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Modelling and measurements of urban aerosol processes on the neighborhood scale in Rotterdam, Oslo and Helsinki

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Abstract

This study evaluates the influence of aerosol processes on the particle number (PN) concentrations in three major European cities on the temporal scale of one hour, i.e. on the neighborhood and city scales. We have used selected measured data of particle size distributions from previous campaigns in the cities of Helsinki, Oslo and Rotterdam. The aerosol transformation processes were evaluated using an aerosol dynamics model MAFOR, combined with a simplified treatment of roadside and urban atmospheric dispersion. We have compared the model predictions of particle number size distributions with the measured data, and conducted sensitivity analyses regarding the influence of various model input variables. We also present a simplified parameterization for aerosol processes, which is based on the more complex aerosol process computations; this simple model can easily be implemented to both Gaussian and Eulerian urban dispersion models. Aerosol processes considered in this study were (i) the coagulation of particles, (ii) the condensation and evaporation of n-alkanes, and (iii) dry deposition. The chemical transformation of gas-phase compounds was not taken into account. It was not necessary to model the nucleation of gas-phase vapors, as the computations were started with roadside conditions. Dry deposition and coagulation of particles were identified to be the most important aerosol dynamic processes that control the evolution and removal of particles. The effect of condensation and evaporation of organic vapors emitted by vehicles on particle numbers and on particle size distributions was examined. Under inefficient dispersion conditions, condensational growth contributed significantly to the evolution of PN from roadside to the neighborhood scale. The simplified parameterization of aerosol processes can predict particle number concentrations between roadside and the urban background with an inaccuracy of ~ 10 %, compared to the fully size-resolved MAFOR model.

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1 Introduction

Motor vehicle exhaust emissions constitute the major source of ultrafine particle (UFP, < 100 nm in aerodynamic diameter) pollution in urban environments (Harrison et al., 2011; Morawska et al., 2008; Pey et al., 2009; Johansson et al., 2007). Ultrafine particles can contain toxic contaminants, such as transition metals, polycyclic aromatic hydrocarbons (PAHs), and other particle-bound organic compounds, which may be responsible for initiating local lung damage, when the particles deposit on the epithelial surfaces (Lighty et al., 2000). Biodistribution studies suggest translocations of UFP from the respiratory system to other organs including liver, heart and the central nervous system, in which they can cause adverse health effects (Oberdörster et al., 2005; Kleinman et al., 2008; Kreyling et al., 2013). In urban environments, ultrafine particles make the most significant contribution to total particle number (PN) concentrations, but only a small contribution to particulate matter (PM) mass. Hence, reliable information on the number concentrations, together with the size distributions, is needed to better assess the health effects of urban particulate pollution.

The exposure of the population in urban areas to particles may be assessed by modelling the spatial distribution of particles emitted from road transport and other sources in various micro-environments (e.g., Soares et al., 2014). Kumar et al. (2011) reviewed aerosol process modelling on urban and smaller scales. Aerosol dynamic models (which are commonly Lagrangian-type process models) have been used to model the spatial and temporal evolution of ultrafine particles in the initial vehicle exhaust plume during the first seconds after emission (e.g. Vignati et al., 1999; Pohjola et al., 2003, 2007). These models can be used to study the further evolution of the plume, if they will be coupled to an urban dispersion model. Particles emitted from road transport, as they are transported from the emission sources, are subject to complex dilution processes (turbulence generated by moving traffic, atmospheric turbulence) and transformation processes (nucleation, coagulation, condensation, evaporation, deposition, and heterogeneous chemical reactions), acting on different time scales. Aerosol

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dynamic processes continuously change the number and size distribution, after the particles have been released into air.

Clearly, dilution is an important process influencing PN concentrations and the spatial distributions in cities (e.g. Zhang et al., 2004; Pohjola et al., 2007; Keuken et al., 2012). An exhaust parcel emitted from the tailpipe of a vehicle first experiences fast dilution by the strong turbulence generated by moving traffic between tailpipe to roadside (Rao et al., 2002; Zhang and Wexler, 2004). On the neighborhood scale in the city, the parcel of exhaust is advected through a network of streets, over and around several buildings. On the city scale, the pollutant plume can extend vertically up to twice the average building height above the city's surface layer, and its dispersion becomes independent of the specific effects of individual buildings (Kumar et al., 2011).

The main aims of the present study are (i) the quantification of the impacts of relevant aerosol processes on the neighborhood and city scales and (ii) the derivation of a reasonably accurate, simplified parameterization of the most important aerosol processes, to be used in urban air quality models. The study is part of the European Union funded research project TRANSPHORM (Transport related Air Pollution and Health impacts – Integrated Methodologies for Assessing Particulate Matter). A related paper by Kukkonen et al. (2015) presents atmospheric dispersion modelling of particle number concentrations in the five target cities of the TRANSPHORM project, as well as on a European scale, and evaluates the predicted results against available measured concentrations. In the present model study, we have used the results of measurements from a campaign in Rotterdam, initiated by the TRANSPHORM project, and those from previous campaigns in Helsinki and Oslo. Our aims are to quantify the influence of selected individual aerosol processes for each measurement campaign and to inter-compare the relative contribution of the processes to PN changes in the selected campaigns and cities.

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2 Materials and methods

2.1 Aerosol process model MAFOR

In order to study the relevance of aerosol dynamics on the fate of PN emitted from traffic in urban areas, the evolution of the particle size distribution with increasing distance from the roadside was modeled using the multicomponent aerosol dynamics model MAFOR (Karl et al., 2011). MAFOR uses a fixed sectional grid to represent the particle size distribution with size bins evenly distributed on a logarithmic scale. MAFOR has been evaluated against laboratory chamber data (Karl et al., 2012) and PN measurements at a motorway (Keuken et al., 2012); it has also been shown to compare well with the sectional aerosol dynamics model AEROFOR (Pirjola and Kulmala, 2001).

Aerosol processes considered in this study were condensation and evaporation of n-alkanes, coagulation of particles due to Brownian motion, and dry deposition (particle deposition in contact with the street surface and other urban structures). In this study 120 size bins were used in the MAFOR model, to represent the aerosol size distribution ranging from particle diameters from 1 nm to 1 µm.

Particles were assumed to be spherical and possible effects of the fractal geometry were disregarded; the exact shape and morphology of particles was not measured in the addressed campaigns. The various aerosol dynamical processes were treated by calculation of the temporal variation of the particle number concentration and the mass concentrations of each chemical component within each size section. Mass transfer of gas molecules to particles was calculated using the Analytical Predictor of Condensation scheme (Jacobson, 1997).

The coagulation coefficients of particles that are smaller than 50 nm in diameter might be enhanced due to Van der Waals forces and viscous interactions; however, these were neglected, due to the large uncertainties involved in the modelling of such processes. Further, effects of turbulent shear on coagulation between exhaust particles can be neglected for the time scale from roadside to ambient (Zhang and Wexler, 2004).

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Dry deposition of particles was modelled according to Kouznetsov and Sofiev (2012), which accounts for the physical properties of both the air flow and the surface, as well as the physical properties of the particle size. In this approach rough surfaces are characterized by two length scales: the aerodynamic roughness and the so-called collection scale, which incorporates the effective size of collectors and a ratio of the airflow velocity at the top of the roughness elements to the friction velocity. Alternatively, MAFOR provides a treatment to calculate size-dependent deposition rates according to Schack et al. (1985) and Hussein et al. (2012).

Two different n-alkanes (i.e., $C_{22}H_{46}$ and $C_{28}H_{58}$) were used to represent vehicular exhaust gases with different volatilities, which can condense or evaporate to or from particles during their transport downwind from the road. Vapor pressure of n-alkanes as function of temperature was adopted from the work by Lemmon and Goodwin (2000). Carbonaceous aerosol in MAFOR is separated into (i) elemental carbon (EC_p) from primary emissions, treated as a non-volatile substance, and (ii) organic carbon (OC_p), treated as a volatile substance. In this study, organic carbon is assumed to be composed of organic acid (for background OC_p) and the two n-alkanes (originated from vehicles). A density of 1200 $kg\,m^{-3}$ (Virtanen et al., 2002) was used for EC_p. The density of OC_p was calculated as the weighted average of the densities of the organic compounds.

2.2 Modelling of the dilution of exhausts

MAFOR is a one-dimensional model; it is therefore necessary to couple it to a dispersion model, to simulate combined atmospheric dispersion and transformation processes. In order to approximate atmospheric dispersion, we used a simplified treatment of dilution of particle numbers. This procedure implies the assumption of a well-mixed state within each cross-wind cross-section of the plume. The assumption of a well-mixed state may overestimate the influence of the processes responsible for the temporal decrease of the PN, due to the non-linear nature of the involved processes

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(condensation and coagulation). Model runs were performed with different dispersion conditions to address the influence of aerosol processes for a wide range of meteorological dispersion regimes.

