. The baseline emission data contains the following substances: $NO_x$, $SO_2$, non-methane volatile organic compounds (NMVOC), $CH_4$, $NH_3$, CO, $PM_{10}$, $PM_{2.5}$, EC (elemental carbon), B[a]P (benzo[a]pyrene) and PN (Denier van der Gon et al., 2014). The PN inventory includes particles in the 10–300 nm size range.

The emission data can be calculated for the individual countries; the official UN ISO3 Country Codes were used. We have used three groups of countries. The EU15+ group is defined to include EU15, Norway and Switzerland. The EU12+ group contains the New Member States, Malta and European Non-EU countries; the latter refers to the other European countries in the United Nations Economic Commission for Europe domain. The EU27+ group consists of EU15+ and EU12+. Emissions from international shipping have been estimated for the various European sea regions.

The first European particle number emission inventory was made in the EU FP6 project EUCAARI (Denier van der Gon et al., 2010a; Kulmala et al., 2011). This inventory was used as a starting point for the present study. For the different transport





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modes (road, rail, air and maritime navigation), a new bottom-up PN emission estimate was made, including also technologies or activities in the future years, 2020 and 2030.

The PN emission inventory includes only anthropogenic sources; the emissions from mainly natural sources such as, e.g., wild-land fires, windblown dust and sea salt are not included. The inventory also does not include vegetation related emissions (e.g., Guenther et al., 1995), or the formation of PNC's from biogenic VOC's (e.g., Paasonen et al., 2012).

To approximate the future year emissions for the non-transport sectors, scaling factors were used based on the IIASA primes baseline scenario for $PM_{2.5}$ (http://gains.iiasa.ac.at/). We have assumed that the PN emissions for the non-transport sectors will follow the corresponding trend in $PM_{2.5}$ emissions.

### 2.2.2 Urban-scale emission inventories in the target cities

#### Emission inventory for Helsinki

The emission inventory included exhaust emissions from vehicular traffic for the network of roads and streets in the Helsinki Metropolitan Area (HMA). The traffic volumes and average travel speeds of each traffic link were computed using the EMME/2 transportation planning system (INRO, 1994). Traffic volume data in 2008 was used as input for the estimation of annual average road traffic emissions in the HMA. The final emission inventory consisted of average hourly emissions for each line source over the year, separately for weekdays, Saturdays and Sundays.

The emission factors for vehicular traffic determined by Gidhagen et al. (2005) in Stockholm have been used. The emission factors corresponding to Stockholm were used, as these were estimated to optimally correspond to the climatic and traffic conditions in Helsinki. These values are $2.70 \times 10^{15}$ and $1.8 \times 10^{14}$ particles vehicle$^{-1}$ km$^{-1}$ for heavy and light-duty vehicles, respectively. These values were determined for driving speeds less than 70 km h$^{-1}$; however, we have applied these values for all urban roads included in the computations.





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In addition to the computations for 2008, we computed the PNC's at the roadside traffic station at Ring Road 1, Malmi (called simply as "Ring Road 1" in the following) in 2012, for model evaluation purposes. For the hourly computations in 2012, the 2008 traffic volume data was scaled using the ratio of the total vehicular mileages ($km\,a^{-1}$) in the HMA in 2008 and 2012. These mileage values were obtained from the national traffic emissions data archive LIPASTO (http://lipasto.vtt.fi/indexe.htm).

Emissions from stationary sources were not included. However, major stationary sources have previously been shown to have negligible effect on the $PM_{2.5}$ concentrations near the ground level in Helsinki (Kauhaniemi et al., 2008); the same was assumed to be approximately valid also for PNC's. Emissions from small-scale combustion were not taken into account, as their spatial distribution was not known with sufficient accuracy. The contribution of small-scale combustion to the total $PM_{2.5}$ emissions in Helsinki Metropolitan Area has been estimated to be 15 % (Niemi et al., 2009).

The importance of the shipping emissions can be evaluated based on Soares et al. (2014). They showed using the STEAM2 shipping emission modelling (Jalkanen et al., 2012) that the contribution of primary shipping emissions of $PM_{2.5}$ to the concentrations of $PM_{2.5}$ are only 3 % on the average in the Helsinki Metropolitan Area (HMA). However, this contribution can be higher than 20 % in the vicinity of the harbours (within a distance of approximately one kilometer).

## Emission inventory for Oslo

Emission factors for traffic exhaust (measured at an ambient temperature of +33 °C) were extracted from the emission database of the TRANSPHORM project (Vouitsis et al., 2014) Emission factors for PN in Oslo and in other studies (Klose et al., 2009; Olivares et al., 2007) have been found to have a significant dependence on ambient air temperature. A dependence of $-3\,\%\,K^{-1}$ has been applied to the Oslo traffic emissions, leading to significantly higher emission factors in the cold winter period (approximately double) than those provided in the emissions database.





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Shipping emissions were based on the STEAM2 emission model (Jalkanen et al., 2012). Emissions for PN were based on the $CO_2$ emissions, converted firstly back to fuel consumption, and then PN emissions were calculated using an emission factor of $1 \times 10^{16}$ particles (kg fuel)$^{-1}$, recommended by Petzold et al. (2010). Shipping emissions were evaluated in a domain of 29 km × 18 km and thus only included shipping in the Oslo fjord area.

Domestic heating emissions of PN, due mostly to wood burning, were calculated based on a previously compiled $PM_{2.5}$ inventory. A conversion factor of $4 \times 10^{14}$ particles $(gPM_{2.5})^{-1}$ emitted was used to convert $PM_{2.5}$ emissions to PN emissions, based on the data presented in Hedberg et al. (2002). Other emissions concerning combustion sources, i.e. agricultural, industrial and mobile sources use the existing $PM_{2.5}$ emissions inventory and convert to PN using a ratio similar to diesel truck emissions; a conversion factor of $3 \times 10^{15}$ particles $(gPM_{2.5})^{-1}$ was applied.

### Emission inventory for Rotterdam

Road traffic data and road characteristics were obtained from a national database (www.nsl-monitoring.nl). Road traffic data contains information about the number of vehicles, speed, congestion and fleet composition in-between traffic links for every major road and motorway in Rotterdam. The road characteristics refer to, e.g., the width and height of buildings along the road.

The following emission factors from COPERT IV (Gkatzoflias et al., 2012) and the TRANSPHORM database have been applied: (i) for motorway traffic, $10^{15}$ particles km$^{-1}$ veh$^{-1}$ for heavy and light duty vehicles, and $0.3 \times 10^{15}$ particles km$^{-1}$ veh$^{-1}$ for passenger cars, and (ii) for urban road traffic, $0.5 \times 10^{15}$ particles km$^{-1}$ veh$^{-1}$ for heavy and light duty vehicles and buses, and $0.3 \times 10^{15}$ particles km$^{-1}$ veh$^{-1}$ for passenger cars.





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## Emission inventory for London

The road traffic data for London have been obtained from London Atmospheric Emission Inventory (LAEI; GLA, 2010). Each road link was characterised by the amount of vehicles per day per vehicle category and mean speed. The traffic activity data were disaggregated by vehicle categories such as motorcycles, cars including taxis, buses, light goods vehicles (LGV) and heavy goods vehicles (HGV). The HGV's are further subdivided into articulated HGV's and rigid HGV's categories. The fleet compositions have been further subdivided as per fuel type, weight, engine size and emission standards.

The emission model in current version of the OSCAR system commonly uses the emission functions and factors based on COPERT IV (Gkatzoflias et al., 2012) and Department for Transport (DfT) emission data base. However, due to the unavailability of emissions in that database for PN's, emission factors from Jones and Harrison (2006) have been used in this study.

