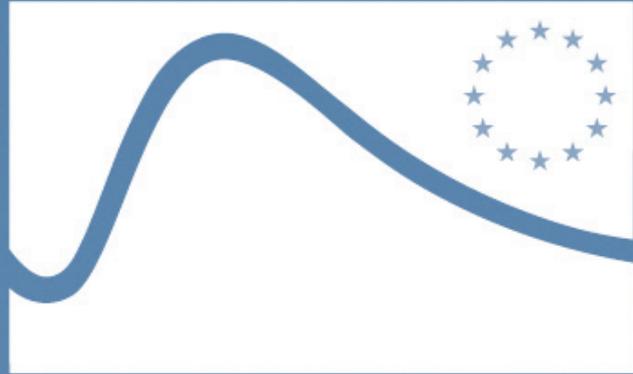


INTERNATIONAL CONFERENCE IN DRESDEN



UFIPOLNET

Ultrafine Particle Size Distributions  
in Air Pollution Monitoring Networks

Ultrafine Particles in Urban Air  
Dresden 23 to 24/10/2007



Freistaat  Sachsen  
Saxon State Agency for Environment and Geology



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UFIPOLNET LIFE04 ENV/DE/000054



**Proceedings of UFIPOLNET Final Conference: Ultrafine Particles in Urban Air, Dresden 23 to 24/Oct/2007**

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# LECTURES

# The Role of Ultrafine Particles in the Urban Atmosphere

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Keywords: air quality, ultrafine particles, soot

The atmospheric aerosol consists of solid and/or liquid particles covering the size range from few Nanometers until 10<sup>th</sup> of Micrometers. One can divide the whole size range into three main classes; coarse particles (greater than 1 µm), fine particles (greater 0.1 and smaller 1µm µm), and ultrafine particles (smaller than 0.1 µm). The coarse mode contains mainly dust, sea salt, pollen but also abrasion from tires near streets. The lifetime is relatively short because they quickly fall out by sedimentation. Fine mode particles are generally more representative for the regional aerosol since their residence time in atmosphere can be from days until few weeks. These particles are mainly grown up from ultrafine particles by condensation of vapours and cloud processes.

Ultrafine particles in the atmosphere are generated mainly by two processes: Homogeneous nucleation from condensable vapours in the atmosphere (new particle formation) and combustion of fossil fuel or biomass.

New particles formation in urban areas can have two sources; 1) solar radiation and a subsequent production of condensable gases by photo oxidation processes, and 2) vehicle emissions the production of condensable vapours after cooling. These particles grow in urban areas to sizes of 10-20 nm and consist mainly of sulphuric acid and organic carbon.

Incomplete combustion of biomass and fossil fuel leads to the direct emission of carbonaceous particles. In urban areas, diesel-driven vehicles are the main sources of soot particles, which consist of elemental carbon and adsorbed organic material. Measurements at the tail-pipe of vehicles or at a kerb-site (Rose et al., 2006) showed that the number peak of soot particles lies in the ultrafine size range between 50 and 80 nm.

Epidemiological studies have shown that aerosol particles can lead to respiratory tract or cardio-

vascular diseases. Ultrafine particles are presently under the discussion to be especially responsible for such diseases (e.g. Peters et al., 1997).

In the respiratory tract, water- and lipid-soluble material of deposited aerosol particles is dissolved and easily removed. Large insoluble particles are absorbed by macrophages and removed afterwards. Especially ultrafine insoluble particles, such freshly emitted soot, are believed to be associated with the greatest risk in relation to the above mentioned diseases. Insoluble ultrafine particles may cross cell borders and can be distributed in the whole human body. Fresh ultrafine soot particles are especially surface-reactive and may additionally carry carcinogenic substances such as PAHs.

The current legal metric to quantify particulate air quality are 24 h averages of PM<sub>10</sub> particle mass concentrations (particles smaller than 10 µm). Since a large fraction of this PM<sub>10</sub> chemical composition (inorganic salts or acids, most of the organic carbon) is less relevant in terms of being responsible for respiratory and cardio-vascular diseases, it might be necessary to add other metrics to the existing measurements. Online soot concentration measurements, especially in traffic-dominated urban areas, would provide a direct measure of surface-reactive soot concentrations and related toxic and carcinogenic compounds.