Emissions from traffic sources commonly contribute to particle size distributions with distinct modes, i.e. nucleation, Aitken, accumulation, and coarse mode. Formation of new liquid particles in the exhaust by nucleation of gases, such as sulfuric acid and semi-volatile organic substances, occurs during the first milliseconds (Kittelson, 1998) after release of the exhaust into the ambient air. On-road measurements by Rönkkö et al. (2007) confirmed that the nucleation mode was already present after 0.7 s residence time in the atmosphere. Thus, it is practical to regard nucleation as a process that has already occurred, when one considers roadside concentrations. The evolution of vehicular emissions from the engine to the roadside concentrations were not considered in this study, as we used the particle size distributions measured at the roadside locations as a starting point.

Idealized scenarios were set up for the study of relevant aerosol processes (i.e. the dry deposition, the growth by condensation of gases and the coagulation of particles) and dilution by background air (see Fig. 1).

We used a simple horizontal particle dilution parameterization, following the numerical power function $y = a \times x^{-b} = a \times (Ut)^{-b}$, where x (in m) is the distance from the roadside and U is the horizontal wind speed perpendicular to the road. The height of the air parcel (plume height), H_m (in m), containing the exhaust emissions, as function of time t (in s) during its travel away from the roadside at a specific wind speed was defined by:

$$H_m(t) = \sqrt{H_{m,0}^2 + \left(a \times \left(10^{-3} \times Ut\right)^b\right)^2} \quad (1)$$

Where $H_{m,0}$ is the initial plume (or air parcel) height at the roadside. The particle dilution rate for use in the aerosol model was obtained by derivation of the above mentioned numerical power function as function of time. The change of particle number concen-

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tration, N (in particles cm^{-3}), in size section i due to dilution with background air is:

$$\frac{dN_i}{dt}|_{\text{dilution}} = -\frac{b}{t}\left(N_i - N_{\text{bg},i}\right) \quad (2)$$

Where $N_{\text{bg},i}$ is the number concentration of background particles in the same size bin.

Dilution parameters a and b that are used in Eqs. (1) and (2) for moderate dispersion conditions were derived from a fit of the modelled total number concentration to measured number concentration in different distances (below 100 m) from a major highway in Helsinki (LIPIKA campaign, case 10; Fig. 5 in Pohjola et al., 2007). It was assured that the PN change in the distance up to 100 m was solely due to dilution with background air. Best fit was obtained with parameter values $a = 40$ and $b = 0.5$. For neutral conditions, the values $a = 86.49$ and $b = 0.923$ were reported for dispersion downwind of a motorway (Petersen, 1980). Similar values were adopted for efficient dispersion conditions in this study ($a = 80.0$ and $b = 0.90$). For inefficient dispersion conditions, a and b were chosen to be typical for atmospheric situations with inversion and stagnant air. Table 1 provides an overview of the set of meteorological and dilution parameters that were tested in sensitivity studies.

The model simulations were started by assuming an initial chemical composition of the aerosol at the respective roadside traffic site and urban background site. Chemical composition of the urban background aerosol was estimated based on the measured $PM_{2.5}$ and the mass fractions of the chemical components in $PM_{2.5}$. Composition of the nucleation, Aitken, accumulation and coarse modes was estimated based on mass fractions for the urban background of Helsinki (Pohjola et al., 2007). Then mass concentrations of the respective lognormal modes were distributed over the discrete size sections of the model. The aerosol composition of the traffic-influenced aerosol at the traffic station was approximated by adding mass concentrations of OC_p and EC_p (from vehicle exhaust emissions) to the mass concentrations of the background aerosol. Fixed modal $OC_p : EC_p$ ratios were used (nucleation mode: 100 : 0, Aitken mode: 80 : 20, accumulation mode 1: 40 : 60, accumulation mode 2: 60 : 40) based on

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the mass composition of vehicle exhaust particle emissions (Pohjola et al., 2007). Finally, the initial model number size distribution was fitted to the observed number size distribution at the traffic site for each of the campaigns, by variation of the geometric-mean mass diameter (by ±30 %) and the geometric standard deviation (within the range 1.5–2.0) in each lognormal mode.

Additional sensitivity tests were carried out to address uncertainties in the modelling with respect to (i) dry deposition of particles to urban surfaces and (ii) assumptions about the roadside concentrations of condensable organic vapors (represented as n-alkanes). Details on the sensitivity test for condensable organic vapors are given in Sect. S1 in the Supplement.

2.3 The effect of different surface types on the dry deposition of particles

As the air parcel containing vehicle exhaust leaves street scale, it can be assumed to be advected through a network of streets, and over and around buildings, defined as the neighborhood scale with a characteristic length scale of 1–2 km. On the neighborhood scale, geometrical features dominate mean flow and mixing. Effects caused by buildings and other structures are disregarded in this study. Instead the flow was assumed to have a long fetch over a statistically homogeneous surface. However, different average surface types may have an impact on dry deposition of particles. In a series of tests the sensitivity of PN changes were studied, caused by dry deposition on various surface types and roughness conditions. Table 2 provides a summary of relevant parameters for dry deposition used in the reference case (all campaigns and dispersion conditions) and in the sensitivity tests (selected campaigns).

The parameterization used in the reference runs is thought to represent dry deposition to typical urban surfaces, i.e. streets and buildings (urban case). Values for friction velocity near surface, u^*, and roughness height, z_0, used in the urban case were adopted from the work of Ketzel and Berkowicz (2004). Sensitivity tests for dry deposition were performed for the campaigns in Rotterdam and Oslo using the dilution parameters for moderate dispersion conditions.

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The methodology by Kouznetsov and Sofiev (2012) considers Brownian diffusion, interception, inertial impaction and gravitational settling as mechanisms for dry deposition to rough surfaces. They define a collection length scale to characterize the properties of rough surfaces. This collection length depends on the ratio U_{top}/u^* (U_{top} is the wind speed at top of the canopy, i.e. at height z_C) and to the effective collector size, d_{col}, of the canopy. The methodology by Hussein et al. (2012) is a three-layer deposition model formulation with Brownian and turbulent diffusion, turbophoresis and gravitational settling as the main particle transport mechanisms to rough surfaces. Hussein et al. (2012) introduced the effective surface roughness length F^+ to relate roughness height and the peak-to-peak distance between its roughness elements. For a hydraulically smooth surface, F^+ approaches zero. Parameters z_0, z_C and d_{col} are only used in the concept of Kouznetsov and Sofiev (2012) while F^+ is only used in the concept of Hussein et al. (2012). Size-dependent dry deposition velocities of particles were calculated with two different methodologies: The methodology of Kouznetsov and Sofiev (2012) (short: KS2012) was applied in the reference case for all simulations. In addition the methodology of Hussein et al., 2012 (short: H2012) was applied for all cases in the sensitivity test.

Figure 2 shows size-dependent dry deposition velocity of particles for the different cases listed in Table 2 for the two methodologies. The curve "KS2012 Urban" (thick black line) represents the parameterization used in the reference runs of this study. Dry deposition velocities calculated by H2012 for the urban case ("H2012 Urban") agree with "KS2012 Urban" within a factor of 3 as function of particle diameter. Results from the KS2012 methodology were not sensitive to changes of friction velocity within a range (0.27–1.33 $m\,s^{-1}$) typical for the urban environment.

A large discrepancy between the two methodologies was found for deciduous forest (green area with forest). H2012 closely matches measured dry deposition velocities over a beech forest by Pryor (2006) when using $F^+ = 2.25$. Kouznetsov and Sofiev (2012) state that their parameterization offsets measured data for broad-leaf forests by 2–3 orders, unless using a very small collector size ($d_{col} < 0.2$ cm). However, their

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parameterization is in close agreement with one wind tunnel measurement for 1 µm particle deposition on natural oak branches by Reinap et al. (2009). It should be kept in mind that "KS2012 Forest" does not necessarily represent realistic dry deposition rates to forests and was rather included as lower limit for particle deposition from the KS2012 method in urban environments.