## Emission inventory for Athens

For Athens PN emissions included traffic, shipping and aviation. Emission factors for traffic exhausts were taken from the TRANSPHORM emission database (Vouitsis et al., 2014). Emissions from shipping and the major ports, and airport emissions were calculated on the basis of the operational action plan for air pollution management in Athens. This plan was developed for 2004, using activity and fuel consumption data (Samaras et al., 2012). The emission factor used for shipping was $10^{16}$ particles $(\mathrm{kg\,fuel})^{-1}$ according to Petzold et al. (2010), and for aviation $6 \times 10^{14}$ particles $(\mathrm{kg\,fuel})^{-1}$, assuming a fuel sulphur content of 1000 ppm (Lee et al., 2010).





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## 2.3 Dispersion and transformation modelling

First, we address the dispersion modelling on a continental scale, which provided the regional background concentrations for urban dispersion modelling. Second, we discuss the urban scale dispersion modelling systems used in the five target cities.

### 2.3.1 Chemical transport modelling on a European scale

The chemistry-transport model LOTOS-EUROS (Schaap et al., 2008) was used in this study to evaluate the regional background PNC's. Compared with other widely used chemical transport models in Europe (Kukkonen et al., 2012), the model is of intermediate complexity. The relevant processes have been parameterized in such a way that the computational demands are modest. The LOTOS-EUROS model has been included in several international model inter-comparison studies that have addressed the dispersion and transformation of ozone and particulate matter (e.g., Stern et al., 2008 and Solazzo et al., 2012). The model performance has in these model inter-comparisons been comparable with other European chemical transport models.

The M7 aerosol microphysics module (Vignati et al., 2004) was coupled to the LOTOS-EUROS model. This module accounts for nucleation and condensation of $H_2SO_4$, and coagulation of particles. The default nucleation scheme originally based on Vehkamäki (2002) was replaced by the activation type parameterization of Kulmala et al. (2006), which is better suitable for the boundary layer.

Formation of $H_2SO_4$ was based on the default gas-phase chemistry of LOTOS-EUROS, using emission inventories provided by the MACC project (TNO-MACC emission inventory; Pouliot et al., 2012) and the TRANSPHORM emission inventories. The PN emissions were converted to values that are compatible with the M7 module, using assumptions on the chemical composition of particulate matter. In addition, sea spray aerosol emissions were included, as well as dust emissions from road resuspension, agriculture and bare soils (Schaap et al., 2009).





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Two sets of simulations were made. (i) The first set was based on the meteorology of 2008, and was used for model evaluation. This set covered Europe on a 0.5 × 0.25 longitude–latitude grid, for a domain from 15° W to 35° E and from 35 to 70° N. The concentrations for particle number were assumed to be negligible at the boundaries of the domain. (ii) The second set of simulations was performed for the meteorology of 2005, with the emissions for the years 2005 and 2020, both of these covering Europe. Additional simulations were performed for each target city, on a finer 0.125 × 0.0625 longitude–latitude grid, for each city in a domain that covered an area of 3° × 1.5°, using the European-scale simulation for boundary conditions.

There are several processes that contribute to uncertainties in the model results. Nucleation mode particles contribute substantially to the total particle numbers. However, several parameterizations for nucleation processes are available, and it is not in all cases clear, which are the optimal ones. The uncertainties associated with the modelling of particle nucleation have mainly an impact on the number concentration of particles smaller than 100 nm (e.g., Fountoukis et al., 2012).

Some atmospheric species are not represented in the M7 module. For example, secondary aerosol formation from biogenic emissions (such as, isoprene and terpene) is not taken into account. Riipinen et al. (2011) investigated the role of condensable vapours on the growth of freshly nucleated particles until the cloud condensation nuclei size, and proposed a semi-empirical modelling approach. Secondary organic vapours can condense on existing particles, and thus contribute to their growth. This process increases the probability of such particles to reach the sizes that are cloud condensation nuclei (CCN) active, before getting scavenged by the background particle population. Secondary organic aerosol from biogenic origin therefore may substantially contribute to the PCN's.

The emissions of condensable gases from combustion processes are also not taken into account in the modelling; these could potentially contribute, e.g., in areas with substantial residential wood burning. In regions with intensive $NH_3$ emissions (e.g., from agriculture and animal husbandry), the impact of secondary inorganic aerosol





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may be significant on number and size distribution of particulate matter; this is not accounted for in the M7 module (Vignati et al., 2004).

The omission of biogenic secondary aerosol causes inaccuracies to the PM size distribution. The inaccuracies are the largest in case of the smallest particles. The modelled sum of the Aitken and accumulation mode particle number concentrations are therefore considered the most appropriate quantity to represent regional background PNC's in this study (compared with using the number concentration of the nucleation mode particles).

### 2.3.2 Urban scale dispersion modelling

For each modelling system, we address (i) the urban dispersion modelling system and its implementation, (ii) the evaluation of meteorological variables (used as input for the urban modelling), and (iii) the assessment of regional background concentrations.

#### Dispersion modelling for Helsinki

The urban scale dispersion of vehicular emissions was evaluated with the CAR-FMI model (Contaminants in the Air from a Road – Finnish Meteorological Institute; Kukkonen et al., 2001; Härkönen et al., 1996). The model computes an hourly time-series of the pollutant dispersion from the line source. The dispersion equation for the line source model is based on a semi-analytical solution of the Gaussian diffusion equation for a finite line source. The dispersion parameters are modelled as a function of the Monin–Obukhov length, the friction velocity and the mixing height. Traffic-originated turbulence is modelled with a semi-empirical treatment.

The receptor grid intervals range from 20 m in the vicinity of major roads to 500 m on the outskirts of the area. The concentration values were computed in 18 692 receptor points.

Input data needed by the dispersion model was evaluated using a meteorological pre-processing model (MPP-FMI) that has been adapted for an urban environ-





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ment (Karppinen et al., 2000c). The MPP-FMI model is based on the energy budget method of van Ulden and Holtslag (1985). The model utilises meteorological synoptic and sounding observations, and its output consists of estimates of the hourly time series of the relevant atmospheric turbulence parameters and the boundary layer height. The computation is based on a combination of the data from the stations at Helsinki–Vantaa airport and Helsinki-Kumpula (3 h synoptic weather observations), and Jokioinen (soundings).

The regional background concentrations for 2008 were based on the predictions of the LOTOS-EUROS model. We used the hourly concentration values predicted by the LOTOS-EUROS model at a grid square (7 km × 7 km) that corresponds to a regional background station for the Helsinki region (the station of Luukki).

The urban background concentrations of PN in 2012 were estimated to be equal to the measured hourly values at an urban background measurement site located in Kumpula in Helsinki. This station is part of the network of stations called "Station for Measuring Ecosystem – Atmosphere Relations", SMEAR-III (Järvi et al., 2009). This data contained PNC's in the particle size range from 3 to 950 nm.

### Dispersion and particle transformation modelling for Oslo

Calculations of concentrations were carried out using the EPISODE dispersion model (Slørdal et al., 2003), which is part of the integrated air quality management tool AirQUIS (Slørdal et al., 2007). The EPISODE model consists of a gridded Eulerian model coupled with a Gaussian line source model for modelling the local contribution at receptor points near roads. The Eulerian grid model uses a 1 km × 1 km grid covering Oslo. There are 13 vertical layers in the model, up to the height of 4000 m, with the lowest layer being 10 m thick. Emissions from traffic sources are placed in the lowest layer, whilst emissions from domestic heating, industry and shipping are placed in the layers between 10 and 35 m.

Receptor points within 500 m of a road include line source calculations, using the Gaussian line source model in EPISODE, otherwise only the Eulerian model con-





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tributes. The model coupling leads to a double counting of the emissions near roads, which has been estimated to contribute a maximum increase of 5–20 % to the model concentrations at receptor points near roads. The receptor points are placed at monitoring sites, and at aggregated home addresses, at the centre of population mass within a 100 m × 100 m grid.

