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OUTDOOR URBAN AEROSOLS AND THEIR TRANSPORT AND FATE INDOORS

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Academic dissertation

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Bjarke Mølgaard
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Abstract

Epidemiological and toxicological studies have clearly shown that air pollution has adverse effects on human health. Exposure to air pollution mainly occurs in the urban and in the indoor environment. Fine particles form a diverse group of air pollutants, which is responsible for some of the health effects, and they originate from many outdoor and indoor sources.

This thesis aims to support efforts for reduction of human exposure to fine particles by development and evaluation of an urban forecast model and by evaluation of an indoor aerosol model and of air cleaners.

Measured particle number size distributions were utilised in all studies, and the urban studies additionally utilised meteorological data. The urban aerosol was investigated by using cluster analysis, and the urban particle number concentration forecast model was based on a regression model with an autoregressive structure on its error term. It was evaluated using particle number concentrations from five cities. The utility of a Multi-Compartment Size-resolved Indoor Aerosol Model (MC-SIAM) for estimating airflows in a building was evaluated by comparison with results from a tracer gas technique. The performance of five portable indoor air cleaners was evaluated by applying a simple aerosol model to data from chamber experiments.

The urban concentration of particles smaller than 50 nm was found to depend on local sources, while for larger sizes distant sources may dominate the concentration. The forecast model was found to perform best for locations with a strong influence of local sources. The estimates obtained with the MC-SIAM of airflows between the indoors and outdoors were good for periods during which good agreement between the modelled and measured particle number size distributions was obtained. The indoor airflow estimates deviated somewhat due to a commonly used simplification. Portable air cleaners utilising fans and filters performed as expected, but an ion generator was ineffective for most particle sizes.

The forecast model can give useful forecasts in its present form under certain conditions but further development is desired to make it more versatile. The MC-SIAM is a valuable tool for the study of indoor aerosols except in case of fluctuating airflows. Portable air cleaners are useful for improving indoor air quality, but not all models perform well.

Keywords: urban aerosols, indoor aerosols, particle number size distribution, modelling

Contents

List of publications

This thesis consists of an introductory review, followed by five research articles. These papers are printed with permission of the journals concerned. In the introductory part, the papers are cited according to their roman numerals.

I Mølgaard, B., Hussein, T., Corander, J., Hämeri, K. (2012). Forecasting size-fractionated particle number concentrations in the urban atmosphere. *Atmos. Environ.*, 46:155–163, doi:10.1016/j.atmosenv.2011.10.004.

II Mølgaard, B., Birmili, W., Clifford, S., Massling, A., Eleftheriadis, K., Norman, M., Vratolis, S., Wehner, B., Corander, J., Hämeri, K., Hussein, T. (2013). Evaluation of a statistical forecast model for size-fractionated urban particle number concentrations using data from five European cities. *J. Aerosol Sci.*, 66:96–110, doi:10.1016/j.jaerosci.2013.08.012.

III Mølgaard, B., Ondráček, J, Šťávová, P., Džumbová, L., Barták, M., Hussein, T., Smolík, J. (2014). Migration of aerosol particles inside a two-zone apartment with natural ventilation: A multi-zone validation of the multi-compartment and size-resolved indoor aerosol model. *Indoor Built Environ.*, In press, doi:10.1177/1420326X13481484.

IV Mølgaard, B, Koivisto, A.J., Hussein, T., Hämeri, K. (2014) A New Clean Air Delivery Rate Test Applied to Five Portable Indoor Air Cleaners. *Aerosol Sci. Tech.*, In press.

V Hussein, T., Mølgaard, B., Hannuniemi, H., Martikainen, J., Järvi, L., Wegner, T., Ripamonti, G., Weber, S., Vesala, T., Hämeri, K. (2014): Fingerprints of the urban particle number size distribution in Helsinki, Finland: Local versus regional characteristics. *Boreal Env. Res.*, In press.

1 Introduction

This dissertation is motivated by the health effects of air pollution. People are exposed to air pollution especially in the indoor and urban environments. The high population density in cities results in a high density of pollution sources, such as, cars, combustion for domestic heating, industry, power plants, harbours, and airports. Due to ventilation pollutants in the outdoor air also penetrate into buildings and thus affect the indoor air quality. Additional indoor air pollution may originate from cooking, cleaning, smoking, domestic heating, furniture, candles, electronics, mould, and building materials. Many of these outdoor and indoor sources are relatively new, but others, such as biomass combustion for cooking and domestic heating, have existed for millennia. Biomass combustion is still the main air pollutant source in many societies in developing countries (Kodgule and Salvi, 2012).

In December 1952 London experienced an extreme air pollution episode which caused thousands of deaths (Bell and Davis, 2001). Reports of a few earlier severe episodes elsewhere exist, but this one took place in a large city rather than some small town in a valley. Perhaps for this reason, or because of the large number deaths, especially this London smog episode stimulated the study of air pollution epidemiology. In the following decades epidemiological studies investigated not only the acute, but also the chronic effects of air pollution. Nowadays, the epidemiological studies have been supplemented by a large number of toxicological studies which have investigated the mechanisms through which the pollution affects human health.