2.4 Experimental data from the measurement campaigns

We have used the measured particle number size distributions at a traffic station and at an urban background (UB) station, during campaigns in the cities Oslo, Rotterdam and Helsinki. The included campaign datasets were:

1. Rotterdam 2011, TRANSPHORM. Traffic site Bentinckplein and urban background location Zwartewaalstraat (6–19 May 2011) at Rotterdam, and the regional background station at Cabauw in the Netherlands (February–November 2011).
2. Oslo 2008, UFP-Oslo. Traffic site Smestad and urban background location Sofienberg park (12 December 2007 to 17 April 2008).
3. Helsinki, SAPPHIRE case I. Traffic site at Herttoniemi and urban background location at Kumpula, Helsinki, 23–28 August 2003 (Hussein et al., 2007).
4. Helsinki, SAPPHIRE case II. Traffic site at Herttoniemi and urban background location at Kumpula, Helsinki, 9–11 February 2004 (Hussein et al., 2007).
5. Helsinki LIPIKA. Traffic site at Herttoniemi and urban background location at Saunalahti bay, Helsinki, 17–20 February 2003 (Pirjola et al., 2006; Pohjola et al., 2007).
6. Helsinki, MMEA. Traffic site at Mannerheimintie and urban background location at Lääkärinkatu, Helsinki, 13–14 December 2010 (Pirjola et al., 2012).

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The measured data for roadside and urban background in the TRANSPHORM campaign at Rotterdam, in the UFP-Oslo ("Measurements of ultrafine particles in Oslo") campaign at Oslo, and in the SAPPHIRE campaigns in Helsinki was obtained simultaneously. Whereas in the LIPIKA and the MMEA campaigns in Helsinki the measured data was obtained with the mobile laboratory "Sniffer" (e.g., Pirjola et al., 2004) at various locations during each measurement day. Quality control (QC) procedures in the measurement campaigns at Helsinki are described in the cited literature. Table 3 compiles the information on the size distribution data from the different campaigns in Rotterdam, Oslo, and Helsinki. In order to obtain an average size distribution for the respective traffic station and urban background station, either median or mean of measured time series of size distributions ($dN/d\log(D_p)$) were calculated, as specified in Table 3. A comparison of measured total PN concentrations between campaigns is shown in Fig. S1 (Supplement).

In Rotterdam, particle measurements were performed at the regional background station at Cabauw near Rotterdam and a traffic location at less than 5 m from the roadside (Bentinckplein) by two Scanning Mobility Particle Sizer (SMPS) instruments: one SMPS 3080 covering size diameters (D_p) 10–480 nm and a CPC 3775 (TSI Inc.) with a with a 50 % cut-off at 4 nm, and one SMPS 3034 with D_p 10–470 nm and a CPC 3010 (TSI Inc.) with a 50 % cut-off at 7 nm. The comparability of both SMPS was tested by parallel measurements, which resulted in a correlation coefficient (r^2) of 0.96. The results of the PNC measurements were corrected for the difference in comparability between both instruments. The traffic volume at Bentinckplein, which is a street canyon (width: 50 m; height: 12 m) was 35 000 vehicles per 24 h with 4 % trucks and buses. The urban background location Zwartewaalstraat in Rotterdam total PN concentrations were measured by a Condensation Particle Counter (TSI 3007). The entire monitoring period at the regional background site was from February until December 2011. QC procedures were derived from the European Supersites for Atmospheric Aerosol Research (EUSAAR) project (Asmi et al., 2011). These involved inter-comparison studies

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of monitoring instruments, 2-weekly checking of the sampling flow and annual calibration of the PN monitors by the manufacturer.

In Oslo, PN concentration and particle size distributions were measured at two stations in the municipality of Oslo for a four month period in winter 2008, using a Grimm 565 Environmental Wide Range Aerosol Spectrometer system (http://www.GRIMM-Aerosol.com). This system combines a Grimm 190 aerosol spectrometer OPC (Optical Particle Counter), and a scanning mobility particle sizer with a condensation particle counter (SMPS+C). The entire system in principle covers the range from 5 nm to 30 µm. Instruments were calibrated by the manufacturer prior to installation on site. Weekly zero filter test and other maintenance was carried out according to the manufacturers guide. In addition to automatic QC in the Grimm software, data from the two sites was compared and aligned with other air quality data from the respective sites. For the analysis of the Oslo campaign, only data from the SMPS was used and the smallest size bin was discarded. The traffic station (Smestad) was at a busy road with an average daily traffic (ADT) of around 50 000 vehicles. The traffic signal at the urban background station (Sofienberg park) showed a continuous shift of the size distribution peak towards larger sizes with decreasing air temperature, i.e. the maximum of the size distribution is shifted from 16 nm at 6 °C to 26 nm at −10 °C. Size distribution data measured at −8 to −12 °C was used as a separate dataset, UFP-Oslo Winter. The complete dataset from the period December 2007 to April 2008 is referred to as UFP-Oslo Tav.

For Helsinki, two cases from the SAPPHIRE campaign, one case from the LIPIKA campaign (both at Herttoniemi), and one case at the city center from the MMEA campaign (Pirjola et al., 2012) were included. The roadside station near the highway Itäväylä at Herttoniemi is located about 6 km east of the center of Helsinki in a suburban area, with substantial local traffic. Particle measurements were performed with a differential mobility particle sizer (DMPS) at the background station and with a twin SMPS at the traffic site. Particle measurements during the LIPIKA campaign were conducted by Sniffer at various locations near the highway Itäväylä (Pirjola et al., 2006).

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The highway consists of six lanes, three lanes to both directions (total width of three lanes: 12 m), and a 6 m wide central grass area between the lanes to both directions, with a speed limit of 80 km h^{-1}. Particle size distributions in the range of 7 nm to 10 µm (aerodynamic diameter) were measured by Electrical Low Pressure Impactor (ELPI, Dekati Ltd.; 12 channels). Nucleation mode particles were measured with high size resolution by a Hauke type SMPS (20 channels); measured size range was 3–50 nm (mobility diameter). The study period included 14 cases of measurements downwind of the highway Itäväylä from wind sector 1 (northwestern wind; Pirjola et al., 2006). The daily traffic density varied between 32 000–54 000 vehicles per day. Based on the traffic density information for year 2001, the vehicle fleet on the highway was composed of 85 % light duty vehicles (of which 11 % were diesel), 12 % vans (of which about 84 % diesel), and 4 % heavy duty vehicles (Hussein et al., 2007).

During the MMEA campaign (Pirjola et al., 2012) the mobile laboratory "Sniffer" was driving along the main street Mannerheimintie (MA) at the city center of Helsinki. MA is about 40 m wide and surrounded by 21 m tall buildings at both sides. The daily traffic flow was 36 300 vehicles day^{-1} (of which ca. 10 % were heavy duty diesel vehicles). On 13–14 December 2010, the northeastern wind was perpendicular to MA, allowing traffic exhaust to be diluted freely between the buildings as in open environments. During rush hours, "Sniffer" was stopping around 10 min at 8, 28 and 56 m distances from the driving lane of MA downwind. Particle size distribution was measured by two SMPS (size ranges: 3–60 and 10–420 nm). The urban background particles were measured while Sniffer was standing at Lääkärinkatu, 300 m north from the measurement sites at MA.

A summary of the meteorological and dispersion conditions for the different campaigns is given in Table S1 in the Supplement. Measured meteorological data was not directly used in the model study of idealized scenarios, but are considered to be important for discussing the relevance of aerosol dynamical processes compared to dilution under real world conditions.

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3 Results

3.1 Traffic-related particle size distributions in the campaigns

Measured PN concentrations based on hourly averages or 10 min averages (in case of the LIPIKA and MMEA campaigns) showed a wide range of PN concentrations (20 000–100 000 particles cm^{-3}) for the traffic sites considered, depending on the season of the year, traffic density, and distance from the road. Size distributions of the measured datasets at the traffic sites from all campaigns were normalized by the measured total PN concentrations (Fig. 3). We have also calculated the average values of the size distribution curves for all traffic sites, denoted as "mean of traffic sites" (black curve in Fig. 3). This size distribution is considered to be representative for the traffic-influenced roadside aerosol in the considered cities.

The "mean of traffic sites" distribution is characterized by a fraction of ultrafine particles (D_p: 10–100 nm) and accumulation mode (Acc) particles (D_p: 100–500 nm) of 80 and 4 %, respectively, while 16 % of the particles were below 10 nm. SAPPHIRE Case I had the highest fraction of < 10 nm particles (38 %); in Rotterdam < 10 nm particles were not measured. For the other campaigns fraction of < 10 nm particles was in the range 6–19 %. The fraction of ultrafine particles was 60 % for SAPPHIRE Case I and 74–87 % for all other campaigns. The fraction of Acc particles was smallest for the SAPPHIRE campaigns (≈ 1 %), and between 4 and 9 % for the other campaigns in Helsinki and Oslo.