The air pollution originated in vehicular traffic tunnels has been modelled assuming that there has been no deposition of particles within the tunnels. The tunnel exits are therefore treated simply as exit points of polluted air.

Meteorology is generated in the model using the diagnostic wind field model MCWIND. The MCWIND model uses meteorological measurements and interpolates these in space, adjusting for topography and atmospheric stability. Measurements from two sites are used (Valle Hovin and Blindern); both sites are centrally located in Oslo. Data required by the dispersion modelling are atmospheric stability, wind speed and wind direction.

Hourly regional background concentrations were derived using predictions from the LOTOS-EUROS model at a number of grid squares surrounding Oslo. The hourly median concentration from these grid squares was extracted for this purpose. These values were further adjusted, based on a comparison of the predicted and observed annual mean PNC measurements at Birkenes (located about 300 km south of Oslo). This procedure resulted in a rescaling of all LOTOS-EUROS predictions by a factor of 0.75.

In Oslo, a parametrization was applied to account for deposition and coagulation processes. This was only applied in the gridded model calculations, but not in the sub-grid Gaussian modelling. This parametrization is based on calculations using the MAFOR aerosol process model for road traffic emissions (Keuken et al., 2012). First, MAFOR calculations were carried out using the complete aerosol process model description and then, for simplicity, the emissions and calculations were binned into three particle size classes. Based on these computations, deposition and coagulation rates in these three size classes were derived.





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The change of the PNC in each size bin caused by coagulation was parameterized in the following simplified form:

$$\left.\frac{\mathrm{dPNC}_i}{\mathrm{d}t}\right|_{\mathrm{coag}} = -\mathrm{PNC}_i^2 \overline{K_{\mathrm{c},i}}, \quad (1)$$

where the subscripts $i$ and coag refer to the particle size class and coagulation, respectively, and $K_{\mathrm{c},i}$ is the coagulation rate derived using the MAFOR model. Dry deposition is described as

$$\left.\frac{\mathrm{dPNC}_i}{\mathrm{d}t}\right|_{\mathrm{depo}} = -\mathrm{PNC}_i \frac{\overline{v_{\mathrm{d},i}}}{H_{\mathrm{grid}}}, \quad (2)$$

where $v_{\mathrm{d},i}$ is the dry deposition rate for the $i$th size class and $H_{\mathrm{grid}}$ is the depth of the lowest model grid layer.

## Dispersion modelling for Rotterdam

In Rotterdam, the contribution of traffic to air quality near inner-urban roads was modelled with a street-canyon dispersion model (Eerens et al., 1993; Vardoulakis et al., 2003), and near motorways up to a distance of 500 m with a line source dispersion model (Wesseling et al., 2003; Beelen et al., 2010; Keuken et al., 2012).

The street canyon dispersion model is based on the results of wind tunnel experiments at different road types, including street canyons. The ratio of the height of the buildings and the width of the street is used to classify the type of street canyon. A source–receptor relationship has been specified as a function of the distance to the street axis for five different road types. All streets in Rotterdam have been categorized in accordance to the model classification. The contribution of traffic emissions to annual average concentrations has been assumed to depend on the emission rate, the annual average wind speed and the road type.





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The line source model is a Gaussian plume model, which assumes that the contribution to ambient air concentrations downwind of the motorway is proportional to the emission rate and inversely proportional to the wind speed (Wesseling et al., 2003; Beelen et al., 2010). The model takes into account vehicle-induced turbulence, the upwind roughness of the terrain, the presence of noise screens near the motorway and atmospheric stability. The meteorological data was retrieved from measurements by the National Meteorological Institute (www.knmi.nl) at the Airport of Rotterdam.

The urban background of PNC's was estimated based on the LOTOS-EUROS model, at a grid square that corresponds to Rotterdam. The model output has a spatial resolution of 10 m × 10 m up to a distance of 300 m from the streets, or alternatively at the housing façade along street canyons, and up to a distance of 500 m near motorways.

### Dispersion modelling for London

The OSCAR air quality assessment system (Singh et al., 2013; Sokhi et al., 2008) has been used to estimate traffic related PNC's across London. The models within the OSCAR system consist of an emission model, meteorological pre-processing model and a line source Gaussian dispersion model. The roadside dispersion model within OSCAR system is the CAR-FMI model. The hourly concentrations were predicted at the receptor points placed at varying distances of 10, 40 and 90 m near both sides of the roads, and 100 m apart in the outskirts.

A range of hourly meteorological parameters are needed, including wind speed, solar radiation, friction velocity and Monin–Obukhov length. These are provided by the dedicated OSCAR meteorological pre-processor GAMMA met, described by Bualert (2002). The meteorological model employs meteorological data, such as solar radiation, roughness length and heat flux, to estimate atmospheric stability parameters, including the Monin–Obukhov lengths and mixing heights. Data from the meteorological station at Heathrow was used as input for the model. The effects of land use characteristics on parameters such as surface roughness, Bowen ratio, Albedo and anthropogenic heat flux are taken into account. The meteorological pre-processor needs six





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input parameters: time, wind speed, wind direction, ambient temperature, cloud cover and global radiation.

Both the regional and urban background levels of PNC were evaluated based on the LOTOS-EUROS simulations. The evaluated regional background is based on the values by the LOTOS-EUROS model both within and around the city. The magnitude of these evaluated values for the urban background were checked, by comparing these with the measured PNC values at the station of North Kensington, which is an urban background site.

### Dispersion modelling for Athens

The modelling system consists of two models: (i) the meteorological model MEMO (Moussiopoulos et al., 1993), and (ii) the chemical transport model MARS-aero (Moussiopoulos et al., 1995, 2012). The MEMO model is a three-dimensional Eulerian non-hydrostatic prognostic model. The MARS-aero model can be used to simulate the transport and transformation of gaseous pollutants and atmospheric aerosols in the lower troposphere. The nesting capability of the modelling system allows for a finer grid simulation to be nested inside a coarser grid simulation.

Meteorological data was generated using the MEMO model. Measured data needed to apply the model were based on upper air soundings for selected meteorological variables (wind speed and direction, temperature), from the Athens International Airport. For evaluating the annual concentration means, a weighting scheme was applied on the daily concentration fields, based on a classification of local meteorological patterns (Sfetsos et al., 2005; Shahgedanova, 1998). The models were applied in a computational domain of 50 km × 50 km, on a spatial resolution of 500 m.

Both the regional background PNC's and the concentrations of other relevant species are needed as boundary conditions for the MARS-aero calculations. A spatially uniform annually average regional PNC background of 1800 particles $cm^{-3}$ was used for the boundary conditions, based on Kalivitis et al. (2008). The regional background values





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of all other relevant species were extracted from the LOTOS-EUROS computations, at the grid squares surrounding the city.

## 3 Results and discussion

### 3.1 Emissions

#### 3.1.1 Emissions in Europe and their associated uncertainties

Total anthropogenic PN emissions in UNECE Europe were estimated using a bottom-up methodology (Denier van der Gon et al., 2014). These are presented in Fig. 2a and b, classified according to both source sector and country group. The transport sectors (i.e., road and non-road transport) contributed approximately 60 % to the total land-based PN emissions in UNECE-Europe in 2005 (Fig. 2a).

The PN emissions are projected to decrease in 2020 and 2030 to less than a half of their value in 2005 (Fig. 2b). International shipping was a dominating source in 2005, but its contribution is expected to substantially decline from 2005 to 2020 and 2030, mainly due to the introduction of low sulphur fuels. The contribution of shipping is more dominant in the current inventory, compared with the first European PN emission inventory made in the EU-funded project EUCAARI (Denier van der Gon et al., 2010a; Kulmala et al., 2011). Another remarkable change compared with the previous inventory is that in the new inventory, aviation is a substantially stronger source of UFP's than previously assumed. Most of these shipping and aviation particulate emissions are not solid, but semi-volatile particles, and may therefore have escaped attention in previous emission factor measurements.

