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# OBSERVATIONS OF URBAN AEROSOLS IN INDIA

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Academic dissertation

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Petteri Mönkkönen

# Observations of Urban Aerosols in India

Juha Petteri Mönkkönen
University of Helsinki, 2005

## Abstract

Continuous and campaign based aerosol field measurements in different environments are essential in understanding fundamental atmospheric aerosols processes and evaluating the effect of the aerosols on global and regional climate, environment and human lives. This thesis investigates the formation and growth dynamics, characteristics of the aerosol number size distribution and seasonal and diurnal concentrations and variations of atmospheric aerosols in a highly polluted Asian mega city.

The experimental results indicate that the formation and growth of atmospheric aerosols is possible in this kind of environment even though the smallest growing particles face effective scavenging by the pre-existing aerosol population. The formation and growth of atmospheric aerosols in the nucleation mode particles can be explained by high source rates of condensable vapours which was found to vary up to four orders of magnitude between the most polluted (New Delhi) and cleanest sites (Finnish Lapland and Antarctica) considered in this study. However, the average growth rates of nucleation mode particles increased only 1-2 orders of magnitude between different sites. The primary reason for this is that large vapour source rates usually appear in concert with large condensation sinks hence balancing the condensable vapour concentrations.

The characteristic of the aerosol number size distribution (3-800 nm), number concentration and the analysis of diurnal modal parameters suggest that there are at least two major sources in New Delhi. Beside the traffic, another source in the evenings increased the geometric mean diameter of the detected particles. It is concluded that vehicular emissions together with biomass and/or refuse burning has significant contribution to New Delhi's atmosphere. This analysis is also supported by the indoor number concentration measurements conducted in urban private houses in the Indian cities of Nagpur and Mysore.

High seasonal and diurnal number and mass concentrations averages, substantial pollution events and the use of fossil fuels in cooking deteriorate ambient air quality in India. Continuous research activities also in other developing countries are needed to gain better scientific understanding not only in evaluating Earth's radiative balance but also in designing technological inventions providing better living conditions for inhabitants in these countries.

## Contents

## List of publications

This thesis consists of an introductory review part, followed by five research articles. Papers are reprinted with the kind permission from Elsevier (I, III and V) and European Geosciences Union (II and IV).

**I:** Mönkkönen, P., Uma, R., Srinivasan, D., Koponen, I. K., Lehtinen, K.E.J., Suresh, R., Sharma, V. P. and Kulmala, M. Relationship and variations of aerosol number and $PM_{10}$ mass concentrations in a highly polluted urban environment – New Delhi, India. *Atmospheric Environment*, 36, 425-433, 2004

**II:** Mönkkönen, P., Koponen, I.K., Lehtinen, K.E.J, Hämeri, K., Uma, R., and Kulmala, M. Measurements in a highly polluted Asian mega city: Observations of aerosol number size distribution, modal parameters and nucleation events. *Atmospheric Chemistry and Physics*, 5, 57-66, SRef-ID: 1680-7324/acp/20055-57, 2005.

**III:** Mönkkönen, P., Koponen, I.K., Lehtinen, K.E.J, Uma, R., Srinivasan, D., Hämeri, K. and Kulmala, M. Death of nucleation and Aitken mode particles: Observations at extreme atmospheric conditions and their theoretical explanation, *Journal of Aerosol Science*, 35, 781-787, 2004.

**IV:** Kulmala, M., Petäjä, T., Mönkkönen, P., Koponen, I.K., Dal Maso, M., Aalto, P.P., Lehtinen, K.E.J., Kerminen, V.-M. On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments. *Atmospheric Chemistry and Physics Discussions*, p. 6943-6966, 2004.

**V:** Mönkkönen, P., Pai, P., Maynard, A., Lehtinen, K.E.J., Hämeri, K., Rechkemmer, P., Ramachandran, G., Prasad, B. and Kulmala, M. Fine particle number and mass concentration measurements in urban Indian households. *Science of The Total Environment*. In press, 2005.

# 1. Introduction

Atmospheric aerosols are extremely interesting research field. Aerosols give challenges for physicists, chemists, meteorologists, epidemiologists, biologists, engineers, mathematicians and for policy makers. Researchers are interested for example on aerosol formation, growth, scavenging, size, shape, concentration, size distribution, chemical composition, life time, production and transport to understand better their contribution to our everyday life, ranging from health effects to climate change.

Aerosols are divided into two broad categories based on their method of formation: primary aerosols and secondary aerosols. Primary aerosols are solid or liquid particles in a gaseous media that are emitted directly into the atmosphere by natural (e.g. wind suspension) or anthropogenic (e.g. burning of fossil fuels) processes. Secondary aerosols are solid or liquid particles created in the atmosphere through gas-to-particle conversion (e.g. homogeneous nucleation) which involves chemical or physical transformations of precursor gases (Seinfeld and Pandis, 1998).

Besides the chemical composition of aerosols, one the most important physical property of aerosols is their size. The size of an aerosol varies from about one nanometer (e.g. a molecular cluster) to hundreds of micrometers (e.g. a dust particle) in diameter. Mass concentration ($PM_{10}$ or $PM_{2.5}$, particle size less than 10 or 2.5 micrometers, respectively) is a measure that defines the mass of the particles in certain volume. The most typical unit used is micrograms per cubic meter ($\mu gm^{-3}$). Dividing particles into different size classes and weighing the classes, researcher can derive mass size distribution of the particles. As a result of developed measuring techniques, aerosol number concentration (number of particles per cubic centimetre, $cm^{-3}$) and number size distribution measurements have become possible research groups in the field.

Continuous and campaign based aerosol field measurements are important in many ways. Firstly, aerosols influence the earth's radiation balance by scattering or absorbing solar radiation. Aerosols scatter solar radiation back to space, thus enhancing the planetary albedo and exerting a negative (cooling) climate forcing. Aerosols also indirectly cause a negative climate forcing through formation of clouds and affecting cloud life time. On the other hand, aerosols cause positive (warming) climate forcing by preventing reflected radiation from earth's surface to escape back to space and by absorbing (aerosols containing BC) solar radiation. The total albedo caused by the aerosols is one of the most uncertain questions in climate change studies (Houghton et al., 2001). Secondly, understanding human exposure to aerosols and health effects caused by poor ambient air is important to enable exposure control and reduction. Several studies have shown the association of aerosols with increased mortality and hospital admissions due to respiratory and cardiovascular diseases (e.g. Brunekreef and Holgate, 2002). In developing countries, indoor air quality deteriorates rapidly because of the use of solid fuels in cooking. In India alone, it has been estimated that the use of biomass fuels causes 400,000-550,000 premature deaths annually among women and children under five because their household roles (Smith, 2000).

In both of the cases the number concentration and the number size distribution are two most important factors. Scattering and absorption of solar radiation are strongly based on the diameter of particles. Recent findings in health related studies have shown that the number concentration of ultrafine particles ($D_p < 100$ nm) could be much better predictor and indicator of the health effects caused by particle matter than the mass concentration (Donaldson et al., 2001, Peters et al., 1997). Aerosol number size distribution measurements in the ambient air are also important for example while investigating particle lung deposition, designing indoor ventilation and filtration systems or developing less polluting diesel engines.