The mean traffic-related size distribution shows three distinct modes with mean diameter at 17, 85, and 250 nm, respectively. The measured average size distributions from the campaigns LIPIKA and MMEA in Helsinki, as well as UFP-Oslo Tav exhibited a similar shape as the mean traffic-related size distribution. The distribution of SAPPHIRE Case II also resembled the constructed distribution but did not show significant particle numbers with diameter > 100 nm. Ultrafine particles measured at the traffic sites in the campaigns were most likely from fresh vehicle exhaust emissions. Particles emitted from diesel engines are usually in the size range 20–130 nm (e.g., Kittelson, 1998),

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somewhat larger than those emitted from gasoline engines, typically being in the range 20–60 nm (Harris and Maricq, 2001; Ristovski et al., 2006). Comparing campaigns in Helsinki, MMEA size distribution peaked at 20–40 nm, while LIPIKA and SAPPHIRE size distributions peaked at 8–25 nm. The higher fraction of heavy duty diesel vehicles at Mannerheimintie (10 %) compared to highway Itäväylä (4 %) could be one possible reason for the peak at relatively larger sizes in MMEA. Different driving conditions may also have contributed to the difference in peaks; at Mannerheimintie rush hour limited the speed to 20–25 km h^{-1} with stop-and go driving, whereas at Itäväylä vehicles could drive 60–80 km h^{-1} very fluently.

The peak of the size distribution for UFP-Oslo Winter was below 10 nm and for Helsinki SAPPHIRE Case II the peak was below 20 nm. Both campaigns were during winter in Northern Europe; ambient temperature in UFP-Oslo Winter ranged from −12 to −8 °C and SAPPHIRE Case II ranged from −15 to −4 °C. The relative increase of nanoparticle numbers in cold conditions may be the result of increased nucleation of semi-volatile compounds post-emission and decreased saturation ratio of the condensing vapors that tend to enhance initial particle growth. It has also been reported that particle emission from light duty diesel vehicles are influenced by low ambient temperatures during the vehicle cold-start (Mathis et al., 2005). However, the primary effect of a cold environment on vehicle cold-start is a number increase of semi-volatile nucleation mode particles, not of the solid particles in the exhaust (Maricq, 2007).

Accumulation mode particles have a longer lifetime in the atmosphere, it is therefore likely that they are either a result from ageing processes on the urban time scale or that they are from short-range or long-range transport of aerosols. Since the size distribution measurements were carried out at traffic sites at distances of a few meters from busy roads, the measured aerosols are expected to be mainly influenced by primary traffic emissions. However, for the campaigns at Rotterdam and Oslo, measurements were not always downwind from the traffic emissions, and could be influenced also by other local particle sources and secondary pollution from local traffic.

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The number size distribution at Rotterdam showed an exceptionally broad peak mode at 30–70 nm and a large fraction of Acc particles. Other sources, such as emissions from harbor activities and refineries situated in the harbor area, could have contributed to the relatively high fraction of Acc particles at Bentinckplein. Average wind direction during the Rotterdam campaign was from southwest, from the direction of the harbor area "Nieuw Mathenesse". At an average wind speed of 3.6 m s^{-1}, the travel time of particles from the harbor and refineries to the traffic site was about 15 min. Ships emit large amounts of particles larger than 20 nm, which consist of soot and volatile material (e.g. Fridell et al. 2008; Petzold et al., 2008; Kasper et al., 2007). Number size distributions of ship emissions in the ports of Helsinki and Turku (Finland) measured by "Sniffer" showed peaks at around 20–30 and 80–100 nm (Pirjola et al., 2014).

Truck traffic in the harbor during loading/unloading of ships also leads to increased particle numbers (Pirjola et al., 2014). Measurements downwind of a harbor at Rotterdam showed that 61 % of the PN concentration was in the size range 25–100 nm while it was 48 % downwind of a motorway (Keuken et al., 2012). Condensation of vapors onto particles emitted from ships during their transport to the traffic site might explain the relatively high number concentration of Acc particles in the Rotterdam campaign. We note that the measured PN concentration of Acc particles in Rotterdam was similar as in Oslo but lower than during the LIPIKA and MMEA campaigns in Helsinki.

3.2 Comparison of the model predictions against the campaign measurements

The evolution of the particle size distribution as function of time up to one hour for all the cases, and by definition, following increasing distance from the roadside (idealized scenario, Sect. 2.2), was studied with the aerosol dynamics box model and compared to measured size distributions data from the respective campaigns. Figure 4 and Fig. S2 in the Supplement show the comparison of modelled number size distributions from the idealized scenarios and the measured number size distribution at the roadside and at the urban background site for campaigns at Oslo, Rotterdam, and Helsinki. As the air

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parcel containing vehicle exhaust leaves street scale it is assumed to be advected over a homogenous surface in the neighborhood with a length scale of a few kilometers and further to the city scale. In the model, initial particle concentrations in all size bins were diluted with background air containing particles with a size distribution that matched the measured size distribution at the urban background site.

The modelled number size distribution calculated for moderate dispersion conditions after ~ 10 min of travel time, corresponding to a distance of 3600 m from the roadside, was generally in good agreement with the size distribution measured at the urban background site. Dilution was the dominant process changing the size distribution between roadside and urban background, as shown by the continuous decrease of concentrations with time. For the campaigns Helsinki SAPPHIRE Case I (Fig. 4c) and Case II (Fig. S2c in the Supplement), as well as Helsinki MMEA (Fig. 4d) the maximum of the particle size distribution was moved to larger diameter. For instance, the modelled size distribution in Helsinki SAPPHIRE Case I showed an increase of the nucleation mode peak diameter from 10 to 18 nm within a distance of 3600 m. This behavior can be explained by dilution transforming the shape of the roadside distribution into the (prescribed) shape of the urban background distribution. UFP-Oslo Winter (Fig. 4a) shows signs of growing small particles by condensation, with peak diameter moving from ca. 5 to 8 nm.

Model simulations using different wind speed and dilution parameters, representative for different dispersion conditions (efficient, moderate, inefficient dispersion; as given in Table 1), were performed for each campaign. The contribution of the various aerosol dynamic processes to the change of total PN at a given travel time was derived by switching off the respective aerosol process in the model calculation. The percentage PN change due to a specific aerosol dynamic process was obtained by division of the total PN change with the total PN change when all processes were considered (PN change defined as difference between initial total particle number and total particle number after a certain travel time). Table 4 summarizes the PN change after 30 min of travel time due to each selected aerosol process in each of the campaigns for efficient,

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moderate and inefficient dispersion conditions. The considered aerosol processes accounted for PN concentration changes of up to 20 % after 10 min and up to ~ 30 % after 30 min (Fig. 5), respectively.

According to the results shown in Table 4, coagulation and dry deposition were relevant aerosol dynamic processes for particle removal in the Rotterdam campaign whereas dry deposition was the predominant aerosol process in the Oslo campaign. Due to identical dispersion conditions and wind speeds used in the comparison, the observed difference is attributed to the different shapes of the initial size distribution measured at the roadside station and the background particle size distributions. The larger fraction of particles with diameter > 25 nm measured at Rotterdam (accounting for 73 % of total PN) explains the higher relevance of coagulation compared to the Oslo campaign. Particles with larger diameter more efficiently scavenge the small (< 25 nm diameter) particles by coagulation.

For the LIPIKA and MMEA campaigns coagulation was the most important aerosol process for particle removal during low wind speed. The size distributions for LIPIKA and MMEA (green and cyan lines in Fig. 3a) peak in a size range between 10–40 nm diameter and show a higher fraction of Acc particles than the SAPPHIRE distributions. Obviously, coagulation becomes a relevant PN loss process once large numbers of particles below 50 nm diameter from vehicle exhaust emissions (e.g. ca. 92 000 particles cm^{-3} at roadside, LIPIKA) are accompanied by a significant PN fraction of larger particles, which originate either from other local sources or from secondary particle formation within the urban area. These results are in agreement with the ones by Kerminen et al. (2007) who estimated that the lower and upper limits for the inter-modal coagulation time scale during the rush hours were 15–20 and 60–80 min, respectively. During the night, the inter-modal coagulation time scale was 2–3 times that during the rush hours (Kerminen et al., 2007).