The PN emission inventory includes in principle all particulate sizes. The PN emissions in two size fractions have been presented in Fig. 2a. The ultrafine particle fraction (UFP) is defined as particles smaller in diameter than 100 nm. As expected, the differ-





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ence between the total PN emissions and the UFP emissions is relatively small, as the PN emissions are dominated by the smaller size fractions.

The corresponding emissions solely for the road transport sector have been presented in Fig. 2b. The PN emissions of road transport are projected to significantly decrease in time (Fig. 3b). The PN emissions due to fuel combustion in road transport and shipping are expected to significantly decrease as a consequence of motor and fuel modifications, such as low-sulphur fuels and particulate matter filters (e.g. Ristovski et al. (2006); Morawska et al., 2008; Fiebig et al., 2014). The EU 15 emissions are estimated to decline strongly in future years, due to implementation of new emission standards in road transport, and the phase-out of the older vehicles that have less stringent emission limits.

To facilitate the modelling of PN on a regional scale, the PN emissions were spatially distributed using available proxy data (Denier van der Gon et al., 2010b; Pouliot et al., 2012). Examples of such proxy data are maps of population density, road networks, shipping tracks, land use, and port capacities. The spatial distribution of the PN emissions has been presented in Fig. 4.

The estimates for PN emissions are associated with a relatively high uncertainty, compared with the emissions of the commonly regulated pollutants. This uncertainty varies substantially in terms of the different source categories. Vehicle-originated PNC's can change on a short timescale after the emissions exit the tailpipe, due to both rapid dilution and microphysical processes. The latter depend on ambient temperature and other environmental conditions, as well as on secondary particle formation. Due to such transformations, the PN concentration flux is not conserved. For some source categories, no PN emission factors were available. In such cases, the PN emission was calculated based on PM measurements and estimated particle size distributions.

For the road transport emission factors reported here, an uncertainty analysis for PM emission has been carried out. This analysis shows an uncertainty between 10 and 20 %, depending on the quality of the country's statistics (Kioutsioukis et al., 2010). Although PN emission factors were not included in the uncertainty evaluation of the





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above mentioned study, it is possible to indirectly estimate also the uncertainties of the PN emissions. The latter were derived by combining the available experimental data on mass and PN emissions with COPERT PM emission factors (Vouitsis, 2014).

Solid particles can be measured more accurately than semi-volatile ones; the emission standards for road transport are therefore currently based on the solid fraction of PN. The PN emissions are influenced by numerous factors, such as, e.g., vehicle category, PN measuring equipment and environmental conditions. The overall uncertainty of vehicular PN emissions can therefore be evaluated to have high uncertainties: (i) 81–144 %, when after-treatment device effects are not included and (ii) 144–169 %, when these effects are included (UNECE, 2010).

Road transport is the most intensively studied source category for PN emissions. It can therefore be expected that the uncertainties for other source categories are at least of the same magnitude. For example, the total PN emission factor is dependent on the set-ups of the measurements. In particular, the measurement can (i) include only solid PN, or solid and volatile PN, and (ii) the lower particle size cut-off used in the measurements can vary, as this is dependent on the instrumental method. Sometimes a lower cut-off of 3 nm is used, but frequently also only PN's for sizes larger than 20 or 30 nm are reported. This definition of lower size cut-off can have substantial effects on the estimates of the total PN emissions. For a more detailed discussion of the various techniques used to measure PN, we refer to McMurry (2000) and Morawska et al. (2008).

Another important uncertainty is caused by the sulphur content in shipping fuels. It is known what the regulatory limit values for the fuel sulphur content are, and in some cases also what the average fuel sulphur content is; however, it is not commonly known what the actual values are. Therefore, for all transport modes the uncertainty is expected to be at least equal to the previously listed uncertainty estimate for road transport; this is in the range of 100–170 %.

On a regional to city-scale, Kalafut-Pettibone et al. (2011) determined average size-resolved and total number- and volume-based emission factors for combustion. They





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estimated that the uncertainty of the PN emission factor is approximately plus or minus 50 %. This uncertainty value is based on longer term temporal averages.

#### 3.1.2 Emissions in the target cities

All of the emission inventories in the target cities included vehicular traffic. However, the details of the treatments for other source categories varied substantially from city to city. The urban inventories for Helsinki, Oslo, London and Athens included also the primary particulate matter emissions from shipping. The stationary sources were included at varying levels of detail for Helsinki, Oslo, London and Athens. For Helsinki, the influence of shipping and major stationary sources was estimated indirectly, but the actual PN emission values for these source categories were not included in the urban emission inventory.

The most detailed emission inventory was compiled for Oslo. The proportions of total emissions in Oslo in 2008 have been presented in Fig. 5. Traffic exhaust emissions were responsible for about 3/4 of the total emissions; the contributions from shipping, heating and other mobile sources are also notable.

### 3.2 Modelled concentrations

#### 3.2.1 Concentrations in Europe

The LOTOS-EUROS model, including the M7 module, was used together with the above mentioned new PN emission inventory, to evaluate the PNC's in Europe.

**Evaluation of predicted concentrations with measured values on a European scale**

The predicted PNC's were compared with the EUCAARI measurements (Asmi et al., 2011), with a focus on the stations of Cabauw, Vavihill and Melpitz. Cabauw is a rural site in an agricultural area with influence from the nearby city of Rotterdam;





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this is the type of region for which the model is most suitable. Melpitz is a rural site in Germany, the concentrations of which are dominated by long-range transport and biogenic emissions. This site was chosen to evaluate, how large the biogenic contribution could be. The site of Vavihill in Sweden is close to the sea; this site is representative for fairly clean background conditions, with occasional influence from shipping and nearby cities. The model performance at other EUCAARI locations have also been investigated, but the results are not discussed here.

Modelled PNC's in the Aitken and accumulation mode were compared with the observed PNC's in size bins 30–50, > 50 and > 100 nm (the latter two bins are partly overlapping). Measured and modelled monthly average PNC's for the three sites have been presented in Fig. 6. The nucleation mode was excluded, and the values correspond to the size fraction 30–250 nm for the observations, and the sum of Aitken and accumulation mode for the LOTOS-EUROS computations (defined as the interval 10–1000 nm).

At Vavihill, the modelled and observed concentrations match fairly well for the whole year. The observed concentrations from January to April at Cabauw are not comparable with the predictions, due to different settings of the measurements. At Cabauw, for the rest of the year, the overall measured and predicted levels of the PNC's were in agreement. However, the modelled concentrations at Melpitz were clearly lower than the corresponding measured values. These relatively high measured values have probably been caused by the substantial contribution of particles formed from biogenic emissions, which were not accounted for in the present model version. In winter, when the biogenic emissions are smaller, the model and observations match relatively better at Melpitz.

However, the modelling did not accurately predict the observed particle number size distributions in the EUCAARI measurements. Modelled values were on the average within a factor of two of the measured values for the Aitken mode, compared to observed particle modes in the range 30–100 nm. However, the number of particles with diameter > 100 nm was underpredicted, whereas the number of particles < 100 nm was





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in most cases overpredicted. Fountoukis et al. (2012) previously reported a similar result; a systematic under-prediction of the number of particles larger than 100 nm, using the original EUCAARI emission inventory and another chemical transport model.

These model evaluation studies indicate that the applied regional scale modelling provides reasonably accurate results for PNC's in the size range larger than 30 nm, in the presence of dominating anthropogenic emissions. In case of substantial biogenic contribution, the predicted PNC's will probably be underestimates. Clearly, the prediction of particle size distributions is a more challenging task, compared with the prediction of the PNC's.