When deposited in the airways and lungs, air pollution may cause inflammation, oxidative stress, and weaken the defence against infections. Through these mechanisms the pollution may cause, or at least increase the risk of, asthma, chronic obstructive pulmonary disease, various infections (e.g. pneumonia) and lung cancer (Loomis et al., 2013; Arbex et al., 2012; Smith et al., 2000). The effect of air pollution is not limited to the respiratory system. There is strong epidemiological evidence that it also affects the cardiovascular and the central nervous system (Rückerl et al., 2011), and possible mechanisms have also been suggested. Araujo and Nel (2009) summarised three possible pathways for the effect on the cardiovascular system. Firstly, pollution may through the respiratory system affect the autonomic nervous system which controls the heart. Secondly, air pollution may also cause inflammation and oxidative stress, which may lead to atheroschlerosis; a chronic inflammatory disease in the blood vessels. This may eventually cause a heart attack or a stroke. Various particles and trace gases may affect through these two pathways. Thirdly, it has been suggested that ultrafine particles (UFP, diameter < 100 nm) may cross the barrier from the lung to the blood

stream and cause harm virtually anywhere in the body. This translocation of particles has been observed in animals, but it is unclear whether it happens in humans (Watkins et al., 2013; Oberdörster et al., 2005). This third pathway leads us to the discussion of how the toxicity depends on particle size. Part of the size dependence is caused by different deposition probabilities in airways and lungs. When inhaled, coarse particles are likely to deposit in the airways due to impaction. Thus, only a small fraction reaches deep into the lungs. Fine particles, on the other hand, are likely to penetrate into lungs and many of them reach the alveolar region. Ultrafine particles have a high deposition probability due to fast Brownian motion, while particles with diameters of a few hundreds of nanometres often exit the lungs with the exhaled air. It is debated which particle size-fraction is most dangerous (Kelly and Fussell, 2012). In general, ultrafine particles dominate the number concentration while coarse particles dominate the mass concentration. A few epidemiological studies have investigated which particle size fraction is more dangerous (Iskandar et al., 2012; Franck et al., 2011; Breitner et al., 2011; Andersen et al., 2010, 2008; Stölzel et al., 2007; Osunsanya et al., 2001), and their conclusions depend on the studied health effects and the pollutant mixture in the city considered. Generally, it seems that mainly the UFP are responsible for cardiovascular effects, while larger particles cause most of the respiratory effects, although the results of Andersen et al. (2008) deviates from this trend. They found that both cardiovascular and respiratory diseases in elderly were mostly affected by PM_{10} (mass concentration of particles with aerodynamic diameter < 10 μm), and that paediatric asthma was more affect by fine particles and NO_x. In general, the results of these studies are somewhat uncertain. Because of large UFP concentration gradients measured concentrations in one or a few locations may not be representative for the whole city, and it is therefore hard to estimate the general UFP exposure. Another difficulty in epidemiological studies is that the effect of particle size cannot be separated from the effect of differences in particle composition.

The 1952 London episode was characterised by emissions of particulate matter and sulphur dioxide from coal burning in industry and for domestic heating. This resulted in high concentrations of SO_2 and soot particles. To avoid similar episodes domestic coal burning was in the following decades abandoned in London and elsewhere in western countries. In the recent decades the main concern has been the vehicular emissions, which comprise nitrogen oxides, CO, SO_2, and various particles. The gaseous compounds and most of the particles smaller than 200 nm originate from the tailpipe emissions. These particles comprise mainly soot and organic carbon, but they also contain traces of many metals (Vouitsis et al., 2009; Geller et al., 2006). Some of these particles (soot) are emitted directly, while others (organic carbon, sulphuric acid) are formed during the cooling of the exhaust (Kittelson et al., 2006). Depending on

driving conditions, wear of tires, brakes, and roads may produce particles in the whole size range from a few nanometres to tens of micrometres (Wahlström et al., 2012; Kukutschová et al., 2011; Mathissen et al., 2011). Moreover, moving cars suspend road dust into the air (Pirjola et al., 2010; Hussein et al., 2008a). However, road traffic is not the only relevant source of urban air pollution. In cities with cold winters the effect of biomass burning for domestic heating is important (Saarikoski et al., 2008). Industrial emissions affect the air quality substantially in some cities (Minguillón et al., 2013; El Haddad et al., 2011). In coastal cities considerable amounts of pollution may originate from ships, especially when the wind is dominated by a sea breeze (González et al., 2011). In general, weather conditions affect pollutant concentrations strongly by affecting sources, deposition, chemical reactions, and pollutant transport (Jones et al., 2010; Seinfeld and Pandis, 2006; Mathis et al., 2005; Chate and Pranesha, 2004).

The pollutants in the urban atmosphere also affect the indoor air quality, because buildings continuously exchange air with the outdoor environment. Some of the particles in the incoming air are lost due to deposition on surfaces along the path through which the air enters the building, while others penetrate into the indoor environment (Mosley et al., 2001). Particles are lost due to deposition indoors as well (Hussein et al., 2009; Lai and Nazaroff, 2000). For these reasons the concentration is lower indoors than outdoors in absence of indoor sources. However, indoor sources are common. Emission rates on the order of 10^{10} particles/s have been observed for cooking activities, cigarette burning, incense burning, candle burning, and vacuum cleaning, use of sprays, and laser printing (Géhin et al., 2008; Evans et al., 2008; Hussein et al., 2006a; Afshari et al., 2005; He et al., 2004). The concentration resulting from these emissions depends on their duration, the volume of the indoor environment, the ventilation rate, and sinks, such as deposition. The concentration may easily reach 10^5 particles/cm^3 during cooking activities (Hussein et al., 2006a; He et al., 2004).

The risk for adverse health effects can be reduced by reducing exposure to pollution. The exposure happens both indoors and outdoors as illustrated in Figure 1. Most people spend most of their time indoors so the indoor exposure is important (Hussein et al., 2012; Schweizer et al., 2007; Kousa et al., 2002). The outdoor exposure may also be substantial, especially for people who spend time close to major roads. Each individual can to a limited extent reduce his or her own exposure, but the society can also reduce the exposure for the entire urban population through regulations. The European Union regulates emissions and outdoor concentrations of a number of air pollutants (Directive 2008/50/EC). The member states are required to do what is needed to keep concentrations below the limits. Moreover, forecasts of the regulated pollutants need to be made available to the public, so that individuals can take action to avoid exposure to high concentrations. Aerosol particle concentrations are regulated