The contribution of dry deposition and coagulation to total PN losses is comparable to those determined in previous measurements and model studies. City scale modelling studies with a multi-plume aerosol dynamics and transport model indicated that

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coagulation and dry deposition can cause total PN losses of 15–30 % between roadside measurement and urban background measurement in Copenhagen (Ketzel and Berkowicz, 2005). Gidhagen et al. (2005), using an urban dispersion model that included aerosol dynamics in Stockholm, concluded that in terms of time-averaged PN concentration, dry deposition may yield particle number losses of up to 25 % in certain locations, while coagulation contributed little to PN losses. During particle peak episodes the removal by dry deposition and coagulation was more substantial (Gidhagen et al., 2005).

Condensation and evaporation of vapors as such is not expected to change the total number concentrations, however can modify the particle size distributions and particle volume. In this study a significant increase of PN (by up to 8 % after 30 min travel time) was evident under inefficient dispersion conditions, when condensation was considered in addition to coagulation and dry deposition. The reason could be the competition between condensation and coagulation. As the air parcel moves away from the roadside, condensation of n-alkane vapors leads to rapid growth of small particles to larger diameters at which they are less affected by coagulational loss. In addition, dry deposition velocity decreases with increasing diameter between 1 and 100 nm. Hence particles grown by condensation of n-alkanes will be less affected by deposition.

Aerosol dynamics are less relevant under conditions with efficient dispersion. When efficient dispersion occurs in the urban canopy, dilution by background air is the only effective process reducing PN concentration with distance from the roadside. In such situations (dilution parameters: $a = 80$, $b = 0.9$) aerosol processes account for PN concentration changes of less than 3 % after 10 min and PN concentration changes of less than 6 % after 30 min, hence modelling of PN as passive tracer is adequate. According to the previously published model study at the Dutch motorway A16; the particle size distribution at $D_p > 40$ nm is not further altered by aerosol processes after a distance of 1000 m from the roadside (Keuken et al., 2012). The distance where the PN level reaches background concentrations depends on dispersion conditions. Background PN levels were reached approximately (within an accuracy of ±5 %) after 1740, 900, and

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160 m in distance from the road for inefficient, moderate, and efficient dispersion conditions, respectively, in box model simulations using the Rotterdam campaign data.

3.3 Effect of dry deposition of particles to different surface types

The sensitivity of modelled PN concentrations towards dry deposition of particles on various surface types and roughness conditions were studied in the campaigns Rotterdam TRANSPHORM and UFP-Oslo Tav under moderate dispersion conditions. Two different deposition methodologies, KS2012 and H2012 (detailed description in Sect. 2.3), were applied. Results from the sensitivity tests are summarized in Table S2 in the Supplement.

Between different KS2012 cases, calculated dry deposition velocity, v_d, spanned about one order of magnitude for all particle diameters. Case "KS2012 Urban" corresponded to the surface characteristics of typical urban environments, i.e. streets and buildings, as used for the reference model runs with MAFOR. KS2012 parameterization was not sensitive to changes of friction velocity or roughness length within a typical urban range of values: reducing friction velocity (case "Low friction") or increasing roughness length (case "High roughness") resulted in negligible (≤ 0.1 %) change of PN loss due to dry deposition compared to case "Urban". Over grassland and forest, modelled PN concentration changes due to dry deposition were smaller by 30 and 50 %, respectively, than over urban surfaces.

Using the deposition methodology H2012 for case "Urban" resulted in 40–50 % lower PN losses by dry deposition compared to KS2012. Between different H2012 cases calculated v_d spans about two orders of magnitude for accumulation mode particles (D_p 100–1000 nm) which can be attributed to the fact that surface roughness becomes a dominant factor in collecting aerosol particles efficiently for that particle size range, where neither diffusion nor inertial processes are significant processes. H2012 parameterization was very sensitive to changes of friction velocity or roughness length. The contribution of dry deposition to PN changes varied by roughly a factor of 5 for Rotterdam and by a factor of 3–4 for Oslo due to changing roughness conditions.

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It has been evident in the literature (e.g., Guha, 1997), that surface roughness can increase v_d by up to two orders of magnitudes, in the size range between particle diffusion regime and diffusion-impaction regime, compared to a smooth surface. This behavior is reflected by the H2012 parameterization, but not by the KS2012 parameterization.

3.4 Effect of condensation of n-alkane

The effect of condensation of two n-alkanes with varying gas-phase concentrations on total PN concentration and on the size distribution was tested. Variation of the roadside mixing ratio of n-alkanes between 1–4 ppbv changed the total PN concentration after 30 min travel time by less than 1.55 % at the most, for the campaigns Rotterdam, UFP-Oslo Tav, UFP-Oslo Winter, and Helsinki LIPIKA under moderate dispersion conditions. Therefore, PN concentration can be considered invariant to variations of the concentration of condensable vapors within the above mentioned concentration range, if dispersion is sufficiently efficient.

Inspection of the modelled evolution of number size distributions in simulations of LIPIKA revealed that variation of n-alkane concentration mainly affected the nucleation mode. Roadside n-alkane concentration of 1 and 3 ppbv caused an increase of the nucleation mode diameter from 5 to 5.2 and 7 nm, respectively, in a distance of 240 m from the road compared to a simulation without condensation. Our sensitivity results are qualitatively in line with the study of Pohjola et al. (2007) who, based on measured PN data and the aerosol dynamics model MONO32 (Pirjola et al., 2003), found that the influence of condensation on PN concentrations was negligible on a distance scale of 200 m near a major road in Helsinki. For example, presence of a condensable organic compound with $\sim$ 0.4 ppbv increased the diameter in the two smallest particle size modes by only 14 and 1.9 %, respectively (Pohjola et al., 2007).

An extreme case to test the relevance of condensing n-alkane vapor and its effect on traffic-related size distributions was the Oslo Winter data (Fig. 4a). Under inefficient dispersion conditions, modelled total PN concentration in UFP-Oslo Winter was 2 and 10 % higher after a distance of 240 m with 0.5 and 3 ppbv n-alkane, respec-

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tively, compared to a simulation without condensation. The growth rate of particles with $D_p < 10$ nm was $\sim 12\,\mathrm{nm\,h^{-1}}$ for 0.5 ppbv n-alkane. Two factors enhanced the effect of n-alkane condensation on the changing size distribution: first, the low temperature causing low vapor pressure (a factor of 90 smaller than at 10 °C) and second, the high fraction of initially present particle numbers with diameter below 10 nm. Growth of particles by condensation of n-alkanes caused a shift of the size distribution to larger diameters, thus increasing the survival probability of very small particles. The growth of small particles ($D_p < 10$ nm) to larger sizes within a distance of 240 m in campaign UFP-Oslo Winter corroborates the finding in a curbside study by Zhang et al. (2004) at two freeways in Los Angeles, that a large number of emitted sub-6 nm particles can grow substantially 30–90 m downwind.

3.5 The recommended simplified parametrizations of aerosol processes

As a first step of the implementation of a treatment of aerosol processes in urban air quality models, a separation of PN to various size categories is required. Three particle number concentration (PNC) categories were defined, as follows: PNC_1 (8.5 nm $< D_p <$ 25 nm; "Nucleation mode"), PNC_2 (25 nm $< D_p <$ 100 nm; "Aitken mode"), and PNC_3 (100 nm $< D_p <$ 500 nm; Acc). The upper boundary of 500 nm is justified because the contribution of large particles (defined here as > 500 nm) to total PN concentration from vehicular exhaust is negligible.

A first-order rate law for PNC in the three size categories (index k) was derived for number concentration change with time due to dry deposition:

$$\ln\left(\frac{\mathrm{PNC}_k}{\mathrm{PNC}_{k,0}}\right) = -\frac{\overline{v_{\mathrm{d},k}}}{H_m} \times t \qquad (3)$$

Where $\mathrm{PNC}_{k,0}$ is initial concentration. The average dry deposition velocity $\overline{v_{\mathrm{d},k}}$ was determined by fitting a linear regression model to the time series of modelled PNC_1,

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PNC_2, and PNC_3, from a MAFOR run initialized with the size distribution "mean of traffic sites" (see Sect. 3.1) and dry deposition as only process.

In Eulerian models, dry deposition of particles can be implemented according to:

$$\frac{\mathrm{dPNC}_k}{\mathrm{d}t}|_{\mathrm{depo}} = -\mathrm{PNC}_k \frac{\overline{v_{\mathrm{d},k}}}{H_{\mathrm{grid}}} \quad (4)$$

Here H_{grid} is the depth of the lowest grid level. Table 5 provides average dry deposition velocity derived from the fit to Eq. (3). If applying the parameterization in a Gaussian model then the deposition velocity is usually used to influence the reflection parameter (α) for the reflected plume, e.g. using the following equation (Hanna et al., 1982):

$$\alpha_k(x) = 1 - \frac{2\overline{v_{\mathrm{d},k}}}{v_t + \overline{v_{\mathrm{d},k}} + (Uh - v_t x) \times \sigma_z^{-1} \left(\frac{\mathrm{d}\sigma_z}{\mathrm{d}x}\right)} \quad (5)$$

Where h is the effective plume rise and σ_z is the vertical dispersion coefficient. Gravitational settling velocity v_t in Eq. (5) can be neglected (set to zero) since only particle sizes below 500 nm are relevant for determining PN concentrations.