## The spatial concentration distributions in Europe

The modelled regional scale PNC's for 2005 and 2020 are presented in Fig. 7a and b. The highest concentrations occur at urban and industrialized areas, and along the most densely trafficked shipping lanes. Annual mean concentrations reach values of up to 10 000 $cm^{-3}$ for 2005.

The PNC's are projected to substantially decline from 2005 to 2020, especially in northern and western Europe. The decline is in agreement with the predicted decrease in emissions (cf. Fig. 1), and will mainly be caused by lower PN emissions from road transport and shipping. However, the PN emissions for the non-EU27+ countries are not expected to decrease substantially. An increase of PNC's in the future is predicted in some specific areas, namely within or in the vicinity of Warsaw and Bucarest, and to a smaller extent within or in the vicinity of Paris and Frankfurt. These are also the locations with very high PNC's in 2005.

Comparing model results for the different size classes suggests that in places, in which PN emissions decrease, the number of nucleation mode particles (and to a lesser extent the number of Aitken mode particles) increases. The possible reason for this could be that in such conditions, $H_2SO_4$ does not condense so intensively on existing particles (as there are fewer particles), but will instead nucleate to form new particles. In addition, smaller particles agglomerate less rapidly to form larger particles





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(as their concentration is lower), which results in a relatively larger amount of smaller particles.

### 3.2.2 The influence of aerosol processes on an urban scale

We did not include a treatment of aerosol processes to all of the urban scale modelling systems used in this study. Instead, their influence was examined in a numerical study performed for Oslo in 2008. We have used a simplified aerosol process parametrization based on the more complex MAFOR aerosol process model and some experimental results. The numerical accuracy of the simplified model, as compared with the more complex model, was evaluated to be approximately 10 %.

The model needs as input values an initial size distribution, which was based on experimental data in Oslo, Rotterdam and Helsinki. An initial size distribution ratio was defined as the initial fraction of the total PN concentration in each size bin ($PNC_1$, $PNC_2$ and $PNC_3$). These model input values have been presented in Table 2.

The impact of this parametrisation was tested in comparison with the measured data in Oslo for a three-monthly period from January to April 2008. In these computations, the upper limit values were used both for the coagulation coefficient and the dry deposition velocity, in order to evaluate the maximum possible effects due to these processes.

Use of the parametrisation resulted in lower PNC levels further from sources. At the urban background station in Oslo (Sofienbergparken), the above mentioned parametrisation resulted in a maximum reduction of PN concentrations by approximately 45 %, compared to treating PN as a tracer. The range of this percentage value, allowing for the uncertainty of the simplified aerosol process modelling, can be considered to be approximately from 40 to 50 %. The impact of deposition was larger than that caused by coagulation; however, the influences of both processes were significant. The model-derived deposition and coagulation rates in the selected three size classes, and the relevant time scales are presented in Table 2.





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### 3.2.3 Predicted concentration distributions in the target cities

The predicted annually averaged spatial concentration distributions in the target cities are presented in Fig. 8a–e. For Oslo, the roads with average daily traffic higher than 1000 vehicles per day are shown. In the case of Rotterdam, the following roads have been marked: the motorways around the city centre and major inner urban roads that have a traffic volume higher than 10 000 vehicles per day. For Athens, a few key locations have also been marked in the figure.

The predicted spatial ranges of the annual average PNC's are approximately from 7000 to 20 000, from 3000 to 30 000, from 10 000 to 50 000, from 5000 to 50 000 and from 500 to 30 000 in Helsinki, Oslo, Rotterdam, London and Athens, respectively. The lower limits correspond to regional background concentrations, as predicted by the LOTOS-EUROS model. As expected, the upper limits of these ranges are relatively higher in the megacities, London and Athens, but also in Rotterdam, and lower in Helsinki and Oslo. The relatively high upper limits in Rotterdam (with respect to the population of the city) are caused both by the high regional background concentrations and the intensive urban traffic.

In all cities, the spatial distributions of PNC's were dominated by road traffic emissions; the major traffic networks are clearly visible. E.g., the main ring road or ring roads (for Helsinki, Oslo, London and Athens) or the main highways (for Rotterdam) surrounding the city centers are clearly visible. The concentrations were also elevated in city centers, especially in case of the megacities, London and Athens.

In Helsinki, the highest concentrations occurred in the vicinity of the most densely trafficked ring roads, and near the junctions of such ring roads and other major roads. In Oslo, the PNC's are enhanced near road tunnel entrances and in the harbor region. It was assumed that there was no deposition of particles within the tunnels; therefore all traffic-originated PN's within the tunnels were treated as emitted at these entrances. In Oslo, the annual mean concentrations were highest in the harbour area near to tunnel entrances. In Athens, there were substantially elevated PNC's near the Athens





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International Airport (denoted in Fig. 8e as A.I. Airport) and near the main harbour regions (Piraeus and Rafina).

However, the urban scale modelling in this study did not explicitly allow for the influence of street canyons. The predicted PNC's at street canyon locations, and more generally in the vicinity of locations that are influenced by high buildings are therefore inaccurate.

### 3.3 Evaluation of model predictions against measured data in the target cities

The model predictions were compared with the available PNC measurements in the target cities. Such measured data were available in four of the cities, as presented in Table 3. The predictions and measurements were compared at two stations (representing urban background and urban traffic environments) in three cities, viz. Oslo, Rotterdam and London. In the case of Helsinki, such comparisons were performed only at one station (urban traffic), due to scarcity of relevant measured data. The comparisons were performed for different years in Rotterdam (2011) and in Helsinki (2012), as the relevant measured data was not available for those cities in 2008.

The comparison in the case of annual averages is also presented in Fig. 9. The predicted and measured annual averages agree within approximately $\leq 36\,\%$ (measured as fractional biases), except for the traffic station in London. The agreement is better at urban background stations, compared with urban traffic stations, in Oslo and London, but vice versa in Rotterdam. The urban traffic station in London is Marylebone Road, which is located in a street canyon and has continuously severe traffic congestion. The measured concentration at the Marylebone Road station is substantially higher than the predicted values. The lower predicted concentration values are probably caused by the fact that the computations in this study did not allow for the effects due to street canyons, and the applied emission factors may under-estimate the influence of severely congested traffic conditions.

It was possible to evaluate the agreement of the measured and predicted hourly time series of PNC's at three stations located in two cities, Oslo and Helsinki (cf. Ta-





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ble 3). The period of these comparisons was one year in the case of Helsinki, and three months in case of Oslo. The indexes of agreement (IA) for these comparisons were 0.75 for the annual time period, and 0.77 and 0.79 for the three-monthly periods; these values indicate a fairly good agreement of measurements and predictions. Clearly, the selection of the computational method also influences on the values of the IA's. In the case of Oslo, the predicted regional background values and predicted local urban contributions were separately modelled, whereas for Helsinki, the predicted values contain measured urban background PNC values and the local contributions.

## 4 Conclusions

We have presented the results of the modelling of PNC's in five European cities in 2008. Novel emission inventories of particle numbers have been compiled both on urban and European scales (the latter is called the TRANSPHORM inventory). It has not previously been possible to conduct such computations on a European scale, due to the deficiencies of the previously available emission inventories. The TRANSPHORM PN emission inventory was based on a previous inventory that was compiled in the EUCAARI project (Kulmala et al., 2011). The new inventory focused on improving the representation of the emissions of the transport sector; major improvements were made to the previous inventory in this respect. The previous emission inventory was also substantially re-structured and improved for particulate matter emissions.