Besides aerosol field measurements, this study is also based on theories explaining: (1) essential atmospheric processes i.e. formation, growth and scavenging of aerosols through nucleation, condensation and coagulation; (2) modal structures (nucleation, Aitken and accumulation mode) of submicron ($D_p$ < 1000 nm) aerosol number size distribution; and (3) modal parameters (total concentration, geometric mean diameter and geometrical standard deviation) of the size distribution.

The aim of this thesis was to improve our understanding of urban air quality and characteristics of aerosol dynamics in a highly polluted urban environment. For this purpose, New Delhi provided an ideal environment to complete this study, since New Delhi is considered to be one of the most polluted cities in the world. The main objectives were:

- to find out the seasonal, weekly and diurnal concentrations and variations of aerosol number and mass concentrations and, if possible, to find out a relationship between the number and mass concentration (Paper I).
- to study diurnal indoor number and mass concentrations, I/O-number concentration relationship and relationship between the indoor number and mass concentration (Paper V).
- to find out characteristics of the aerosol number size distribution, formation rate for 3 nm-size particles, growth rate of the particles and source rate of condensable vapours (Paper II and III)
- to compare the formation and growth rates of the particles and source rates of condensable vapours between clean (Antarctica, Finnish Lapland and Boreal Forest) and polluted urban (Athens, Marseille and New Delhi) environments (Paper IV).

# 2. Urban aerosols in India

Chapter two presents a short review of the past and recent aerosol measurements conducted in India. The review is focused on the physical properties of urban aerosols. A review published by The Energy and Resources Institute (TERI, 2001) summarizes the chemical characterisation of particulate matter and other pollutants.

## 2.1 Particulate mass concentration

Under the Air Prevention and Control of Pollution Act, 1981, the Central Pollution Control Board (CPCB) initiated National Ambient Air Quality Monitoring (NAAQM) programme in the year 1984 and this programme was later renamed as the National Air Monitoring Programme (NAMP). Over the years, the number of stations has increased and presently, the network comprises 290 stations spread over 92 cities and towns distributed over 24 states and 4 Union Territories. In addition to the NAMP, many State Boards have set up ambient air quality monitoring stations under their own programme known as Ambient Air Quality Monitoring (AAQM) programme. National Environmental Engineering Research Institute (NEERI) monitors ambient air quality in 30 stations covering 10 major cities. In addition to the monitoring stations, operated by the Central/State Boards and Research Organisations, major industries and research institutes like The Energy and Research Institute (TERI, former Tata Energy Research Institute) have set up monitoring stations as part of the compliance of the consent conditions (CPCB, 2000, TERI, 2001).

Beside national monitoring network, several research organizations have their own research programmes. It is still difficult to state, however, whether any city has been adequately studied. In comparative terms, Agra, Calcutta, Delhi and Mumbai have the most studied ones among Indian cities and towns. Even though fairly extensive monitoring network the pollutants monitored should be reconsidered. Instead of monitoring Total Suspended Particles (TSP, $D_p < 100$ μm) levels which are widely acknowledged not to be important from the point of view of public health impact, smaller particles such as $PM_{10}$ ($D_p < 10$ μm) and $PM_{2.5}$ ($D_p < 2.5$ μm) should be monitored regularly. In India, the concept Suspended Particulate Matter (SPM) is used to describe TSP (TERI, 2001).

SPM and $PM_{10}$ levels are relatively high in all Indian metropolitan cities. The trend in SPM annual average value indicated upward movement up to 1995 in residential areas and thereafter declined because of various measures for air quality improvement, but in industrial areas the trend has been fluctuating very widely due to industrial emission generation as well as enforcement by regulatory bodies. The trend of SPM in residential areas has been fluctuating in most cities like Calcutta, Bhopal, Nagpur, Patna, Ponhcherry and Bangalore. The downward trend has been observed in residential areas of Jaipur, Mumbai, Ahmedabad and Howrah. Maximum SPM values have been recorded at Delhi, Calcutta and Kanpur. The lowest values have been observed in South Indian cities e.g. Bangalore, Hyderabad and Chennai (CPCB, 2000).

SPM levels are high in New Delhi and exceed many times the National Ambient Air Quality Standards (NAAQS). SPM was monitored at six locations for the period 1989 to 2000. The annual average concentrations of SPM ranged between 255 $\mu gm^{-3}$ and 443 $\mu gm^{-3}$ at locations in residential areas and between 282 $\mu gm^{-3}$ to 510 $\mu gm^{-3}$ at locations in industrial areas. The NAAQS levels for SPM in India are 140 $\mu gm^{-3}$ (annual average) and 200 $\mu gm^{-3}$ (24-h average) in the residential area and 360 $\mu gm^{-3}$ and 500 $\mu gm^{-3}$ in the industrial area (CPCB, 2001).

The continuous measurement of $PM_{10}$ in New Delhi conducted by CPCB started in March 1998. Monitoring of $PM_{10}$ is carried out for 24 hours with 8 hourly sampling. $PM_{10}$ is measured gravimetrically with GFA/EPM 2000 filter paper using respirable dust samplers. (CPCB, 2001). The variation of SPM and $PM_{10}$ mass concentrations between March 1998 and December 2000 at B.S.Z. Marg measuring station are presented in Figure 1. During this period, the maximum amount of $PM_{10}$ out of SPM was 69 % and minimum was 26 %, respectively.