## References:

- Peters, A., Wichmann, H.E., Tuch, T., Heinrich, J., Heyder, J. (1997) Respiratory effects are associated with the number of ultrafine particles. *Am.J.Respir.Crit.Care Med.* 155, 1376-1383.
- Rose D., Wehner B., Ketzler M., Engler C., Voigtlander J., Tuch T., Wiedensohler A.. (2006) Atmospheric number size distributions of soot particles and estimation of emission factors. *ACP Vol. 6*, 1021-1031.

## Ultrafine Particles Chemical Content

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Keywords: UFP, Ions, OC/EC, Organics

In the first part of this presentation the current knowledge on particle chemical composition and its size-dependence will be summarized. The main groups of particle constituents are reviewed and it will be outlined what can be learned from the quantitative chemical analysis of particle constituents

The IFT chemistry department has performed a number of particle characterisation field campaigns with extensive size-resolved sampling and chemical characterisation over the last decade and mostly in the urban environment. Sampling was mostly done with the so-called six stage Berner impactor, the first stage of which samples particles with aerodynamic diameters in the range 50-140 nm and hence covering the larger fraction of ultrafine particles. Results from different campaigns centered on the chemical composition of the first Berner impactor stage will be reviewed. The substance groups of interest cover OC, EC and TOC, the inorganic anions and cations and single organic species. In selected experimental campaigns also metals were determined.

Methods for the determination of single organic compound are summarized and their potential for different applications such as mass closure or source apportionment is elucidated. Unfortunately, many currently available methods for the determination of single organics require input masses of material which could only be produced by sampling ultrafine particles in unrealistically long sampling efforts.

Efforts have been started to extend particle sampling and subsequent chemical analytics into the range beyond 50nm as aerodynamic. Several approaches are being described such as an extended cross comparison of the nano-Moudi sampler with DMPS (i.e. particle size distribution) measurements which lead to considerable discrepancies especially for very small particles. An outlook is given on ongoing and future sampling development.

As an alternative to sampling followed by offline chemical analysis, the application of particle realtime analysis with the Aerodyne Aerosol Mass spectrometer (AMS) will also be discussed.

A summary will be given on the current state of the art for chemical sampling and analysis of particles and ultrafine particles together with an outlook.

## **LIFE and LIFE + .....from program to projects**

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Keywords: LIFEIII, LIFE+ .

With the preparation of the new structural funds period 2007 - 2013, other EU-funding programmes have parallel redesigned their overall scope as well as their framework regulations. The LIFEIII - programme, the funding source of UFIPOLNET, has been redesigned to the LIFE+-Programme.

Whilst some company-oriented key-contents are now covered by other programmes, LIFE+ remains an important and viable instrument for the support of innovations in the environmental sector and for the realisation and promotion of nature protection and biodiversity.

The presentation explains the way from LIFE III to LIFE+ by highlighting the main criteria for the new shape of the programme. It also gives an overview on how the different topics covered by the former programme are now covered by other funding sources. Furthermore, the concrete possibilities for project initiators are listed up and implementation examples are proposed. Furthermore, funding rules as well as deadlines are presented.

## UFIPOLNET: Purpose - Partner - Project

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Keywords: atmospheric aerosols, instrument development, number concentration, number size distribution

The objective of the project UFIPOLNET (Ultrafine Particle Size Distributions in Air Pollution Monitoring Networks) is to demonstrate that the newly developed Ultrafine Particle Monitor is able to perform adequately in routine air monitoring networks.

Epidemiological studies show a relationship between high concentrations of UFP and adverse health effects. However, there are only a limited number of long-term ultrafine particle (=UFP) measurements in Europe.

Therefore the European Commission addressed its needs to get more information about UFP concentrations within the CAFE process and the Thematic Strategy on Air Pollution.