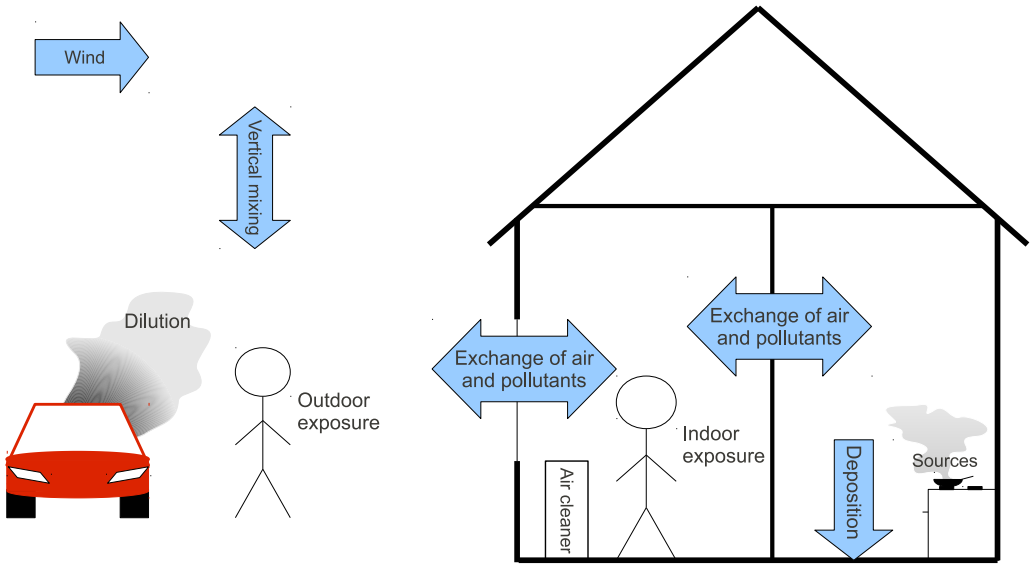

Figure 1: Illustration of the most important processes related to the urban population exposure to particles.

through limits on the particle mass concentrations PM_{10} and $PM_{2.5}$, and on the lead content of these particles. The particle number concentration is not regulated, although the ultrafine particles only contribute to a small fraction of $PM_{2.5}$ even at high number concentrations.

Efforts to reduce the pollutant concentrations outdoors also contribute to getting cleaner indoor air. Exposure to air pollution indoors can further be reduced through regulations on ventilation systems and on building materials. Individuals can reduce their exposure in their own homes by removing pollution sources, ventilating, and by using air cleaners. During the recent decades awareness towards the health effects of tobacco smoke has increased, and smoking is forbidden in many indoor environments. However, other sources of indoor air pollution receive much less attention, and large parts of the population may be unaware of them. Because of the focus on energy efficiency, new buildings are more tight than old ones. Thus, the ventilation rate may get low in new buildings if the mechanical ventilation is absent, and pollutants may accumulate in the indoor air. Indoor air cleaners can improve the indoor air quality, but they should not replace ventilation, because air cleaners only remove part of the pollution.

In summary, the health effects of air pollution are adverse, both the outdoor and the indoor exposures are important, and the regulated mass concentrations are not enough for quantifying the toxicity of aerosols. This dissertation focuses on particle number concentrations and particle number size distributions of fine particles outdoors as well as indoors. There is a lack of forecast models for urban particle number concentrations and size distribution. In the development of such models knowledge of the sources is useful. For indoor aerosols models exist, but these require knowledge of the outdoor particle number size distribution and of several input parameters, such as indoor airflows. These input parameters can be estimated by applying the indoor aerosol model to measured particle data, but the reliability of such estimates is hard to assess. The effect of an air cleaner can easily be handled in indoor aerosol models provided that its Clean Air Delivery Rate (CADR) is known. Many manufacturers fail to give this information. The lack of the CADR information is also a problem for the consumer, who wants a good air cleaner.

The aims of the dissertation are:
- the development and evaluation of a forecast model for size-fractionated urban particle number concentrations (**Papers I** and **II**),
- the investigation of the urban background aerosol in Helsinki including the assessment of sources (**Paper V**),
- the evaluation of the utility of an indoor aerosol model for estimating airflows in a building (**Paper III**), and
- the quantification of size-resolved CADR and general evaluation of the performance of portable air cleaners for indoor use (**Paper IV**).

2 Background

An aerosol comprises a gas (or a mixture of gases) and solid or liquid particles suspended in this gas. The air we breathe is an aerosol which contains particles from various sources. These particles are very diverse in size, shape, and chemical composition, but the information available from aerosol measurements is usually limited to a few characteristics.

2.1 Aerosol measurements

The particle number concentration can be measured with a Condensation Particle Counter (CPC). A CPC detects particles optically after making the particles grow by condensation of a vapour (e.g. buthanol). The detection efficiency is high for particles in a wide size range (Yli-Ojanperä et al., 2012).

To get more information on the aerosol one can measure its particle size distribution. There are various instruments for this purpose. The particle size distributions used in this thesis were measured with Differential Mobility Particle Sizers (DMPS), Scanning Mobility Particle Sizers (SMPS), an Optical Particle Sizer (OPS), and an aerodynamic particle sizer (APS). The DMPS and the SMPS (Wiedensohler et al., 2012; Aalto et al., 2001) consist of the same three main three parts. First the aerosol passes through a bipolar charger (also known as a neutraliser) in which some radioactive material provides ionising radiation. Because of this there are many air ions and the aerosol quickly reaches the equilibrium charge distribution, which is a known particle size dependent charge distribution. Then the aerosol passes through a Differential Mobility Analyser (DMA) in which an electric field causes the trajectories of charged particles to deviate from the airflow. Particles with electrical mobilities in a certain range are selected and their number concentration is measured with a CPC. The electrical mobility is the product of the charge and the mechanical mobility, which depends on particle size and shape. The chosen electrical mobility depends on the electric field in the DMA, so by scanning over a range of DMA voltages it is possible to obtain the concentrations of particles with various electrical mobilities. Based on these data and knowledge of the flows, air viscosity, and other parameters the particle number size distribution can be derived. In the DMPS the DMA voltage is changed in discrete steps, and in the SMPS the DMA voltage change is continuous. These instruments measure the particle number size distribution in the sub-micrometer range only.