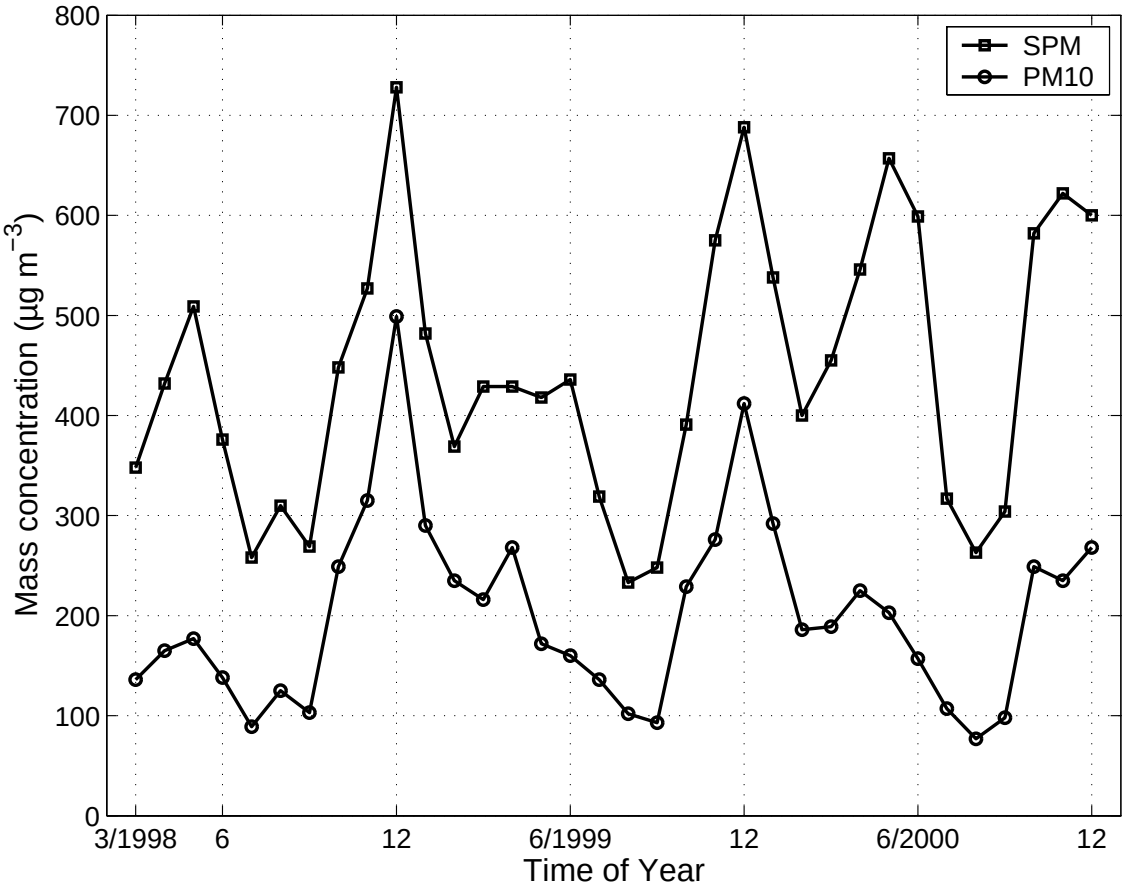


Figure 1. The monthly mass concentration (µgm$^{-3}$) of SPM and $PM_{10}$ at B.S.Z. Marg in New Delhi between 3/1998 and 12/2000. Data from CPCB (2001).

**2.2 Mass size distribution**

Only few studies have examined aerosol mass size distributions in India. These studies have been mostly of short duration and hence seasonal variations on mass size distribution are not documented. Mass size distribution measurements have been conducted in New Delhi (Gadi et al., 2000, Balachandran et al., 2000), Mumbai (Sharma and Patil, 1991), Agra (Kulshrestha et al., 1998, Parmar et al., 2001) and in Pune (Maheskumar et al., 2001, Momin et al., 1999) and Thiruvananthapuram (Momin et al., 1999). Studies conducted in New Delhi, Agra, Pune and Thiruvananthapuram used an eight or nine stage Andersen impactor for the particle sizes 0.43-10.0 µm. Parmar et al. (2001) used a four-stage cascade impactor (CPS-105) which fractionates particles in the sizes ranging between 0.7 and > 10.9 µm. In Mumbai a Quartz Crystal Microbalance Cascade Impactor (QCM-CI, Model PC-2E) with the size range 0.05-25.0 µm was used.

The results in all these cities indicated bimodal mass size distribution. The peak in the fine mode in New Delhi and Agra was close to 1 μm and another peak in the coarse mode (5 μm < $D_p$ < 6 μm). In Mumbai the peaks were found close to 0.1 μm and 2.0 μm and a third peak in the industrial site was found close to 0.05 μm. The main purpose of these studies and the studies conducted in Mumbai by Tripathi et al. (1999 and 2004) and Venkataraman et al. (1999 and 2001) was to define the chemical mass size distribution.

### 2.3 Number concentration

The aerosol number concentration in the measuring site in Mumbai was defined by Sharma and Patil (1992) from the observed mass size distribution assuming a spherical shape for all the particles. They also approximated the number size distribution using the Junge distribution. Their results indicated that the number concentration in the Aitken nuclei range (< 100 nm) varies considerably with location; however it remained almost the same in the coarse size range.

During the Indian Ocean Experiment (INDOEX) campaign Murugavel and Kamra (1999) used an Electrical Aerosol Analyzer (EAA, TSI Inc.) at a coastal station in Thiruvananthapuram to measure the number concentration and number size distribution of aerosols in the size-range of 3 nm to 1 μm. They found two peak hours in the number concentration. The morning peak hour was at about 7.30 a.m. and the evening peak hours at 7-9 p.m. The number concentrations were also higher in the evenings. Diurnal 30 minutes mean concentrations varied form 2,500 $cm^{-3}$ (2 p.m.) to 29,000 $cm^{-3}$ (9 p.m.).

Similar diurnal patterns were also detected in New Delhi (Paper I) and Nagpur (Paper V), but the concentrations observed in New Delhi and Nagpur were much higher since they were urban sites. Figure 2 presents the diurnal outdoor number concentration measured during March and April in New Delhi and Nagpur 2002. Clear diurnal pattern during morning (6-8 a.m.) and evening (5-10 p.m.) in both cities were observed. The 24 hours average number concentration in the measuring site in New Delhi was more than two times higher both in March and April. In this study, also diurnal indoor number concentrations ($cm^{-3}$) in urban Indian households have been presented for the first time (Paper V).

The outdoor number concentration values in New Delhi were higher compared for example to European city Athens. The concentration levels in New Delhi varied from 20,000 to 200,000 $cm^{-3}$ during the day and in Athens from 2,000 $cm^{-3}$ to 90,000 $cm^{-3}$, respectively. In clean environments like Finnish Lapland, concentrations smaller than 100 $cm^{-3}$ can be detected.