Coagulation of particles can be implemented, rate according to:

$$\frac{\mathrm{dPNC}_k}{\mathrm{d}t}|_{\mathrm{coag}} = -\mathrm{PNC}_k \times \left(\overline{K_{\mathrm{coag},k}} \times \mathrm{PNC}_k^0\right) \quad (6)$$

Where $\overline{K_{\mathrm{coag},k}}$ (in units $\mathrm{cm}^3\,\mathrm{s}^{-1}$) is the average coagulation coefficient in a size category k derived from MAFOR calculations, provided in Table 5. The expression in Eq. (6) neglects the production terms of coagulation. The superscript 0 indicates the number concentration at the start of the time step calculation in Eulerian models. For Gaussian models this is the calculated concentration before the inclusion of the decay rate due to any physical and chemical processes considered for PNC.

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Dry deposition and coagulation terms are applied separately for the three PNC classes. This means that coagulation between different size categories is not calculated explicitly with the parameterization. However, inter-modal coagulation is partly taken into account through the average coagulation coefficient derived from a model calculation that included coagulation between all size bins. Since the average coagulation coefficient of a given size category depends on the number concentrations in the other size categories, the predicted coagulational loss for PNC_1 and PNC_2 of roadside size distributions that differ from the size distribution "mean of traffic sites" will be somewhat inaccurate.

The accuracy of the presented parameterization for aerosol processes for prediction of PN concentrations is limited by three factors: first, by the averaging of process parameters over a certain size range; second by the simplified treatment of coagulation; and third by neglecting condensation. The uncertainty of the parameterization was studied by comparison with PN concentrations resulting from a detailed aerosol dynamics calculation with MAFOR as reference. For the case "mean of traffic sites", calculated total PNC after 10 min travel time deviated from the reference solution by only 1 %, implying that the error introduced by size-averaged process parameters is negligible. When applying the parameterization to campaign data, the deviation of the total PNC to the reference solution was up to 10 %. UFP-Oslo Winter was excluded from the evaluation due the obvious influence of condensation as shown in Sect. 3.4.

Results of PN concentration modelling for Oslo using the simplified parameterization for dry deposition and coagulation in the Eulerian urban dispersion model EPISODE (Slørdal et al., 2003) are presented in Kukkonen et al. (2015).

4 Conclusions

We have evaluated the significance of aerosol processes during the atmospheric transport of particles on a timescale of one hour, i.e., from the roadside to the neighborhood scale, based on measurement campaigns and modelling in three European ma-

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jor cities. Most of the previous studies have been based on the results of one specific measurement campaign. Our analysis included size distribution data from several campaigns that were performed in different urban settings (street canyon, highway, and suburban main road), exhibiting different traffic characteristics and dispersion conditions, at different times of the year. Monitoring was done with stationary or mobile platforms, and size distributions were measured with various aerosol instruments. An advantage of this study is therefore that the results and conclusions about the relevance of aerosol processes do not depend critically on the specific conditions in terms of emissions, meteorology, and dispersion of a single campaign.

We have used the one-dimensional multicomponent aerosol dynamics model MAFOR to predict PN concentrations and number size distributions. We coupled this one-dimensional model with a simplified treatment of the dilution of particle numbers. Three dispersion cases that are common for northern and central Europe were simulated, ranging from stagnant conditions to efficient dispersion. Despite the simple representation of atmospheric dispersion, size distributions predicted by the aerosol model after approximately 10 min of travel time ($U = 3\,\mathrm{m\,s^{-1}}$) compared well with the size distributions measured at the respective urban background sites.

A limitation of this study was that the chemical transformation of gas phase compounds was not taken into account. It was not necessary to evaluate the nucleation of gas-phase vapors to form new particles, as the model simulations of this study were started at roadside conditions (instead of the exit of the tailpipes of vehicles). It was investigated how condensational growth might influence the shape of the particle size distribution between roadside and the neighborhood scale. Condensational growth did not substantially affect the temporal evolution of the PN concentrations in the presence of efficient and moderate dispersion conditions. The present study shows that growth by condensation can increase the survival probability of very small particles. Condensation removes the smallest particles ($D_p < 15$ nm; Ketzel and Berkowicz, 2004) from the size distribution by growing them to larger sizes, which are less affected by removal through dry deposition and coagulation. However, a significant increase of PN concen-

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tration was found between roadside and the neighborhood scale due to condensational growth under inefficient dispersion conditions. This result differs from that in some previous studies, which stated that the total number concentration between roadside and ambient is not substantially influenced by condensation and evaporation (e.g., Ketzel and Berkowicz, 2004).

It was found that dry deposition and coagulation of particles were generally relevant for PN concentrations on timescales of the neighborhoods. However, as expected, these processes were less relevant in efficient dispersion conditions. The relative relevance of coagulation compared to dry deposition depended on the concentrations of nanometer size particles (< 50 nm).

Coagulation is especially important for the removal of nanoparticles, in this study defined as particles of the sizes 8–25 nm, which accounted for 70 % of the total PN of the mean traffic-related aerosol. The timescale of coagulation is similar to that of dry deposition for the nanoparticles. The typical time scale of dry deposition of particles with 8–25 nm diameter in the urban environment using different deposition schemes was 0.5–3 h. Average dry deposition velocities were in the range of 0.2–0.9 $\mathrm{cm\,s^{-1}}$; this range is similar with the range of 0.6–0.9 $\mathrm{cm\,s^{-1}}$ estimated by Ketzel and Berkowicz (2004). Large differences between the two considered deposition schemes were evident for very rough urban surfaces and for forests. Most of the urban environmental surfaces are rough, and the influence of surface roughness on the dry deposition seems to be pronounced, especially for those particles that are not deposited efficiently by diffusion and inertial processes (Hussein et al., 2012). A future refinement of the dry deposition parameterization should take into account the dependence of the deposition velocity on the underlying urban surface. The lack of measurements of deposition velocities for ultrafine particles to various urban surfaces currently impedes such a refinement.

A simple parameterization of dry deposition and coagulation for urban air quality models was derived. The parameterization of aerosol processes can predict particle number concentrations between roadside and the urban background within an inac-

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curacy of ∼ 10 %. The main source of inaccuracy is the simplified treatment of coagulation, which does not explicitly account for coagulation between size categories. Inclusion of more PN data form other traffic sites and cities might improve the overall accuracy of the parameterization. Potentially, the process of condensational growth might be included in the framework of the current parameterization. However, new particle formation events in the urban background air, frequently associated with a prominent nucleation mode with peak diameter $D_p < 10$ nm (Hussein et al., 2014), probably cannot be sufficiently accurately represented by such a simplified parameterization.

Mitigation policies for ultrafine particle pollution in the future would require the need for operational modelling of PN on urban scales. The presented simplified parameterization can be implemented in both Gaussian and Eulerian models. However, it would be recommendable to evaluate such modelling systems against measured PN data in various urban settings.

Code availability

The computer code of the MAFOR aerosol dynamics model, version 1.8, can be made available upon request (contact: Matthias Karl on email matthias.karl@hzg.de). The code is written in FORTRAN 90.

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Table 1. Meteorological and dilution parameters used in the numerical computations on the evolution of the particle size distribution and PN between roadside and neighborhood time scales. Notation: U = wind speed at a height of 10 m, H_m = initial plume height at the roadside station, a, b = Parameters of the particle dilution parameterization ($y = a \times x^{-b}$, where x is the distance from roadside in meter). The moderate dispersion conditions were used for the reference case.

Dispersion cases	Wind speed	Initial plume	Dilution parameter	
	U [m s^{-1}]	$H_{m,0}$ [m]	a	b
Moderate dispersion	3.0	0.88	40.0	0.5
Efficient dispersion	4.0	0.67	80.0	0.9
Inefficient dispersion	1.0	2.59	20.0	0.2

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Table 2. Dry deposition of particles to urban surfaces. Parameter values used in the modelling of the reference case (all campaigns) and in the sensitivity cases for the dry deposition process. Notation: u^* = friction velocity, z_0 = roughness height, d_{col} = effective collector size, z_C = canopy height, F^+ = effective roughness length. Values for F^+ were adopted from Hussein et al. (2012) for corresponding surface and vegetation types.