However, there are still unresolved issues on PN emissions. The semi-volatile particulate matter should also be allowed for, in addition to solid state particles. Another challenge is to allow for the short-term temporal transformations of particulate matter, after the exhaust of pollutants from an engine or an industrial process. PNC is not a conserved quantity, and the emission values are therefore dependent on the detailed definition of emissions; especially on the assumed spatial distance from the emission source. Clearly, the transformation is dependent on ambient conditions, especially on the ambient air temperature. The values of measured PN emissions are also depen-





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dent on the selected lower particulate matter limit; this is commonly determined by the capabilities of the experimental techniques. The impacts of fuel quality and the sulphur content of fuels on PN emissions are also not currently sufficiently understood.

We have compiled urban scale emission inventories in the five target cities; these are detailed and extensive for most of these cities. However, the present knowledge is not sufficiently accurate regarding the variation of PN emission factors in terms of the various source categories, especially for shipping and small-scale combustion, and for various environmental conditions. In future work, an in-depth inter-comparison of such urban emission inventories would be valuable, in terms of both the physical assumptions and the numerical emission values.

We have conducted dispersion modelling on both European and urban scales. The European scale computations included aerosol process modelling; however, it was not possible to include a detailed treatment of aerosol processes to all of the urban scale modelling systems. Instead, the influence of coagulation and deposition was examined numerically for the background air pollution in Oslo in 2008. These processes were estimated to reduce the background air PNC's maximally by approximately 40–50 % in the considered environmental conditions. The urban scale modelling also did not explicitly allow for the influence of urban buildings and other structures.

As expected, the most important local source category in terms of the PNC's was local vehicular traffic in all the target cities. In several target cities, the highest concentrations occurred in the vicinity of the most densely trafficked roads, and near the junctions of such roads and other major roads. The concentrations were also elevated in city centers, especially for the megacities of London and Athens. In Oslo, the PNC's were also enhanced near road tunnel entrances and in the harbor region. In Athens, there were substantially elevated PNC's near the airport and the main harbour regions. The highest values of the predicted PNC's were relatively higher in the megacities, London and Athens, and also in Rotterdam, whereas these were relatively lower in Helsinki and Oslo. The relatively high values in Rotterdam were probably caused by the high regional background concentrations and the intensive urban traffic.





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The predicted and measured annual average PNC's in four cities agreed within approximately $\leq 36\,\%$ (measured as fractional biases), except for one traffic station in London. We consider this agreement to be reasonable, considering the many potential uncertainties associated with the PNC modelling. The indexes of agreement (IA) for the comparisons of hourly measured and predicted time-series in Oslo and Helsinki ranged from 0.75 to 0.79, indicating a fairly good agreement. However, the amount of experimental data that could be used for model evaluation was modest: only one or two stations per city, and no relevant data was available in the case of Athens. More long-term hourly measurements of PNC's would therefore be valuable for a more extensive model evaluation in various urban locations.

## Code availability

The computer code of the LOTOS-EUROS model can be made available upon request (contact: Astrid Manders on email astrid.manders@tno.nl). The code is written in FORTRAN 90 and uses NetCDF libraries and python scripts.

The access to the CAR-FMI model for educational and non-commercial research use can be granted after signing a collaborative agreement with the Finnish Meteorological Institute (contact: Jaakko Kukkonen on email jaakko.kukkonen@fmi.fi). The code is written in FORTRAN 77.

The computer code of the EPISODE model can be made available upon request (contact: Leonor Tarrason on email leonor.tarrason@nilu.no). The code is written in FORTRAN 90.

The OSCAR model can be configured for any urban area in collaboration with the Centre for Atmospheric and Instrumentation Research (CAIR) at the University of Hertfordshire, UK. Access to the model for educational and non-commercial research use can be granted after signing a collaborative agreement with the University of Hertfordshire. The code has been developed to assess air quality and exposure to air pollution at local scales across cities (contact: Ranjeet S. Sokhi on email r.s.sokhi@herts.ac.uk).





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The model code is written in FORTRAN 90, except for emission model, which is written in Matlab.

The MEMO and MARS-aero models can be obtained for educational and non-commercial research use, after signing an end-user license agreement from the Aristotle University of Thessaloniki (contact: George Tsegas on email gtseg@aix.meng.auth.gr). The code is written in FORTRAN 95 and uses OpenMP and MPI directives.

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**Table 1.** Overview information on the computational methods and the evaluation of predictions in the five target cities for 2008.

| | Helsinki | Oslo | Rotterdam | London | Athens |
|---|---|---|---|---|---|
| Traffic flows and urban scale emissions | Traffic planning model, vehicular emission factors based on Gidhagen et al. (2005), shipping emission model STEAM2 | Local traffic data, TRANSPHORM emission database (Vouitsis et al., 2014) with temperature correction, STEAM2 | Local traffic data, COPERT IV (Gkatzoflias et al., 2012) and TRANSPHORM emission database | Local traffic data, emission factors from Jones and Harrison (2006) | Local traffic data, TRANSPHORM emission database, Petzold et al. (2010) and Lee et al., (2010). |
| Meteorological data and its pre-processing | Meteorological pre-processor model MPP-FMI, based on measured sounding data and other data from two stations | Diagnostic wind field model, based on measured data at two sites | Measured data from local airport | Meteorological pre-processor model GAMMA-met, based on measured data at one station | Prognostic model MEMO, based on measured data at one location |
| Urban source categories included | Vehicular traffic, importance of shipping and major stationary sources separately evaluated | Vehicular traffic, shipping, small-scale combustion, industry, other sources | Vehicular traffic | Vehicular traffic | Vehicular traffic, shipping, aviation, stationary sources |
| Regional or urban background concentrations and their evaluation | LOTOS-EUROS, values at a regional background grid square, and urban background values at the station of Kumpula | LOTOS-EUROS, regional background values at the grid square that contains the city | LOTOS-EUROS, regional background values at a grid square that contains the city | LOTOS-EUROS, regional background based on values both within and around the city | The PNC values by Kalivitis et al. (2008) and the values of other relevant compounds by LOTOS-EUROS, the latter at grid squares surrounding the city |
| Urban modelling system | CAR-FMI, PN treated as tracer | EPISODE, Aerosol process parameterisation included | URBIS: street-canyon and line-source models; PN treated as tracer | OSCAR, PN treated as tracer | MARS-aero, PN treated as tracer |
| Evaluation of predictions against measured concentrations | At one measurement station for one year | At two measurement stations, for three months | At two measurement stations for one year | At two measurement stations, for one year | Measurements were not available for 2008 |





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**Table 2.** Data and coefficients required for the implementation of the PNC parameterization used in Oslo. Typical predicted time scales associated with deposition and coagulation are also presented.

| Size class | Size range (nm) | Initial size distribution ratio | Dry deposition velocity $v_d$ (cm s$^{-1}$) | MAFOR derived $K_{c,i}$ (cm$^3$ #$^{-1}$ s$^{-1}$) | Typical deposition time scale (h) | Typical coagulation time scale (h) |
|---|---|---|---|---|---|---|
| $PNC_1$ | 8.5–25 | 0.79 | 0.904 | $6.31 \times 10^{-9}$ | 0.6 | 0.9 |
| $PNC_2$ | 25–100 | 0.20 | 0.202 | $5.58 \times 10^{-9}$ | 2.7 | 2.9 |
| $PNC_3$ | 100–400 | 0.01 | 0.032 | $8.82 \times 10^{-10}$ | 17 | 292 |





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**Table 3.** The comparison of measured and predicted PNC's in four target cities. IA is the index of agreement, FB is the fractional bias and NA refers to data that is not available.

| City | Name of station | Classification of station | Period | Mean of the observed values ($10^3$ particles cm$^{-3}$) | Mean of the predicted values ($10^3$ particles cm$^{-3}$) | IA (based on the hourly means) | FB |
|---|---|---|---|---|---|---|---|
| Helsinki | SMEAR III, Kumpula | Urban background | Whole year 2012 | 7.1 | NA | NA | NA |
| | Ring Road I | Urban traffic | Whole year 2012 | 19.5 | 20.0 | 0.75 | +0.02 |
| Oslo | Sofienbergparken | Urban background | Three months, Jan–Mar 2008 | 9.3 | 10.8 | 0.77 | +0.15 |
| | Smestad | Urban Traffic | Three months, Jan–Mar 2008 | 24.0 | 19.8 | 0.79 | – 0.19 |
| Rotterdam | Zwartewaalstraat | Urban background | Whole year 2011 | 14.5 | 10.1 | NA | – 0.36 |
| | Rotterdam, Bentinckplein | Urban traffic | Whole year 2011 | 17.7 | 20.1 | NA | +0.13 |
| London | North Kensington | Urban background | Whole year 2008 | 14.0 | 10.8 | NA | – 0.26 |
| | Marylebone Road | Urban traffic | Whole year 2008 | 36.7 | 15.7 | NA | – 0.81 |





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**Figure 1.** The target cities of this study.