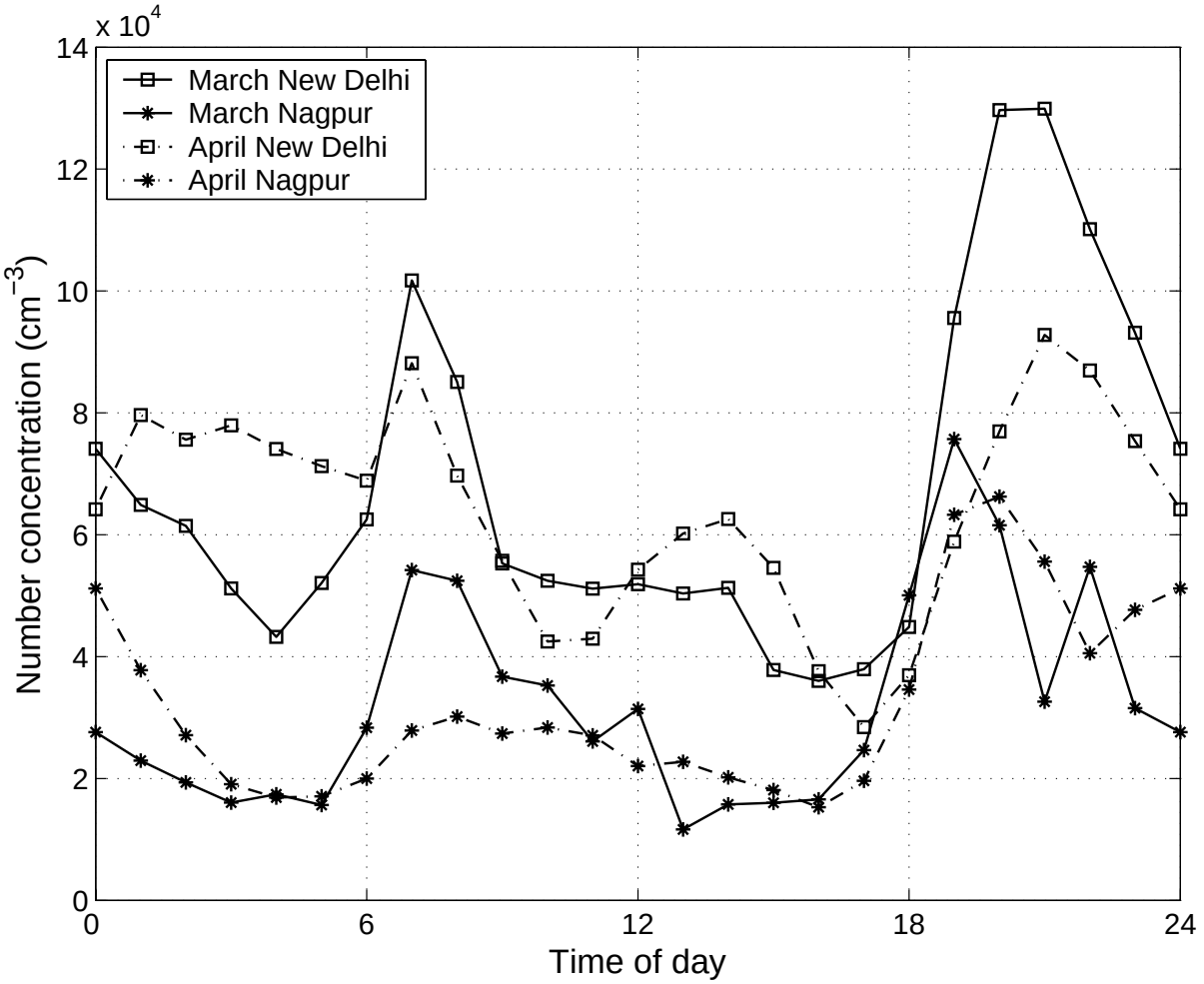


Figure 2. Diurnal one hour mean outdoor number concentration ($cm^{-3}$) measured during March and April in New Delhi and Nagpur 2002.

**2.4 Number size distribution**

The Indian Ocean Experiment (INDOEX) conducted above the Indian Ocean was the most ambitious research project so far in India in this field. A primary goal of INDOEX was to quantify the direct and the indirect aerosol forcing from observations. The INDOEX observations were made over the tropical Indian Ocean during the Northern Hemisphere dry monsoon (December to April). Since 1996, an international group of scientists has been collecting aerosol, chemical and radiation data from ships, satellites and surface stations. This culminated in a major field experiment with five aircraft, two ships, several satellites and numerous surface observations conducted during January through March, 1999 (Ramanathan et al., 2001). During INDOEX field campaigns aerosol number size distributions were measured on ships and aircrafts (e.g. Kamra et al., 2003 and de Reus et al., 2001).

Before this study, only few aerosol number size distribution measurements with various methods at inland and coastal stations have been performed. Murugavel and Kamra (1999) defined the size distribution by Electrical Aerosol Analyzer (TSI Inc.) in the size range 3-1000 nm at coastal station in Thiruvananthapuram. Maheskumar et al. (2001) retrieved number the size distribution in the size range 80-4000 nm from a multi-channel solar radiometer in an urban station in Pune. Badarinath et al. (2004) calculated the number size distribution of aerosols caused by biomass burning in

Mizoram (Northeast) by using inversion of spectral variation of aerosol optical depth data. Madhavi Lata et al., (2003) derived the number size distribution from mass size distribution measured by Quartz Crystal Microbalance for the particles in the size range 260-4,000 nm by assuming a spherical shape for all the particles with the density of 2 g $cm^{-3}$ in Hyderabad and Anantapur. Aerosol number size distribution (3-800 nm) data presented in this study (Paper II and III) was measured in India Habitat Centre (IHC/TERI) in New Delhi from 26$^{th}$ October to 9$^{th}$ November 2002. The measuring site was located next to a traffic line in a residential area at the altitude of 15 meters.

Figure 3 presents the number size distribution (a), number concentration (b) and selected 1 h mean number size distribution (c and d) in the size range 3-800 nm measured in New Delhi Nov. 6$^{th}$ 2002. This size range has been divided into three different modes: Nucleation mode (particle size range 3-25 nm), Aitken mode (25-100nm) and accumulation mode (100-800 nm). The figure illustrates a typical day during the measuring period in New Delhi in October and November 2002 (Paper II). From Fig. 3a and 3c we can see that the accumulation mode particles were dominant right after midnight until 5 a.m. After 5 a.m. The number concentration increased clearly after 6 a.m. from 38,000 $cm^{-3}$ to 240,000 $cm^{-3}$ (8 a.m.) This increase can be explained by the increase of number concentration in Aitken mode (Fig. 3c). In Fig. 3c the Aitken mode was dominant already between 6 and 7 a.m. and became very dominant between 8 and 9 a.m. After 8 a.m. the number concentration decreased back to 60,000 $cm^{-3}$ before 11 a.m. At the same time the size distribution returned to a similar type as measured in the morning (6-7 a.m.). Aitken mode particles remained dominant the whole day, but towards the evening (9 p.m.) the number concentration of accumulation mode particles increased. The analysis and discussion of the diurnal pattern of the number concentration and number size distribution in New Delhi is presented in Papers I-III.

### 2.5 Chemical characterisation

The quantity of the published work related to the chemical characterisation is overwhelming compared to the physical characterisation of the particulate matter in India. However, most of the chemical studies are concentrated on heavy metals, particularly lead. Beside vehicular sources, incineration has been cited as an important source of lead. Lead concentrations have been observed to decrease only along the dense traffic corridors and not necessarily in other parts of urban areas. Also re-suspension of dust is an important source of airborne lead. The concentrations of other elements have been found to be correlated with the characteristics of the site and anthropogenic activities taking place in its vicinity. For example Tripathi et al. (1993) reported high concentrations of Pb, Cd, Cu and Zn in Deonar, Mumbai. The site was affected by the emissions from chemical industries. Also soil has been found to be a significant contributor in several studies (TERI, 2001).

In this study only the CO, $NO_2$ and $SO_2$ concentrations were monitored simultaneously with the number and mass concentration measurements (Paper I and V). Gas concentration measurements were conducted to support the analysis of the possible sources. Sharma et al. (2003) conducted a preliminary analysis of the organic compounds from the samples collected from the same location as where the number concentration and the number size distribution measurements were performed in this study in New Delhi. Their results suggested that biomass and/or refuse burning and motor vehicle exhaust emissions were significant contributors to the organic fraction of ambient $PM_{10}$ in this area. Also a study conducted by Reiner et al. (2001) above Indian Ocean indicated an important contribution from biomass or biofuel burning sources. They measured the ratio dX/dCO (X=acetone, acetonitrile, sulfur dioxide, potassium, or sulfate) during the flights during INDOEX. According their measurements most of the CO in the continental outflow is due to biomass or biofuel burning, whereas the majority of the aerosols results from fossil fuel burning.