Case	Surface type	u^* [cm s^{-1}]	z_0 [m]	d_{col} [cm]	z_C [m]	F^+ [–]
Urban	Street and building	133	0.13	0.20	10.0	0.55
Low friction	Street and building	27	0.13	0.20	10.0	0.55
High roughness	Street and building	133	1.00	0.20	10.0	1.60
Green area without trees	Grassland	36	0.01	0.40	0.20	0.50
Green area with forest	Deciduous forest	75	1.00	1.00	12.0	2.25

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Table 3. Campaign number size distribution data used in this study. Notation: RT = roadside traffic site, ST = street canyon traffic site, UB = urban background, RB = regional background.

City	Campaign / case	Time period	Classification of location	Name of station	Data averaging method	Average total PN [particles cm^{-3}]	References
Rotterdam	TRANSPHORM	6–19 May 2011	Suburban	Bentickplein (ST) Zwartewaalstraat (UB) Cabauw (RB)	Mean $dN/d\log(D_p)$	20 300 14 100 10 200 [a]	This Study
Oslo	UFP-Oslo	12 Dec 2007 – 17 Apr 2008	Suburban	Smestad (RT) Sofienberg park (UB)	Mean $dN/d\log(D_p)$ [b]	24 000 9300	This Study
Helsinki	SAPPHIRE case I	23–28 Aug 2003	Suburban	Highway Itäväylä, Herttoniemi (RT) at 65 m distance Kumpula (UB)	Median $dN/d\log(D_p)$ [c]	32 000 7200	Hussein et al., 2007
Helsinki	SAPPHIRE case II	9–11 Feb 2004	Suburban	Highway Itäväylä, Herttoniemi (RT) at 65 m distance Kumpula (UB)	Median $dN/d\log(D_p)$ [c]	55 100 11 300	Hussein et al., 2007
Helsinki	LIPIKA case I	17 Feb 2003	Suburban	Highway Itäväylä, Herttoniemi (RT) at 65 m distance Saunalahti bay, Herttoniemi (UB)	1 data record (10 min average) Mean $dN/d\log(D_p)$	186 100 13 400	Pirjola et al., 2006 and Pohjola et al., 2007
Helsinki	MMEA	9–11 Feb 2004	Suburban	Mannerheimintie (ST), Herttoniemi (RT) at 0 m (or 8 m) distance Lääkärinkatu (UB)	1 data record (10 min average) Mean $dN/d\log(D_p)$	51 000 (25 800) 13 700	Pirjola et al., 2012

[a] Annual average (2011) at Cabauw.
[b] Weekdays, between 6 a.m. and 3 p.m.
[c] Excluding night-time between 10 p.m. to 6 a.m.

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Table 4. Contribution of aerosol processes coagulation (Coag), dry deposition (Dry dep) and condensation (Cond) to percentage change of PN concentration (%) between roadside station and neighborhood environment after 30 min transport time for different dispersion conditions (i.e. ($\Delta PN_{process}$/(PN(initial) – PN(end))) × 100 %; with $\Delta PN_{process}$ being the change due to aerosol dynamic processes after 30 min).

City and campaign	Efficient dispersion			Moderate dispersion			Inefficient dispersion			All dispersion conditions Range (min – max)		
	Coag	Dry dep	Cond	Coag	Dry dep	Cond	Coag	Dry dep	Cond	Coag	Dry dep	Cond
Rotterdam TRANSPHORM	4.7	0.9	0.0	7.5	4.3	−0.1	12.9	16.6	−0.9	4.7–12.9	0.9–16.6	−0.9–0.0
Oslo UFP-Oslo Tav	0.5	0.4	0.0	0.8	2.6	−0.1	4.1	15.1	−2.9	0.5–4.1	0.4–15.1	−2.9–0.0
Oslo UFP-Oslo Winter	0.5	0.4	0.0	0.8	2.8	−0.1	4.4	16.8	−5.9	0.5–4.4	0.4–16.8	−5.9–0.0
Helsinki SAPPHIRE Case I	0.4	0.3	0.0	0.7	3.2	0.0	4.6	18.3	−4.5	0.4–4.6	0.3–18.3	−4.5–0.0
Helsinki SAPPHIRE Case II	0.7	0.4	0.0	1.2	2.4	0.0	7.3	14.8	−3.0	0.7–7.3	0.4–14.8	−3.0–0.0
Helsinki LIPIKA	1.1	0.2	0.0	2.2	1.0	−0.1	12.5	8.7	−1.1	1.1–12.5	0.2–8.7	−1.1–0.0
Helsinki MMEA	1.3	0.3	0.0	2.2	1.6	−0.1	12.0	9.2	−1.0	1.3–12.0	0.3–9.2	−1.0–0.0
All campaigns Range (min–max)	0.4–4.7	0.2–0.9	0.0	0.7–7.5	1.0–4.3	−0.1–0.0	4.1–12.9	8.7–18.3	−5.9– −0.9	**0.4–12.9**	**0.2–18.3**	**-5.9–0.0**

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Table 5. Data required for the implementation of the PNC parameterization for dry deposition according to three different methodologies and for coagulation. Typical urban times scales for dry deposition (τ_{depo}) and for coagulation (τ_{coag}) is given as reference. MAFOR uses a large number of bin sizes so the extracted coefficients for the three size categories are based on an integral/average over a number of bins in the model. The initial size distribution ratio is the PN fraction in each PNC category for the "mean of traffic sites" distribution. Dry deposition velocity and time scale was calculated with three different methods: KS2012 (Kouznetsov and Sofiev, 2012), H2012 (Hussein et al., 2012), and S1985 (Schack et al., 1985).

Size category	Size ranges [nm]	Initial size distr. ratio [–]	v_d KS2012 [cm s^{-1}]	v_d H2012 [cm s^{-1}]	v_d S1985 [cm s^{-1}]	K_{coag} [cm^3 s^{-1}]	τ_{depo} KS2012 [h]	τ_{depo} H2012 [h]	τ_{depo} S1985 [h]	τ_{coag} [h]
PNC_1	8.5–25	0.70	0.53	0.20	0.87	4.51×10^{-9}	1.1	2.8	0.6	1.9
PNC_2	25–100	0.29	0.12	0.08	0.19	3.10×10^{-9}	4.7	6.7	2.9	6.6
PNC_3	100–500	0.01	0.02	0.07	0.03	8.82×10^{-10}	24	8.5	17	589

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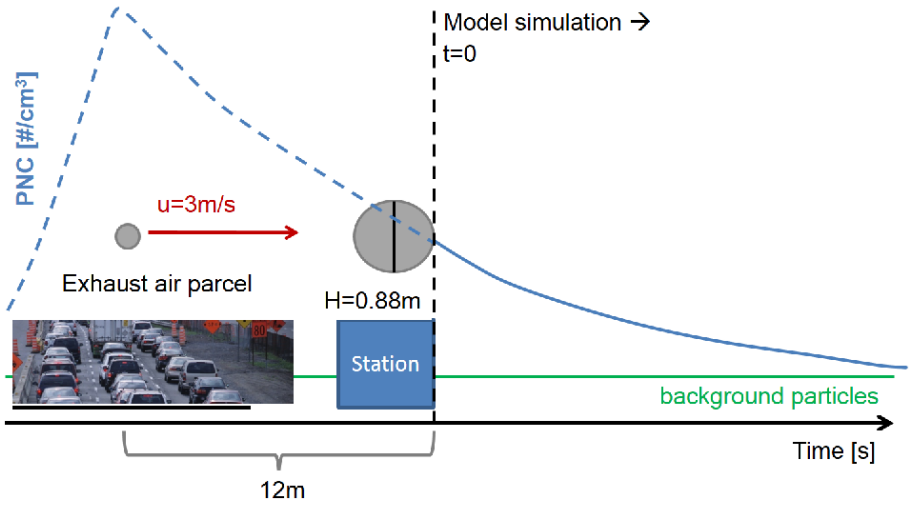

Figure 1. Idealized scenario for model simulations with MAFOR to study aerosol processes between roadside and neighborhood scale. Model simulations start at the point where the exhaust parcel (plume height typically 0.88 m) has approached the roadside traffic station. The simulations are initialized with PN concentration and size distribution measured at roadside. Particle concentrations in the exhaust air are diluted by background air with constant PN concentration.