To obtain the particle number size distribution for larger particles as well an OPS was used in **Paper IV** and an APS was used in **Paper III**. These instruments both detect

particles optically. The OPS (Heim et al., 2008) irradiates the particles and estimates the size of particles based on the amount of scattered light, which also depends on particle composition and orientation. This was not a problem in our application, where most of the particles larger than 300 nm were spherical and consisted of the same material. In an APS (Peters and Leith, 2003) particle sizes are distinguished based on their inertia. When the air flow is accelerated, heavy particles are accelerated less than lighter ones. Thus, the particles will for some time have different velocities, which are determined by passing the particles through two laser beams and recording the time between the peaks of scattered light. The APS gives the particle number size distribution in terms of the aerodynamic diameter which can easily be converted to the geometric diameter for spherical test particles of known density.

2.2 Aerosol dynamics and modelling

The particle size distribution in an air parcel is affected by many processes. Direct emissions and new particle formation increase the concentration, and deposition decreases the concentration. Coagulation decreases the number concentration without affecting the mass concentration of the aerosol. Condensation and evaporation of vapours, on the other hand, change the particle mass concentration without affecting the number concentration.

When a particle hits a surface it usually gets attached to it. This process is known as deposition. There are several ways in which a particle can get in contact with a surface. For coarse particles gravitational settling and impaction are important (Guha, 2008; Hinds, 1999). Impaction happens when the inertia of a particle prevents it from following the streamlines of the air in which it is suspended and thereby causes it to hit a surface. In the other end of the size scale, for UFP eddy and Brownian diffusion are responsible for most of the deposition. Brownian diffusion is only effective at small length scales, but turbulent mixing (eddy diffusion) brings particles into the region in which Brownian diffusion affects (Lai and Nazaroff, 2000). For particles in the accumulation mode the mentioned deposition mechanisms are ineffective. Interception happens when a streamline brings a particle so close to a surface that the particle touches the surface (Hinds, 1999). A special case of deposition is wet deposition which happens when raindrops or snowflakes remove particles from the atmosphere (Paramonov et al., 2011; Chate and Pranesha, 2004; Laakso et al., 2003). Deposited particles may be resuspended into the air. The force causing the resuspension may originate from the wind or from an object in contact with the surface on which the particle was deposited (Boor et al., 2013).

Coagulation happens when particles collide and stick to each other. The reason for collision is usually Brownian motion or gravitational settling. Particle collision usually involve particles of rather different sizes, because small particles have fast Brownian motion and large particles have a large cross-section (Yu et al., 2013). If the particles contain enough liquid, the resulting particle will be spherical, but if the particles are solid the result is a particle of irregular shape. In the latter case the coagulation is also known as agglomeration.

Particle sizes change due to condensation and evaporation of vapours. Water is an important example due to its abundance in the atmosphere, but there are several other compounds, mostly organics, which may condense to or evaporate from particles. The concentrations of these compounds may change due to emissions or chemical reactions, and saturation ratios change when the temperature changes. In case of sufficiently high supersaturation of one or more compounds new particle formation occurs. This may happen during the rapid cooling of an exhaust plume from a car (Kittelson et al., 2006) or due to photochemical production of non-volatile compounds such as sulphuric acid (Kulmala et al., 2013; Sipilä et al., 2010; Kulmala et al., 2004). New particle formation is also common indoors where non-volatile products of reactions between terpenes (e.g. from cleaning agents) and ozone often are the cause (Coleman et al., 2008; Vartiainen et al., 2006). Particles formed in the atmosphere are called secondary particles as opposed primary particles originating from direct emissions.

So far only processes which take place in an air parcel were described. However, when assessing the particle size distribution in some location, indoors or outdoors, transport mechanisms should be taken into account. This brings a lot of complexity to the aerosol dynamics, especially outdoors. On large scales, one needs to consider the wind driven horizontal transport and the vertical transport due to turbulent mixing. This turbulent mixing depends on the wind and the stability of the atmosphere. At smaller scales, buildings affect the air movement and thereby also the transport of particles. In street canyons the air at street level may move in the direction which is opposite to the overall wind direction (Olivares et al., 2007).

2.2.1 Urban aerosol modelling

Because of the complex transport mechanisms and the general lack of knowledge of input parameters, dispersion models for particle number concentrations or size distributions are hard to implement on urban scales. Nevertheless, a few implementations exist. Gidhagen et al. (2005) implemented a city-scale Eulerian model with 500 m horizontal resolution to data from Stockholm. Ketzel and Berkowicz (2005) modelled

the evolution of the particle number size distribution from the sources to a roof top location in Copenhagen using a multi-plume Lagrangian model, and Roldin et al. (2011) developed a Lagrangian model for urban plume studies.

Because dispersion models require detailed information on weather conditions and pollutant emissions, an alternative approach is often used. Statistical models have been used in a large number of studies of urban air pollution. In general, these models are easy to implement and they can run on normal desktop computers. For the implementation of statistical models, sufficiently large data sets of good quality are needed. Statistical models have been used for estimating the effect of various weather parameters on pollutant concentrations (Clifford et al., 2011; Hussein et al., 2006b; Aldrin and Haff, 2005), and for assessing temporal trends (Anttila and Tuovinen, 2010; Carslaw et al., 2007). The information extracted from measured data can be used for various purposes. Many studies seek the underlying physical reasons for the extracted dependences on co-variates, and new knowledge on the pollution sources or the relevance of various atmospheric processes may be obtained. The information can also be used for forecasting of future pollutant concentrations. Many models for the forecast of gaseous pollutants and particle mass concentrations exist (Moustris et al., 2012, 2010; Cobourn, 2010; Chelani and Devotta, 2006; Goyal et al., 2006; Pérez et al., 2000), but there is a lack of forecast models for urban particle number concentrations.