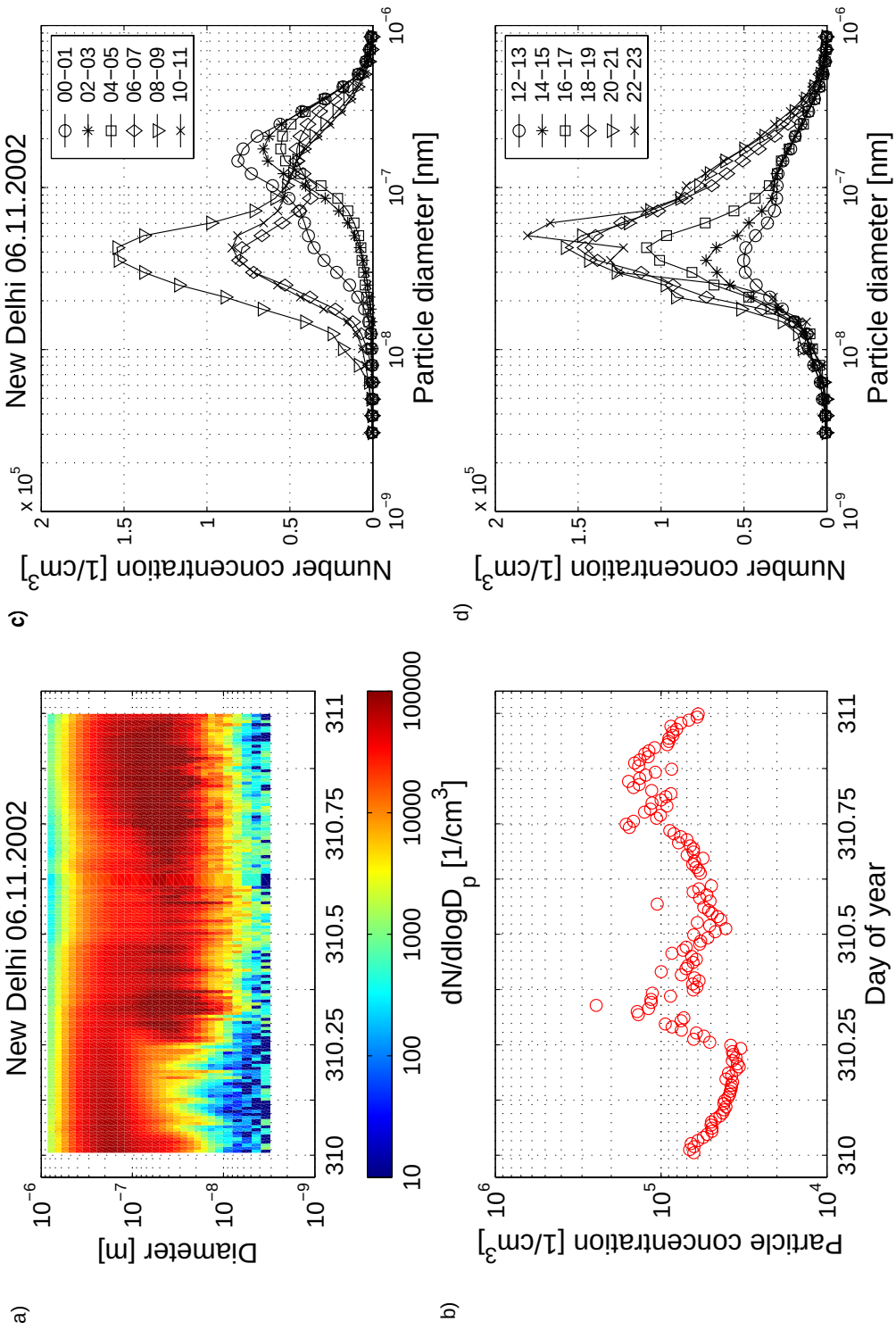


Figure 3. Measured aerosol number size distribution (a) and number concentration (b). The x-axes in Fig. 3a and 3b represent the time of day and the y-axis in a) particle diameter (m) and in 3b) particle number concentration ($cm^{-3}$) for the same period. Figs. 3c and 3d represent selected 1 h mean number size distribution on the same day. Fig. 3c presents the size distribution before noon and Fig. 3d presents the size distribution after noon.

# 3. Aerosol measurements in India

## 3.1 Description of the measuring sites

In India all the experiments were performed in three different cities from March to December in 2002 (Figure 4). Details of the other measuring sites outside of India are presented in Paper IV.


Figure 4. Locations of the measuring sites of this study are represented by a square.

In the Indian capital city of New Delhi the experiments were performed at India Habitat Centre (IHC/TERI, 28º 35' N, 77º 12' E, 218 m asl) between March and November (Paper I) and from 26$^{th}$ October to 9$^{th}$ November (Papers II-IV). The measuring site was located in a residential area a few kilometers south from one of the city's centers called Cannaught Place. The equipment were placed 20 meters from the traffic line at a height of 3 meters ($PM_{10}$ Sampler and CPC 3007) and 15 meters (DMPS). There were two big parks very close to the sampling site and there were no industries nearby (4 km radius). New Delhi is a land locked city. The distance from the sea and cold waves from the Himalayan region gives Delhi an extreme type of continental climate. The weather remains hot and humid during the summer months (March to May) and is cold during the winters (December to February). The average temperature of New Delhi ranges from 25 ºC to 46 ºC during summer and 2 ºC to 5 ºC during winter.

The data presented in the Paper V was collected in the private house in the city of Nagpur and in several private houses in the city of Mysore between March and December 2002. The city of Nagpur (21º 06' N, 79º 03' E, 247 m asl) is located in the eastern part of the Indian state of Maharashtra 1094 km South from New Delhi. The city lies almost at the geographical centre of India and hence the climate is mainly influenced by its inland position and the prevalence of

continental air during a major part of the year. The climate of Nagpur follows a typical seasonal monsoon weather pattern. The onset of monsoon is usually from the end of June extending up to September peaking during July and August. The monthly average rainfall during monsoon season varies from 180 mm (September) up to 320 mm (July). During other seasons only showers are observed. Nagpur faces high variations in temperature with very hot summers and cold winters. The maximum temperatures as high as 48º C are usually reached in May and June. The minimum temperatures (6º C) are recorded through December to January. The measuring site located in a private house in a residential area about three kilometres east from the city's centre called Sitabuldi.

The city of Mysore (12º 18' N, 76º 42' E, 770 m asl) is located in the Southern Indian state of Karnataka 136 kilometres from the city of Bangalore. Mysore does not have as extreme types of climate as New Delhi and Nagpur. Mysore is located 770 m above sea level and hence, enjoys pleasant climate around the year. Temperature varies between 15 ºC and 35 ºC throughout the year. The rainy season is from June to August but it rarely rains all day.

### 3.2 Instrumentation

#### 3.2.1 Particulate matter ($PM_{10}$) sampler

Particulate matter (PM) concentrations are widely monitored worldwide. Organization like World Health Organization (WHO) and national environmental protection agencies (e.g. U.S. Environmental Protection Agency) have defined ambient air quality standards in which one factor is the level of $PM_{10}$ (particles size less than 10 µm by aerodynamic diameter) in ambient air.