For these reasons a group of different organisations, such as research institutes, universities, monitoring networks, as well as big and small companies, were used to develop the project idea from 2003 to 2004. The goal was to create an easy to use and reasonably priced Ultra-fine Particle Monitor. The idea was supported by the LIFE-program of the European Commission DG Environment.

The resulting instrument produces a number size distribution in 6 size classes between 20 – 800 nm. In addition the sampling conditions were harmonised at all sampling stations.

The instruments have run since February 2007 in Dresden, Augsburg, Stockholm and Prague. All sites are near busy roads; Augsburg is an urban background site. The data of all sites are transferred to the different databases of the central measurement network station. The data of the 4 stations are sent monthly to the central database where it is evaluated. The different size classed number concentrations are correlated with other airborne pollutants and traffic numbers. Comparable results will allow absolute differences between ultrafine aerosol size distributions at many polluted sites to be analyzed over long periods.

It is planned to run the 4 instruments for a longer period over several years.

First comparisons with a DMPS for ambient aerosols show a good correlation. Also NOx and other traffic caused pollutants correlate good.

Comparison between the 4 different stations showed different factors between particle number concentrations and to NOx in August 2007.



Figure 1: Ultrafine Particle Monitor at 4 stations in EU

### Acknowledgements

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# A new particle measurement system for environmental ultrafine particles

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Keywords: ultrafine particles, instrumentation, measurement

It is well known that ultrafine particles have an impact on human health. Consequently it is necessary to monitor the exposure in cities and urban areas. There are some instruments commercially available for this task like SMPS, DMPS or FMPS but their original field of application is contradictory to the utilization in air pollution monitoring networks.

These sensitive devices had been developed primarily for scientific purposes in the lab environment, they are servicing-intensive and delicate to handle. The use of radioactive sources in the charger complicates the transport of the devices. Furthermore a compromise has to be found between the necessary accuracy in the field of air monitoring and the costs of the devices. The high accuracy of these devices is not needed within the scope of air pollution monitoring and lead to too high costs.

Based on the requirements of monitoring networks a new particle measurement system was developed and tested in the project UFIPOLNET – using of a diffusion charger instead of a radioactive source and an electrometer instead a CPC:

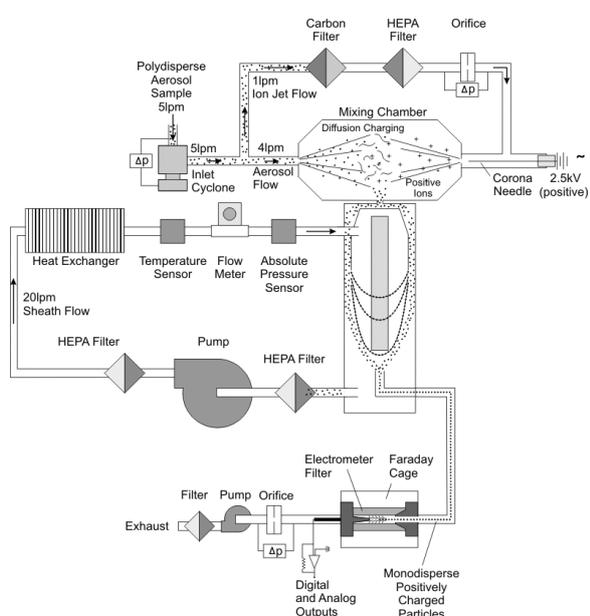


Figure 1. Schematic of the new particle measurement system for ultrafine particles.

Additional components are a long DMA (TSI 3081) with sheath air circuit, a complete control unit (PCB)

and a single board computer for data processing and a database driven data storage.

The data inversion delivers the particle number concentration in the following size classes:

CH1 (nm)	CH2 (nm)	CH3 (nm)	CH4 (nm)	CH5 (nm)	CH6 (nm)
20...30	30...50	50...70	70...100	100...200	>200

In the field of environmental aerosols particular attention has to be paid to the sampling system.. Basically it consists of a PM1-inlet, a membrane dryer and an equalizing tank. The membrane dryer requires no maintenance and induces only minimal particle losses.