2.2.2 Indoor aerosol modelling

In the simplest cases the indoor air is assumed to be well-mixed, and the only transport mechanism to consider is the air exchange with the outdoors (Chao et al., 2003; Long et al., 2001). Usually, this treatment is too simplistic, especially when walls divide the indoor environment into separate rooms. Instead one can then divide the indoor space into a number of zones and assume that the air in each of these is well mixed (Hussein and Kulmala, 2008; Sohn et al., 2007; Hussein et al., 2005; Miller and Nazaroff, 2001). In this case the time derivative of number concentration $N_{k,i}$ of particles in size section i in zone $k \in \{1, 2, ..n\}$ is described by the balance equation

$$\begin{aligned}\frac{dN_{k,i}}{dt} =& \frac{1}{V_k}\sum_{j=0}^{n}(Q_{jk}P_{jk,i}N_{j,i} - Q_{kj}N_{k,i}) \\ &+ S_{k,i} - \beta_{k,i}N_{k,i} + J_{coag,k,i} + J_{cond/ev,k,i},\end{aligned} \tag{1}$$

where V_k is the volume of zone k, Q_{jk} is the air flow rate from zone j to k, $P_{jk,i}$ is the penetration factor for particles of size section i following this flow, index $j = 0$ refers to the outdoors, $S_{k,i}$ is a particle source (primary or secondary) or sink in compartment k, β is the deposition rate, J_{coag} is the change rates due to coagulation, and $J_{cond/ev}$

is the change due to condensation and evaporation. The latter two terms depend on the concentrations in other size sections. If all parameters on the right hand side are known, it is simple to calculate the evolution of the particle number size distribution in each of the zones. However, this is rarely, if ever, the case for real buildings. Therefore, it is common to make assumptions about some of the parameters, and to ignore one or more of the terms. Often this or a similar model is applied in the analysis of measured particle number size distributions in order to estimate some of the parameters on the right hand side (Hussein et al., 2011, 2006a, 2005; Chao et al., 2003; Long et al., 2001; Vette et al., 2001). When the assumption of well-mixed air in each of the zones is invalid, one can instead use computational fluid dynamics (CFD) to describe the transport of pollutants (Liu and Zhai, 2007). However, when using CFD detailed knowledge of the building in question is needed.

3 Methods

In all the five studies we used experimental data. Table 1 lists the devices used for particle measurement and the size ranges of the detected particles. Additionally, we used supporting data including meteorological parameters, vehicle counts, and indoor tracer gas concentrations.

3.1 Urban forecast model

In **Paper I** we developed a forecast model for size-fractionated particle number concentrations in the urban atmosphere. We chose the statistical approach because of its simplicity and because of lack of spatial data. Particle number concentrations and size distributions are measured continuously in only one location in Helsinki, and this is too little for testing dispersion models which are spatial. Spatial modelling would otherwise be desirable, because the general exposure of the urban population to the particles is unlikely to correlate well with the measurements in just one location. However, a model for forecasting the particle number concentration in one point is a good first approach. For cities with measurements in several locations it can be applied to data from each measurement station, or perhaps it can serve as the first step in the development of a spatial model.

We used a model of the following form (**Papers I** and **II**):

$$\log(N) = g(T, U, RH, Tr, \varphi) + f(t) + \varepsilon, \tag{2}$$

where N is the particle number concentration for some size section, T is the temperature, U is the wind speed, RH is the relative humidity, as the traffic parameter Tr we used the hourly vehicle count at a major road, φ is the wind direction, t is time, g and f are parametric functions, and ε is the error term.

Table 1: Size ranges of particles detected with instruments which provided data for this dissertation.

Device	Lower detection limit	Upper detection limit	Used in Papers
CPC	7 nm	A few μm	**II**
DMPS	3 – 6 nm	700 – 950 nm	**I**, **II**, **V**
SMPS	10 – 20 nm	300 – 760 nm	**II**, **III**, **IV**
OPS	300 nm	10 μm	**IV**
APS	540 nm	20 μm	**III**

Although this model, like statistical models in general, includes no equations for the aerosol dynamics, the aerosol dynamics was considered in its development, because it helped us choosing the covariates and the structure of the model. In general, the particle number concentration depends on sources and weather conditions. Vehicular traffic is a very important source of particles and it varies widely in time. At urban locations the concentration is generally lower at high wind speeds U, because the wind removes particles from the city (Jones et al., 2010; Hussein et al., 2006b). The dilution due to vertical mixing also has an important effect on the concentration, but the vertical mixing is hard to quantify. However, it is associated with the temperature T, and this is one of the reasons that a dependence on temperature is often observed. Another reason is that some sources are temperature dependent. There is evidence that the relative humidity RH has an effect or is correlated with something that has an effect on secondary particle formation in the boreal forest (Hyvönen et al., 2005), and our data suggested that RH could have an effect in Helsinki as well. The wind direction φ is also important, because of the heterogeneity of the surroundings. When the wind carries particles from a source the concentration in the plume quickly decreases due to dilution, deposition, and coagulation (Pohjola et al., 2007; Gidhagen et al., 2005). Thus, higher concentrations are measured when the wind comes from directions where strong sources are nearby. At different wind directions there are different mixtures of sources and surface properties, which affect the deposition and vertical mixing. Therefore it is possible that the dependence on other covariates (Tr, T, U, RH) depends on the wind direction (Hussein et al., 2006b). Our choice of g (see equations 5 and 6 in **Paper II**) allows for such a wind dependence, and thereby our model differs from many other models which treat the wind direction dependence in a simpler way (Clifford et al., 2011; Aldrin and Haff, 2005).