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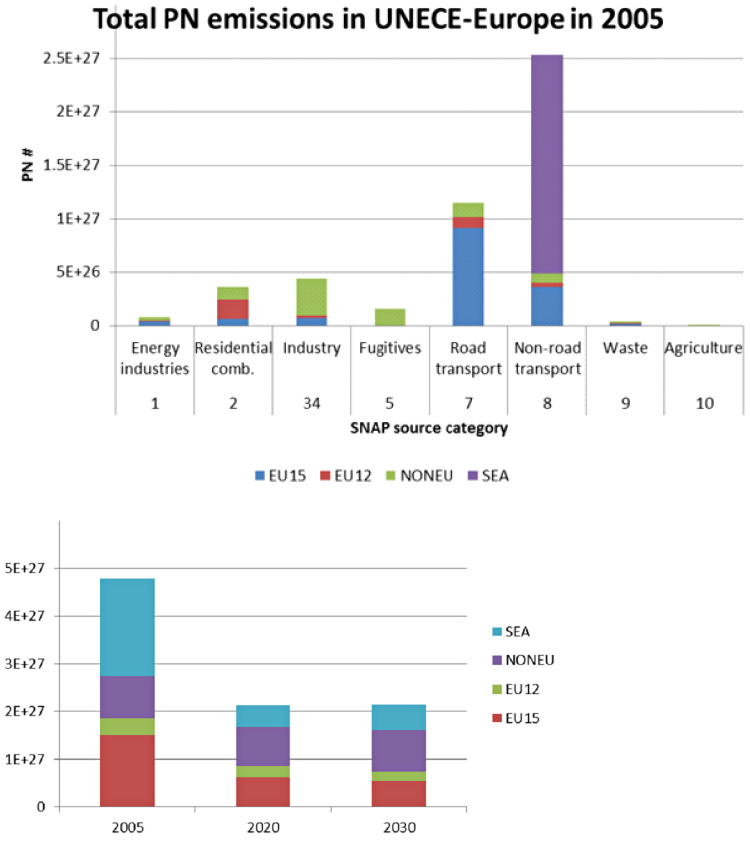


**Figure 2.** Total anthropogenic particle number emissions in the United Nations Economic Commission for Europe, **(a)** classified by the source sector for 2005, and **(b)** classified by the country group for 2005, 2020 and 2030. "Sea" refers to international shipping.





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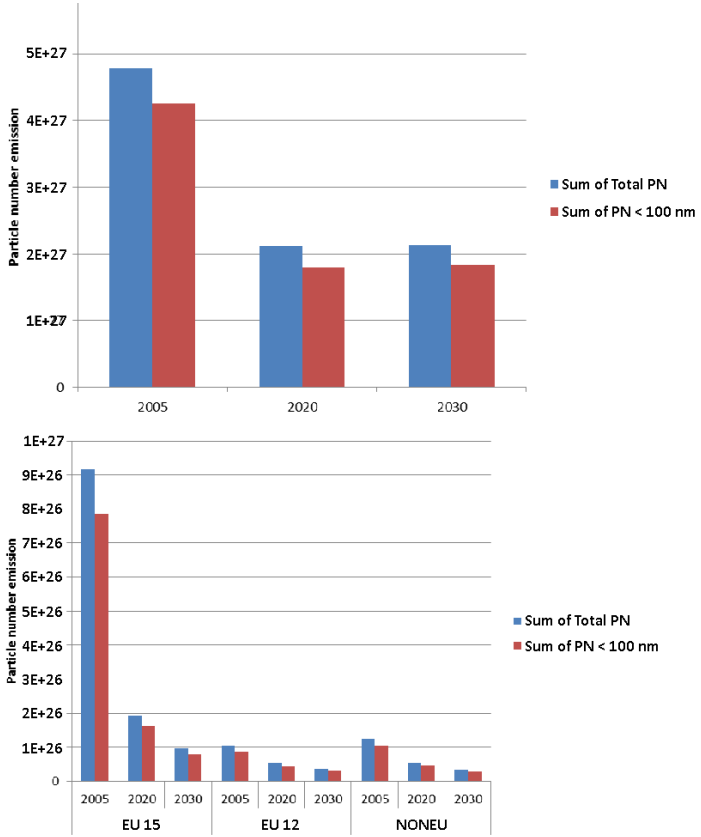


**Figure 3. (a)** Total anthropogenic particle number emissions and total particle number emissions in the particle size range of 10–100 nm for UNECE-Europe for 2005, 2020 and 2030 and **(b)** the same emissions exclusively for road transport, segmented by country group.





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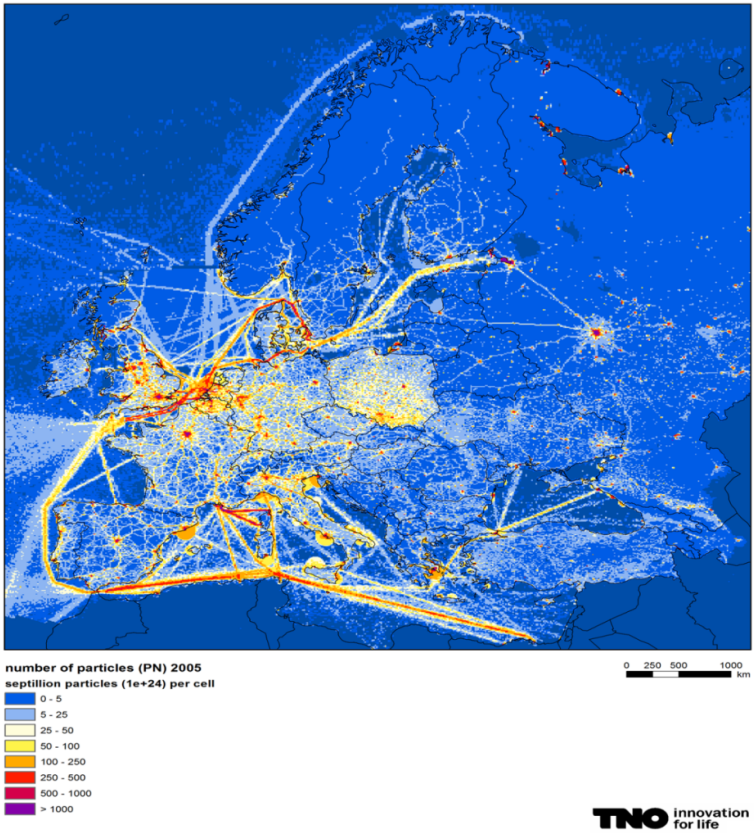


**Figure 4.** Spatial distribution of anthropogenic PN emissions in Europe in 2005, on a longitude vs. latitude grid, on a resolution of $1/8^\circ \times 1/16^\circ$. The unit of the legend is $10^{24}$ particles per computational cell per annum.