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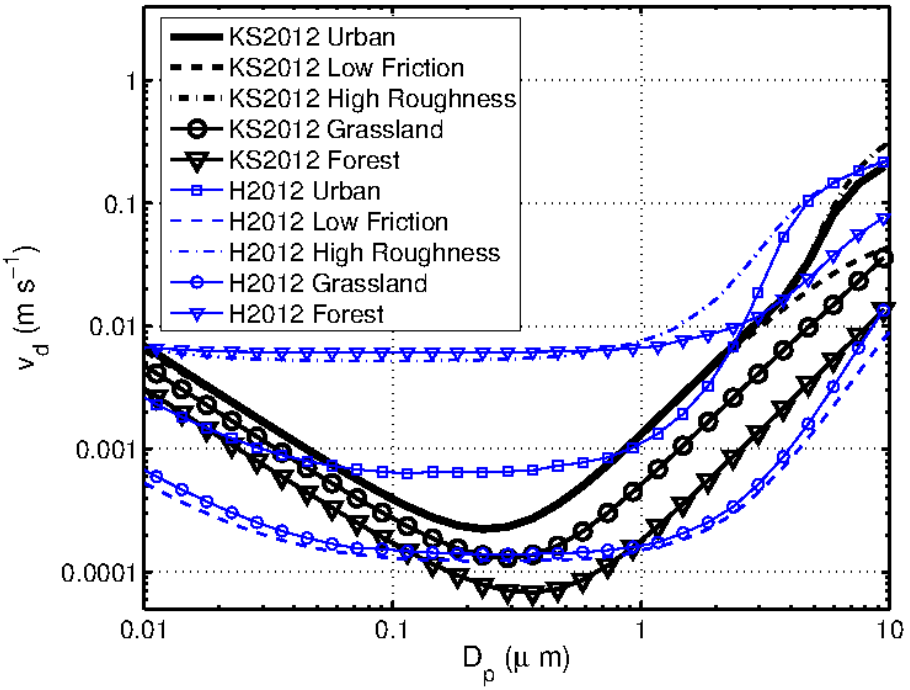

Figure 2. Dry deposition velocity, v_d (in $m\,s^{-1}$) as function of particle diameter D_p (in µm), using a particle density of $1400\,kg\,m^{-3}$. The results with the model of Kouznetsov and Sofiev (2012) (KS2012) are shown as black lines and the results with the model of Hussein et al. (2012) (H2012) are shown as blue lines. The curve of "KS2012 Urban" (thick black line) represents the dry deposition parameterization that is used in all model runs with MAFOR. The curves for cases "KS2012 Low Friction" (dashed black line) and "KS2012 High Roughness" (dash-dotted black line) partly overlay with the curve for "KS2012 Urban".

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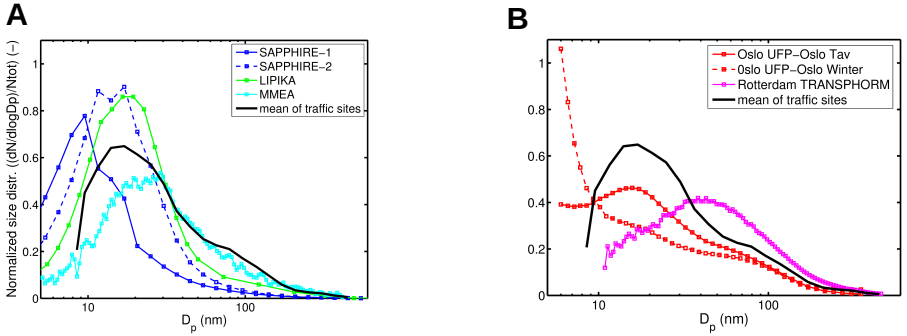

Figure 3. Measured size distribution data normalized to total PN concentration at different traffic stations: **(a)** Helsinki SAPPHIRE Case I, Helsinki SAPPHIRE Case II, Helsinki LIPIKA, Helsinki MMEA, and **(b)** Oslo Smestad Tav case, Oslo Smestad Winter case, Rotterdam Bentinckplein. Urban background concentrations have not been subtracted. The "mean of traffic sites" curve (solid black line) was constructed based on the mean of the size distribution curves for all traffic sites (Bentinckplein, Smestad, Itäväylä, Mannerheimintie) in all campaigns, after synchronization of the size bin diameters. The "mean of traffic sites" curve is displayed in both panels **(a, b)**.

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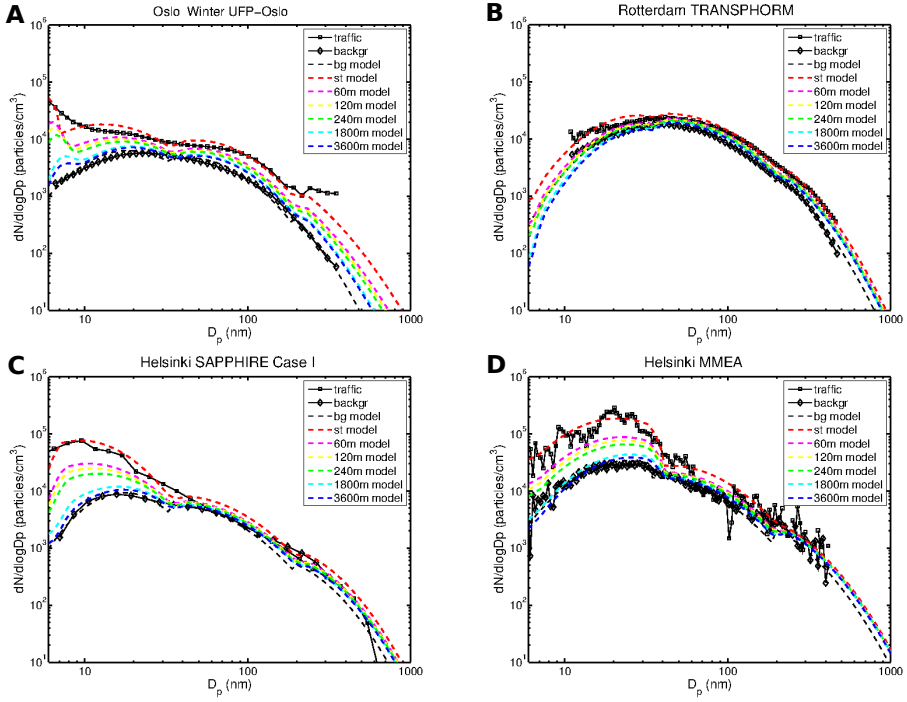

Figure 4. Size distributions ($dN/d\log D_p$ in particles cm^{-3}) downwind of roads in selected campaigns: **(a)** Oslo, UFP-Oslo Winter, **(b)** Rotterdam, **(c)** Helsinki SAPPHIRE Case I, and **(d)** Helsinki MMEA. The plots show the measured distribution at roadside (black squares connected by line), the measured distribution at urban background (black diamonds connected by line), the initial model distribution (roadside: dashed red line, background: dashed black line) and the modelled distributions (resulting for moderate dispersion conditions) at distances of 60, 120, 240, 1800, and 3600 m, respectively. Size distributions are shown with a lower size cut-off at 6 nm.

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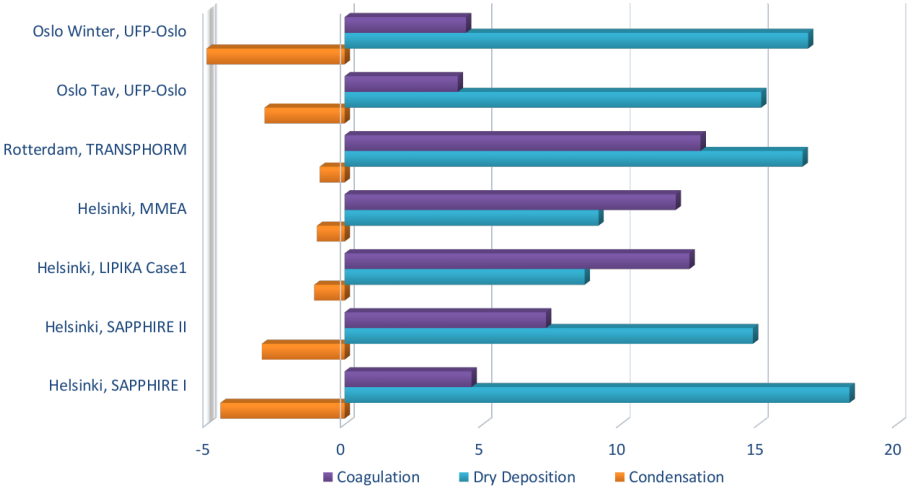

Figure 5. Contribution of aerosol processes to the percentage change of PN concentration (%) between roadside station and neighborhood environment for inefficient dispersion conditions after 30 min transport time.

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