To avoid that the parameters for temperature dependence would just describe the difference between summer and winter, parameters for the seasonal dependence was included in the function f as seen in Equation 4 of **Paper I**. The parameters Tr, T, U, RH, and φ do not explain all of the diurnal variation. Part of the reason is that the vertical mixing has diurnal cycle and peaks in the afternoon. This cycle depends on the season, so along with parameters for the dependence on time of day we included parameters for the combined effect of time of day and time of year in f. Also a linear trend was included, because long term changes in pollutant concentrations are common (Anttila and Tuovinen, 2010).

As most other atmospheric parameters particle number concentrations are strongly autocorrelated, so the error term ε in Equation 2 is autocorrelated. We defined another error term u which is related to ε through Equation 4 in **Paper II** (at one hour resolution). This error term u had negligible autocorrelation and it was assumed to

be normally distributed. Therefore the likelihood function in Equation 7 of **Paper I** equalled the following product $\prod_i \frac{1}{\sigma\sqrt{2\pi}} \exp(-u_i^2/2\sigma^2)$, where u is calculated based on the data and parameters using the mentioned equations. We applied Bayes' law and a simple non-informative prior to obtain the posterior distribution of the model parameters, which was needed for producing forecasts (details in section 2.1.3 of **Paper I**). When testing the model the first year of each data set was used as learning data only, and forecasts were produced for the rest in a sequential manner. The forecast were made for one day at a time, assuming that all data was available until noon on the previous day.

3.2 Investigation of the urban aerosol

Half-hour median particle number size distributions were divided into seven groups of similar distributions by the aid of cluster analysis (**Paper V**). We used the clustering procedure described by Beddows et al. (2009). Moreover, we classified the days according to the following criteria: Clear/unclear traffic influence, new particle formation event/non-event/undetermined (Hussein et al., 2008b), and long-range transport/short-range transport/no transport episode. By comparison of the occurrence of clusters and these classifications information of the origin of particles at various sizes was derived.

3.3 Indoor aerosol models

The models used in **Papers III** and **IV** are both simplifications of the model in Equation 1. In both studies we ignored condensation and evaporation. In agreement with previous studies (Hussein et al., 2006a, 2005; Miller and Nazaroff, 2001) we additionally assumed in **Paper III** that the penetration factors for the internal flows were one ($P_{jk,i} = 1, j \neq 0$). Furthermore, $S_{k,i}$ was assumed to be zero, except around the time of particle injection. We simulated the indoor particle number size distributions using many different values of the model input parameters. The values of these input parameters were estimated by comparing simulated and measured particle number size distributions. The quality of the airflow estimates was assessed by comparison with results from modelling of tracer gases.

In **Paper IV** we tested air cleaners in an 81 m^3 emission chamber. The model was simplified to

$$\frac{dN_i}{dt} = \lambda N_{vent,i} - (\lambda + \beta_i + \gamma_i)N_i + J_{coag,i}, \quad (3)$$

where $N_{vent,i}$ is the concentration in the air coming in from the ventilation duct, λ is the ventilation rate and γ_i was the cleaning rate. There were three unknowns (λ, β, and γ) so we needed three equations to find them. The two additional equations were obtained by further simplification of the equation above. During part of the experiment the air cleaner was off so $\gamma = 0$ during this period. During another period $N_{vent,i}$ was close to zero. After obtaining the cleaning rate γ we multiplied it with the volume of the chamber to get the CADR.

4 Results

The coefficient of determination (R^2) can be used as a measure of forecast accuracy. In **Paper I** we applied the model to a data set from Helsinki and for hourly median concentrations the R^2 values were 0.63 for UFP and 0.52 for accumulation mode particles. At three hour resolution the values were 0.67 and 0.57, respectively. R^2 values obtained with the same model in **Paper II** are given in Table 2.

In **Paper III** the reliability of the airflow estimates obtained with the aerosol model was assessed by comparison with estimates based on tracer gas measurement and modelling. A summary of the results is given in Table 3.

The size-resolved CADR values in Figure 5 of **Paper IV** are the main result of that paper. At the chosen setings, the three air cleaners which relied on filtration (F1, F2, and F3) had CADR above 80 m^3/h for the whole size range, and the CADR of the electrostatic precipitator (ESP) ranged from 60 to 90 m^3/h. For the ion generator (IG) the CADR was below 50 m^3/h for particles with diameters above 100 nm, but for UFP the CADR was higher and reached up to 140 m^3/h for particles with diameters around 30 nm.

In **Paper V** we used cluster analysis for grouping similar particle number size distributions together. For each of the obtained clusters we assessed the origin of the particles. One cluster included a particle mode with geometric mean diameter of 5 nm. Particle size distributions belonging to this mode was mostly seen on days with new particle formation. Two clusters with median diameters between 10 and 30 nm were attributed to fresh traffic emissions. A cluster with median diameter around 40 nm was mainly observed when the wind came from the centre of Helsinki and is most likely dominated by aged traffic emissions. Two clusters with median diameters above 50 nm were the result of an elevated concentration of particles originating from outside the city. Finally, one cluster had particles from a mixture of sources.

Table 2: Coefficient of determination (R^2) for one day in advance forecasts of hourly mean particle number concentrations. D_p is the particle diameter. Results from **Paper II**.

	Size section		
Site	$D_p < 100\,\text{nm}$	$D_p > 100\,\text{nm}$	$D_p > 7\,\text{nm}$
Helsinki	0.60	0.51	-
Stockholm, Street	-	-	0.70
Stockholm, Background	-	-	0.52
Copenhagen	0.40	0.48	-
Leipzig	0.39	0.26	-
Athens	0.49	0.43	-

Table 3: Airflows [m^3/h] obtained by aerosol modelling and by tracer gas modelling.