In India, $PM_{10}$ concentrations are usually monitored by High Volume Samplers with average flow rate more than 1.1 $m^3$/min. The most common instrument is an APM dust sampler, which is manufactured by Environtech Instrument Ltd., New Delhi. The APM sampler first separates the coarser particles (larger than 10 µm). This size limit is not sharp; the cyclone and impactor pre-separators remove half of the particles at the cut size and larger particles with increasing efficiency. Particles less than 10 µm are filtered out on 0.5 µm pore-size filter. The filter is weighed before and after sampling. The weight increase is the mass of particles smaller than 10 micrometers. The mass concentration of $PM_{10}$ particles (µg$m^{-3}$) is determined by dividing the particulate mass by the sampled air volume. By this instrument the remaining air is passed to gas samplers where concentrations of various gases like $NO_2$ and $SO_2$ can be measured.

#### 3.2.2 DustTrak

The DustTrak uses a light scattering technique to measure the aerosol mass concentration in an air flow that passes through an impactor assembly. The amount of scattered light is proportional to the volume concentration of the aerosol. The scattered light decreases as a function of the sixth power for the particles diameter smaller than third the wavelength of the laser (780 nm), thus limiting the smallest detectable particles to approximately 0.1 µm (TSI Incorporated, Aerosol Monitor Theory of Operation Application Note).

In this study the DustTrak Aerosol Monitor TSI model 8520 was used to collect the data for the Paper V. A $PM_{2.5}$ impactor was used for the inlet of the DustTrak allowing measurements in the size range 0.1–2.5 µm. The flow rate of the instrument was 1.7 l/min.

### 3.2.3 Condensation Particle Counter (CPC)

The number concentration of submicron aerosol particles can be measured by Condensation Particle Counters (CPC). In CPC small aerosol particles are grown to optically detectable sizes by condensing some vapour on them.

There were three laminar flow types of CPCs used in this study: TSI models 3007 (TSI Incorporation, operation manual), 3010 (Quant et al., 1992) and 3025 (Stolzenburg and McMurry, 1991). In TSI CPC 3025 the aerosol flow is divided in two sections – to sheath air (90% of the flow) and polydisperse aerosol (10%) flow. The sheath air is first saturated by alcohol vapour in a slightly heated saturator. The polydisperse aerosol flow is directly led to a condenser. After the saturator the temperature of the alcohol-aerosol mixture is decreased in a condenser so that the alcohol becomes supersaturated and condenses onto the aerosols. Through condensation the aerosols are grown to detectable sizes. The flow is then focused in a nozzle and introduced into a detection unit. A cut size 3.0 nm (with the 50% detection efficiency) is received with the TSI 3025 and a cut size of 6.0 nm with the TSI 3010. In the case of CPC 3010, the cut off size is lower than given by the manufacturer, since we have increased temperature difference between the saturator and the condenser. For the TSI 3007 the cut size is 10.0 nm. The equipments were calibrated before the measuring campaign. Different calibration methods are described e.g. by Aalto et al., (2001). The TSI 3007 was calibrated by Hämeri et al., (2002).

### 3.2.4 Differential Mobility Particle Sizer (DMPS)

The Differential Mobility Particle Sizer (DMPS) is a device that measures the particle number size distribution. The DMPS consists of three main parts in which: 1) a charger charges particles, 2) a Differential Mobility Analyzer (DMA) classifies particles according their electrical mobility, 3) and a CPC detects the particle number concentration after the classification (Hoppel et al., 1978). The analysis is differential since only a small mobility fraction can be analyzed at a time.

Determination of the number size distribution over the size range 3-800 nm is possible by using two parallel DMPS systems. The instrumentation used to collect the data for the Papers II, III and IV was similar to instrumentation described by Aalto et al. (2001).

### 3.2.5 Gas analyzers

There are numbers of commercially available gas analyzers. Is this study a portable Enmet OMNI-4000 gas detector was used to measure CO concentrations (Enmet Corporation, OMNI-4000 manual). This detector uses new sensor technology that takes a standard sensor and allows interchangability in an instrument without the need to calibrate each sensor to the instrument (OMNI-4000 manual). The concentrations of $SO_2$ were measured by the improved West and Gaeke method with ultra-violet fluorescence and $NO_2$ concentrations by the Jacob–Hochheiser modified method (Na-Arsenic) with gas phase chemiluminescence (Stern, 1968, APHA, 1977).

## 4. Characteristics of the aerosol formation and growth dynamics

Atmospheric particle formation and growth are complicated key processes in determining atmospheric aerosol dynamics. These processes involve nucleation, growth of the recently formed particles by condensation and coagulation of growing nuclei into larger pre-existing aerosols. Nanometer-size atmospheric aerosol formation and growth events have been observed at a number of sites around the world (Kulmala et al., 2004) and now also nucleation events have been observed in a highly polluted urban environment in India (Fig. 5).

In this study we have used a method presented by Kulmala et al. (2001) in the analysis of the experimental data presented in Papers II-IV. This method allowed the direct calculation of the particle growth rate (GR) and the condensation sink (CS) from the particle size spectra obtained from at one measurement site. From this information, the concentration of vapours (C) and their source rate (Q) can be estimated (see also Kulmala et al., 1998). The equations from which GR, CS, C and Q are calculated are presented in the Paper IV.

We have also used the coagulation sink (*CoagS*) concept in our analysis (Kulmala et al., 2001). The coagulation sink determines how rapidly nanometer-size particles are removed through coagulation. $CoagS$ was determined from

$$CoagS = \sum_j K_{ij} N_j \qquad (1)$$

in which $K_{ij}$ is the coagulation coefficient (Fuchs, 1964, Seinfeld and Pandis, 1998).

The formation rates (or nucleation rate) $J_3$ presented in the Paper III can be solved from the equation

$$\frac{dN_i}{dt} = J_i - S \qquad (2)$$

in which

$$S = CoagS \cdot N_i + \theta \qquad (3)$$

In the equation (3) $N_i$ is the aerosol number concentration in the size class $i$ and $\theta$ is the loss due to condensation growth (Kulmala et al., 2001). Hence the effect of the sinks to the formation rate ($J_3$) at event start time was

$$S = CoagS \cdot N_{(3-6)} + \frac{\Delta N}{\Delta d_p} \cdot GR$$

$$= (13.00 \cdot 10^{-4} \cdot 3008 + \frac{3008}{3} \cdot \frac{16.0}{3600}) \text{ cm}^{-3}\text{s}^{-1}$$

$$= 8.37 \text{ cm}^{-3}\text{s}^{-1}$$

in New Delhi on 5th of November 2002. The corresponding $dN_{(3-6)} / dt$ was 5.6 $cm^{-3}s^{-1}$ and after the correction $J_3$ was 13.97 $cm^{-3}s^{-1}$. Hence, the effects of the sinks have to be taken into consideration while defining formation rates, especially in the areas where formation rates are relatively low.