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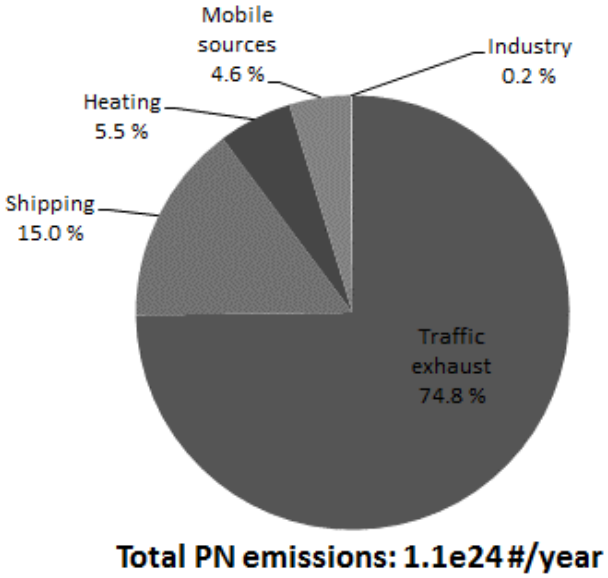


**Figure 5.** The contributions of various source categories on the total emissions of PN in Oslo in 2008.





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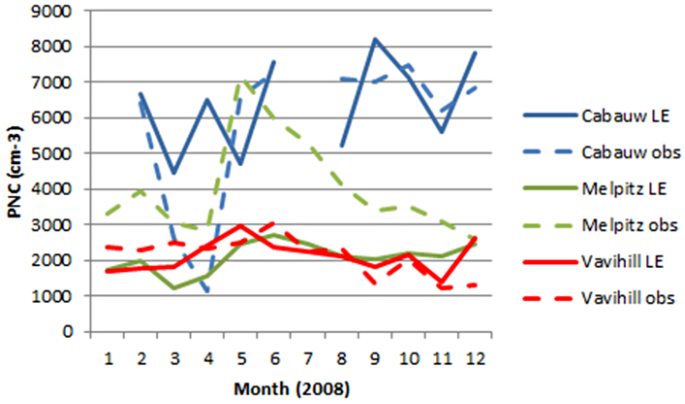


**Figure 6.** A comparison of the seasonal variation of the monthly averaged model predictions and observations of the PNC's, at three selected measurement sites in 2008. LE refers to the predictions of the LOTOS-EUROS model. The nucleation mode has been excluded; the values correspond to the size fractions 30–250 and 10–1000 nm for the observations and the model computations, respectively.





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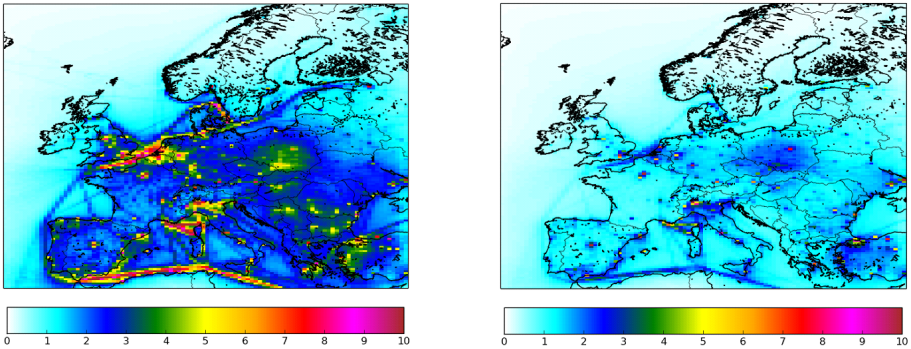


**Figure 7.** Predicted anthropogenic PNC's in Europe **(a)** for 2005 and **(b)** for 2020. The particulate matter size range is from 10 to 1000 nm. The unit in the legend is $10^3$ particles $cm^{-3}$.





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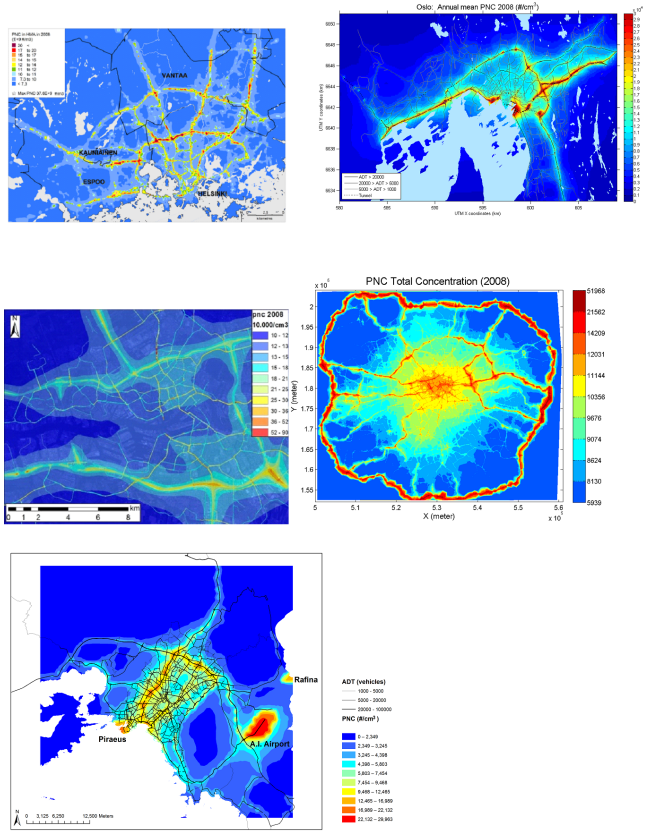


**Figure 8. (a–e)** The predicted spatial PNC distributions in the target cities in 2008. The cities in the top row are Helsinki and Oslo, in the middle Rotterdam and London and in the bottom row Athens. ADT is the average daily traffic. The concentration units in the legend are $10^9$ particles per $m^3$ for Helsinki (i.e., $10^3$ particles per $cm^3$), $10^4$ particles per $cm^3$ for Oslo, $10^3$ particles per $m^3$ for Rotterdam, and particles per $m^3$ for both London and Athens.





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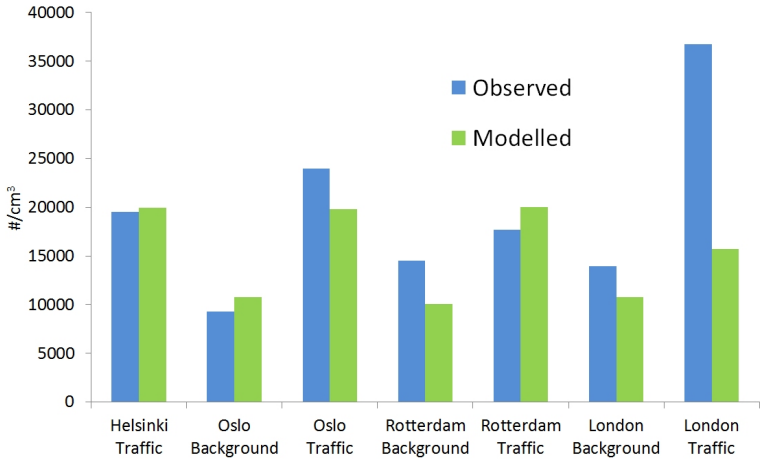


**Figure 9.** Comparison of the predicted and measured annual average PNC's in five cities. The names of the stations have been specified in Table 3. "Background" refers to urban background stations. The unit of PNC's is particles $cm^{-3}$.





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