Scenario (Experiments)	Method	Q_{01}	Q_{10}	Q_{02}	Q_{20}	Q_{12}	Q_{21}
I (1–2)	Aerosol	2.9–14	1.3–13	1.3–2.5	1.0–3.4	0–2.2	0–1.7
I (1–2)	Tracer gas	10–12	9.4–11	-0.6–1.1	0.6–1.5	1.3–2.5	0.3–1.8
II (6–9)	Aerosol	3.1	1.3	1.5	3.3	11	9.3
II (6–9)	Tracer gas	1.9–2.3	1.0-5.0	0.7–1.4	-1.6–2.4	7.6–15	10–14
III (18–19)	Aerosol	6.6–7.5	5.7–6.4	33–43	34–44	0.9–5.3	0–4.2
III (18–19)	Tracer gas	4.5–13	8.3–19	29–30	24–26	3.5–5.9	9.5–9.6
IV (17,20)	Aerosol	75–92	75–96	2.2–4.3	0.3–1.6	0.3-0.4	0.9–4.4
IV (17,20)	Tracer gas	70–75	68–75	-0.8–2.4	1.0–2.2	0.7–2.2	0.4–0.8

5 Discussion

The model in **Paper I** was developed for forecasting particle number concentrations in the urban background. The forecast accuracy was good, especially for UFP the R^2 was high. Presumably the difference in the results for the two size sections is caused by the fact that the origin of the accumulation mode particles is further away. When testing the model with data measured in four other cities (Stockholm, Copenhagen, Leipzig, and Athens; **Paper II**), we obtained results which were generally not as good (Table 2). This was to some extent anticipated because our model was developed using a data set from Helsinki and only parameters which improved the performance for this data set were selected. During our work with **Paper I** we got an idea for how to optimise the model based on the Deviance Information Criterion (DIC, Spiegelhalter et al., 2002). As mentioned in **Paper I** and its supplementary material, we tested the model with a variety of parametrisations, and when calculating the DIC based on a fit to the first year of the data set (learning data), we found that the best forecast accuracy was obtained for the parametrisations which gave the lowest DIC values. The idea of the DIC (and other information criteria) is to estimate whether parameters improve a model, so this also made sense from a theoretical point of view. An algorithm which calculated the DIC for a lot of parametrisations and chose the parametrisation with the lowest DIC value was implemented. To make a faster version of this algorithm, the DIC was also replaced with the Akaike Information Criterion (AIC, Akaike, 1974). However, this optimisation procedure only caused small changes in the forecast model performance relative to the differences between performances at various sites and did not in general improve it (**Paper II**). The reason that our model performed better for Helsinki (in terms of R^2) is that there the concentration depends more strongly on local weather and traffic. The surroundings of the measurement location are very heterogeneous (Järvi et al., 2009), and at certain wind directions the sampled aerosol is strongly affected by the emissions at the nearby road. Therefore, the measured UFP concentration is likely to be poorly correlated with the urban background concentration elsewhere in Helsinki, and it is a poor measure of the concentration the urban population is exposed to. In Copenhagen the measurements were performed at a location even closer to a major road than in Helsinki, but there the inlet was on a roof top 20 m above ground level. Due to the wind the horizontal transport of particles is usually much faster than the vertical. Therefore the measurement station in Copenhagen is a background station despite the proximity of a major road. The urban background concentrations in Stockholm and Leipzig were also measured at roof top level. The sub-urban background station in Athens was located in a vegetated area, and the traffic emissions were found to only have a minor influence on the aerosol at this site. The concentrations measured at this site are not suitable for estimating the exposure of the population, because in most

other locations the effect of traffic emissions is expected to be much stronger. Although people generally spend little time on roofs, the roof level concentrations measured in Copenhagen, Stockholm and Leipzig are much more relevant for exposure estimations, because they are likely to be well correlated with concentrations in other locations which are not directly affected by nearby particle sources (Costabile et al., 2009), and mechanically ventilated buildings may have ventilation inlets on the roof. The particle concentrations on streets are also highly relevant for exposure assessments, but there the concentration gradients are substantial. Therefore, the forecast of concentrations in one location in a street canyon has little value even if the forecast is fairly accurate as it was for Hornsgatan in Stockholm (**Paper II**). Perhaps the model could anyway be useful for the forecast of concentrations at streets. By removing the effect of the wind direction one could obtain forecasts which represent the overall concentration close to the street better. If this would be done for a few street locations in a city, the overall exposure in the streets is likely to correlate well with the mean of these forecasts. However, some research would be needed before putting this idea to practice.

As described in Section 3.1 the choice of the structure of the statistical model in **Paper I** was to a large extent based physical understanding of the aerosol. In **Paper V** we improved our understanding of the aerosol in Helsinki. With respect to model development the important lesson learned is that particles smaller than 50 nm mainly originate from local sources and thus their concentration is likely to be well described by local parameters, while particles transported from outside of the city often dominate the concentration of particles larger than 50 nm. For these particles it may be useful to include information on the history of air masses in the model (Cobourn, 2010), but our model in **Papers I** and **II** only included local parameters. Therefore, it was expected to perform better for ultrafine particles than for larger ones, and the results in **Paper II** were generally in agreement with this expectation. Despite the distant origin of many accumulation mode particles, the forecast performance was decent for this size section, except in Leipzig. Likely reasons are that the model contains parameters for the seasonal variation, its autoregressive part includes lags up to one week, and the local wind direction and air temperature are associated with the air mass origin.