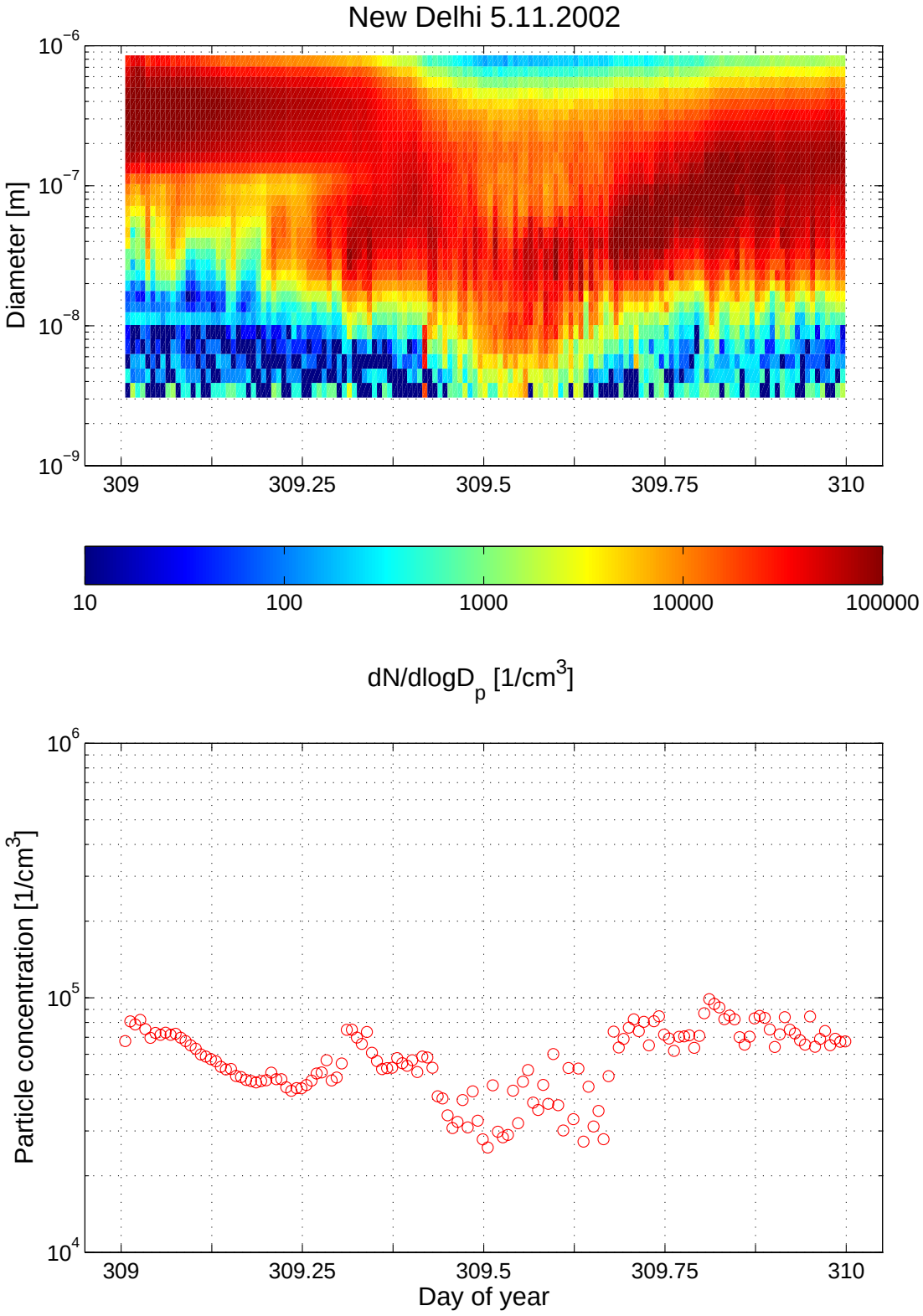


Figure 5. Evolution of the particle number distribution of the total number concentration as a function of time for 5th November 2002. A nucleation event is observed after noon.

## 5. Review of papers

The Papers I-III and V reviewed below are based on the field experiments conducted in India in 2002. The data presented in Papers I, II and III was measured in a residential area in New Delhi and the data presented in Paper V was measured in a private flat in the city of Nagpur and in several private urban houses in the city of Mysore. Besides the number size distribution data collected from New Delhi, data from five other measuring locations (Antarctica, Athens, Marseille, Hyytiälä and Värriö) is used in Paper IV.

Paper I describes diurnal, weekly and seasonal variations of aerosol number, $PM_{10}$ mass, $NO_2$, $SO_2$ and CO concentrations measured in a residential area in New Delhi. Diurnal one hour mean number concentration varied from 16,000 $cm^{-3}$ (June, 5 a.m.) to 130,000 $cm^{-3}$ (March, 9 p.m.) while diurnal four hour mean $PM_{10}$ mass concentrations varied from 255 µg $m^{-3}$ (10 a.m. – 2 p.m.) to 325 µg $m^{-3}$ (10 p.m. – 2 a.m.). The correlation of the aerosol number and $PM_{10}$ mass concentration indicated positive correlation for small values of the mass concentration (below about 300 µg $m^{-3}$). However, with even higher values of $PM_{10}$, the increased coagulation sink diminishes the number concentration values. In addition to the observation, this interesting phenomenon was demonstrated through a dynamic model for a coagulating aerosol with sources.

Paper II presents the diurnal variation of number size distribution (particle size 3-800 nm) and modal parameters (geometric standard deviation, geometric mean diameter and modal aerosol particle concentration) measured in field campaign conducted during October and November 2002 in New Delhi. Besides the analysis of possible sources, the analysis of new particle formation and growth in this kind of an environment is presented. The results indicated clear increase in Aitken mode (25-100 nm) particles during traffic peak hours, but towards the evenings there were more Aitken mode particles compared to the mornings. Also high concentrations of accumulation mode particles (> 100 nm) were detected in the evenings only. In the evenings, biomass/refuse burning and cooking are possible sources beside the traffic. The formation rate of 3 nm particles (J3) of the observed new particles formation events varied from 3.3 to 13.9 $cm^{-3}s^{-1}$ and the growth rate varied from 11.6 to 18.1 $nmh^{-1}$ showing rapid growth and high formation rate, which seems to be typical in urban areas.

In Paper III the observed death of nucleation and Aitken mode particles during a significant pollution event, in which the accumulation mode particles comprised practically 100% of total particle number concentration, is explained using a simple aerosol dynamic model based on basic aerosol physics. According to the model the limits for existence of nucleation mode as well as Aitken mode particles in the atmosphere can be evaluated. For the analysis, the coagulation and condensation sinks values were calculated from the measured number size distribution data. The coagulation sink values ranged from 0.673 $s^{-1}$ to $2.04 \times 10^{-4}$ $s^{-1}$ for particles of diameter 1 nm to 100 nm and the condensation sink values from 0.075 to 1.29 $s^{-1}$.

In Paper IV studies on the formation and growth properties of nucleation mode particles in clean (Antarctica, Finnish Lapland and Boreal Forest) and polluted urban (Athens, Marseille and New Delhi) environments are presented. The concentration of condensable vapours and their source rate have been estimated with the help of the diameter growth rate and condensation sink obtained from the measured number size distribution dynamics. The estimated source rate of condensable vapours varied up to four orders of magnitude between the most polluted (New Delhi) and cleanest sites (Finnish Lapland and Antarctica) considered in this study. Preliminary results indicated that sulfuric acid plays a more important role compared to the condensable organic vapours in the growth of nucleation mode particles in polluted environments. Despite large variations in vapour sources, the

growth rate of nucleation mode particles in different sites varied no more than 1-2 orders of magnitude. In polluted environments large vapour source rates usually appeared with large condensation sinks hence balancing the condensable vapour concentrations. The increase of average particle growth rate with the level of pollution is explained by the fact that low particle growth rates cannot be observed in highly polluted urban environment due to the effective scavenging of the smallest aerosols.