As people spend most of their time indoors, for exposure assessment it is important to understand to what extent particles are transported from outdoors to indoors, how particles migrate indoors, and how quickly they are removed. Aerosol models are a useful tool for this purpose. Airflows, penetration factors, deposition rates, and emission rates are critical parameters, which are often unknown. Several studies have used indoor aerosol models along with particle measurements to estimate some or all of these parameters (Hussein et al., 2011, 2006a, 2005; Chao et al., 2003; Long et al., 2001; Vette et al., 2001). In **Paper III** we assessed the reliability of such

estimates by comparing the obtained airflows to airflow estimates based on tracer gas measurements. We used data from a measurement campaign in a naturally ventilated apartment, which comprised two rooms. We found that the airflow estimates obtained with the two methods generally agreed, although there were a few problems. When using the aerosol model the internal flows (Q_{12} and Q_{21}) were often underestimated (Table 3). The reason is most likely that we used the common simplification of setting the penetration factor for particles following the internal flows to one (Hussein et al., 2006a, 2005; Miller and Nazaroff, 2001). There were also periods during which it was impossible to obtain good simulations of the aerosol with reasonable model input parameters, and thus reliable airflows could not be obtained with the aerosol model. Variable airflows are a likely reason, because when the ventilation is natural it depends strongly on weather conditions. Because this method only gives good airflow estimates for some periods, the use of tracer gases is preferable. When reliable airflow values are available, it is easier to estimate the other input parameters. After the typical values of all input parameters have been obtained, the model can be used to predict indoor concentrations at various (forecasted) outdoor concentrations and in presence/absence of various indoor sources.

Air cleaning systems are often used, because buildings are often surrounded by polluted urban air and indoor sources are hard to avoid. In buildings where the incoming air enters through designated ventilation ducts, it is common to install filters in these ducts. However, in many buildings the air enters through leaks and cannot be cleaned while entering the building. In these buildings portable air cleaners can be a good alternative. These air cleaners also remove pollutants originating from indoor sources. In **Paper IV** we tested five portable air cleaners and quantified their ability to remove particles in terms of the Clean Air Delivery Rate (CADR), which is a standard measure of air cleaner performance. The idea is that with respect to each pollutant the effect of the air cleaner is equivalent to injecting clean air at some rate. The CADR is a parameter which can easily be used in indoor aerosol models like the MC-SIAM (Hussein et al., 2005, **Paper III)**. This is done by adding the term $-\frac{CADR_i}{V_k}N_{k,i}$ into Equation 1 for the zone k in which the air cleaner is placed.

We found that four of the air cleaners were effective for all particle sizes (Figure 5 of **Paper IV**). The remaining air cleaner (IG) was only effective for ultrafine particles. For all air cleaners except IG, the CADR could be adjusted by changing the fan speed. The usage of portable air cleaners and other systems, which clean and recirculate the indoor air, is sensible when the outdoor air is polluted and when there is a need to maintain a temperature difference between indoors and outdoors, because in these cases high ventilation rates are not a good solution. Especially for people with allergies it makes sense to clean the indoor air, no matter if the allergen source is indoors or outdoors. Air

cleaners also have a few drawbacks, such as noise, electricity demand, and maintenance requirements. Moreover, electrostatic precipitators and ion generators often produce ozone, and filters emit volatile organic compounds (Schleibinger and Rüden, 1999). Especially dusty filters can reduce the air quality (Bekö, 2009).

6 Review of papers and the author's contribution

Paper I presents the developed particle number concentration forecast model and its implementation in detail. Furthermore, it presents forecast model performance measures. Results from the implementation with a data set from Helsinki are reported. I wrote most of the article and had the main responsibility for the development of the model, and I wrote the algorithms.

Paper II investigates the performance of the model developed in **Paper I** when applying it to data from five European cities. It also presents and investigates the performance of two procedures for automatic optimisation of the model. I collected and handled data, and I wrote the algorithms and most of the article.

Paper III investigates the utility of the MC-SIAM for estimating the airflows between indoor and outdoors and between indoor zones by comparing such estimates with estimates based on tracer gas measurements. I did most of the work with the aerosol model and improved the tracer gas analysis. I wrote substantial parts of the paper, including most of the results and discussion section.

Paper IV evaluates the performance of five portable indoor air cleaners. Particle size resolved Clean Air Delivery Rates are quantified and presented, and the general performance is discussed. I analysed data and wrote most of the paper.

Paper V investigates the urban background aerosol in Helsinki and its origin based on meteorological data, traffic data, and particle number size distribution measured in Helsinki and at a rural background station. I worked with the cluster analysis including the interpretation of the clusters and wrote a minor part of the article.

7 Conclusions

The human exposure to particulate air pollution, which may cause a variety of diseases and even death, mainly happens in the urban and in the indoor environment. The indoor exposure is important because people tend to spend most of their time indoors. A substantial fraction of the indoor exposure is to particles originating from the outdoors. When adding this to the outdoor exposure the importance of outdoor air quality becomes evident.

The particle number concentration outdoors depends on weather conditions and on sources nearby and further away. In Helsinki, most of the particles with diameters below 50 nm originate from sources within the city, while a substantial fraction of the larger particles come from further away. Therefore, it was no surprise that our forecast model performed somewhat better for ultrafine than for accumulation mode particles. In general, the model provides more accurate forecasts for concentrations which depend strongly on the covariates used in the model. Especially for concentrations strongly affected by traffic emissions the performance was good. The relevance of the forecasts depends on how well the forecasted concentrations correlate with the exposure. Both of these issues should be considered when assessing the utility of the model in a given location. Preferably, the model should be used to forecast concentrations in several locations in a city, but that requires continuous measurements in each of these locations.

Indoor aerosol models are useful for estimating the indoor exposure provided that their input parameters and the outdoor concentration are known. Often the input parameters are estimated using the model and particle measurements. This method was evaluated and good estimates of the airflows were obtained for periods during which the indoor aerosol could be simulated well. The indoor airflows were however somewhat underestimated, because the balance equation was simplified by setting the penetration factor between the zones to unity.

Five portable air cleaners were tested. Four of them used fans for blowing the air through filters or a combination of filters and an electrostatic precipitator. These four were all found to be effective in a wide size range. Under normal conditions, any of these can easily reduce the particle concentration in a small flat to less than half. In contrast, an ion generator with no fan or filters was ineffective except for ultrafine particles. With the obtained CADR values, the effect of an air cleaner can easily be estimated using an indoor aerosol model.

The forecast model can be used to obtain urban size-fractionated particle number concentrations one or a few days in advance. If the building specific input parameters

are available, the indoor aerosol model can use the outdoor forecast for forecasting indoor concentrations.

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