Paper V presents first time initial information on indoor number concentration ($D_p > 10$ nm, $cm^{-3}$) levels, indoor/outdoor-number concentration ratio (I/O), exposure patterns and correlations with CO and aerosol mass concentration (appr. $PM_{2.5}$, $\mu gm^{-3}$) in urban Indian households. The results indicate remarkably high indoor number and mass concentrations and I/O-number concentration ratios caused by cooking. Very high particle number concentrations (300,000 $cm^{-3}$) and mass concentrations (> 1000 $\mu gm^{-3}$) were detected in same houses. High indoor concentration can partly be explained by poor ventilation systems, but in some areas, also outdoor combustion processes had a negative impact on indoor air quality.

## 6. Summary and Conclusions

The seasonal, weekly and diurnal aerosol number ($D_p > 10$ nm, $cm^{-3}$) and mass (appr. $PM_{10,}$ $\mu gm^{-3}$) concentrations and variations have been investigated in the measuring site located in New Delhi, India. The results indicated clear seasonal and diurnal variations. Seasonal variations were biggest in mass concentration. The mean mass concentration measured in November was almost three times higher than in June. Clearest diurnal variations were detected in number concentration. Two peak hours in number concentration were observed at traffic peak hours 7-8 a.m. and 7-10 p.m. The mass concentration increased towards the evening and decreased after midnight, but exact diurnal variations in the mass concentrations could not be observed since the time resolution of the $PM_{10}$ measurements was four hours. The variations in the concentrations between the weekdays and weekends were not such clear as seasonal and diurnal variations. The weekly number concentrations were higher indicating emissions from transportation, but the mass concentrations were observed to be higher during weekends. This can be explained by different sources of emissions in New Delhi. The increase in all studied concentration variables towards the evening and midnight can also be explained by domestic use of fossil fuels and biofuels, however possible mixing within the boundary layer can not be overlooked either. The relationship between the number and $PM_{10}$ mass concentration indicated a positive correlation for small values on mass concentration (below about 300 $\mu g\ m^{-3}$). However, with even higher values of $PM_{10}$, the increased coagulation sink diminishes the number concentration values. This interesting phenomenon was demonstrated through a dynamic model for a coagulating aerosol with sources (Paper I).

Seasonal and diurnal indoor and outdoor number concentration and variations as well as I/O-relationship were investigated in the measurements conducted in a private flat in the city of Nagpur. Also diurnal indoor number ($D_p > 10$ nm, $cm^{-3}$) and mass (appr. $PM_{2.5,}$ $\mu gm^{-3}$) concentrations, variations and relationship were defined in the measurements conducted in five urban houses in the city of Mysore. In these households the main cooking fuel was LPG (Nagpur) or LPG/kerosene (Mysore). Clear seasonal variations were observed. The indoor number concentration (24-h average) was lowest in October (21,964 $cm^{-3}$) and highest in the end of April (40746 $cm^{-3}$) and the outdoor number concentration was lowest in August (25,280 $cm^{-3}$) and highest in March (32,258 $cm^{-3}$), respectively. Normal indoor living activities, especially cooking, clearly increased indoor particle number and mass concentration in all measuring sites, but also in some cases the outdoor sources had a major effect on the exposure levels indoors. Maximum indoor number concentrations more than 300,000 $cm^{-3}$ and indoor mass concentrations more than 3,000 $\mu gm^{-3}$ were detected. Usually the outdoor concentrations were higher, but I/O-number concentration ratios more than 4.00 were detected. A high correlation between the number and CO concentration (r=0.729) indicated that the outdoor particle concentration during the measuring period was strongly influenced by different outdoor combustion processes, i.e., burning of fossil fuels (traffic and domestic use of LPG and kerosene) and biomass/refuse. Despite the cleaner fuels, the cook stoves that used LPG and kerosene produced significant fine particle concentrations to the ambient air in all measured urban Indian households. It is possible to reduce exposure risk and health effects caused by poor indoor air in urban Indian households by improving indoor ventilation especially during cooking processes and reducing the penetration of outdoor particles (Paper V).

The characteristic of the aerosol number size distribution (3-800 nm) is presented for the first time in a highly polluted Asian mega city. The analysis of diurnal modal parameters revealed that there are at least two major sources in New Delhi. Beside the traffic, which is frequently repeated every day, another source in the evenings increased the geometric mean diameter of the detected particles. Based on this study and the study conducted by Sharma et al. (2003), vehicular emissions together with biomass and/or refuse burning has significant contribution to New Delhi's atmosphere. We

have also shown that nucleation events are possible in this kind of atmosphere showing rapid growth and high formation rate, which seems to be typical in urban areas (Kulmala et al., 2004). However, these events were not as clear events as observed in rural sites. In the case of every observed event, the condensation sink was at minimum during event starting time. A typical condensation sink at the event starting time was $6 \times 10^{-2}$ $s^{-1}$. The source of condensable vapour molecules is seen to be 100 times higher than corresponding source in rural area (Kulmala et al., 2001). This shows that the formation of new secondary aerosol particles in polluted urban environment is possible, but requires high vapour sources in order to be able to overcome a high coagulation sink (Paper II).

Beside traffic and combustion processes, special pollution events like fireworks deteriorate ambient air quality in New Delhi. As a result of significant sources during Diwali festival in November 2002, the death of nucleation and Aitken mode particles from the atmosphere was detected for the first time. During this event the accumulation mode particles comprised practically 100% of total particle number concentration. The non-existence of nucleation and Aitken mode particles during this event can be explained by substantial coagulation sink values, ranging from 0.673 $s^{-1}$ to $2.04 \times 10^{-4}$ $s^{-1}$ for particles of diameter 1 nm to 100 nm and by high and increasing condensation sink values from 0.075 to 1.29 $s^{-1}$ (Paper III).

The formation and growth of atmospheric aerosol in the nucleation mode in different environments have been studied by using an analytical tool. From the aerosol number size distribution spectra the growth rate, condensation sinks were calculated and with the help of them the concentration of condensable vapours and their source rate has been estimated. The estimated source rate of condensable vapours was found to vary up to four orders of magnitude between the most polluted (New Delhi) and cleanest sites (Finnish Lapland and Antarctica) considered in this study. The growth rate of nucleation mode particles varied only 1-2 orders of magnitude between different sites. The primary reason for this is that large vapour source rates usually appear in concert with large condensation sinks hence balancing the condensable vapour concentrations. The average particle growth rates increased with the level of pollution, which can be explained by the fact that low particle growth rates can not be detected in highly polluted environments due to the effective scavenging of the smallest growing particles by the pre-existing aerosol population (Paper IV).

This study has revealed that new particle formation is possible even in a highly polluted urban environment. On the other hand, during substantial pollution events even Aitken mode particles disappeared through coagulation. The formation and growth rates were found to be at similar magnitudes with the observations in other urban locations (Kulmala et al., 2004).

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