In comparison measurements between the new spectrometer and a DMPS at a street canyon site in Leipzig a good correlation was found:

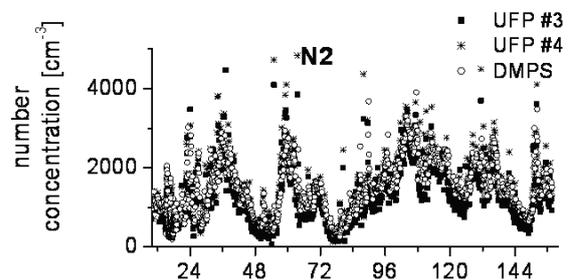


Figure 2. Comparison measurements between the new spectrometer and a DMPS (30 – 50 nm).

Within the project UFIPOLNET at four measurement stations in Europe prototypes were installed with identical sampling systems. The instruments are running over a longer period and experiences of the users will also be presented.

UFIPOLNET ([www.ufipolnet.eu](http://www.ufipolnet.eu)) is financed by the LIFE financial instrument of the European Community under No. LIFE04 ENV/DE/000054.

# Characterization of environmental ultrafine particles with the UFP 330, system calibration and evaluation

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Keywords: electrical effects, charged particles, instrumentation, measurement

The UFP 330 is an aerosol spectrometer basing its measurement on the classification of charged particles in an electric field. Accurate particle size detection requires a well-defined charge status for the aerosol which is achieved by diffusion charging. This process generates a high charge level of the particles making the data inversion difficult (Hillemann, 2007).

The aim of this work is to describe the calibration of the UFP 330 and to give some results of the evaluation process during the project.

## SYSTEM CALIBRATION

The aerosol spectrometer UFP 330 combines an electrostatic classifier and an electrometer to measure the mobility distribution of an aerosol. A measurement cycle delivers the mobility distribution of the particle-bound electrical charge. The inversion problem to calculate the size distribution  $f(x)$  from the measured current distribution  $g(y)$  is described by the Fredholm-equation.

$$g(y) = \int_a^b K(x,y) f(x) dx$$

The kernel data  $K(x,y)$  of the UFP-system is recorded by a parallel quantification of a monodisperse aerosol by SMPS, CPC and UFP.

The inversion algorithm employed in the UFP 330 uses constraints like the typical shape of an environmental particle size distribution and boundaries for the particle size.

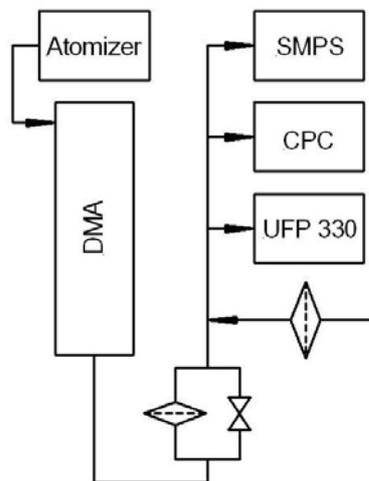


Figure 1. Experimental setup for the system calibration

## DATA EVALUATION

Since January 2007 a prototype of UFP 330 was installed in an air-quality monitoring station in Dresden. Once per month it has been dismantled and transferred to an aerosol lab to measure the counting efficiency with several test aerosols. This comparison of the UFP 330 to SMPS and CPC delivered well comparable results.

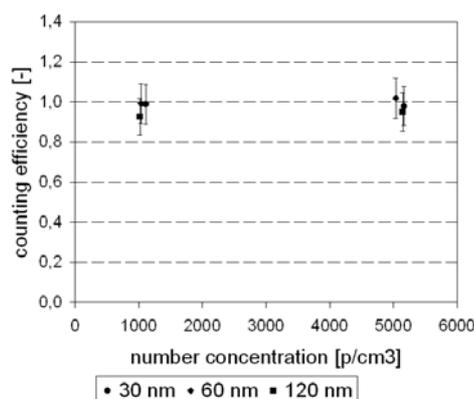


Figure 2. Measured counting efficiency for different particle sizes

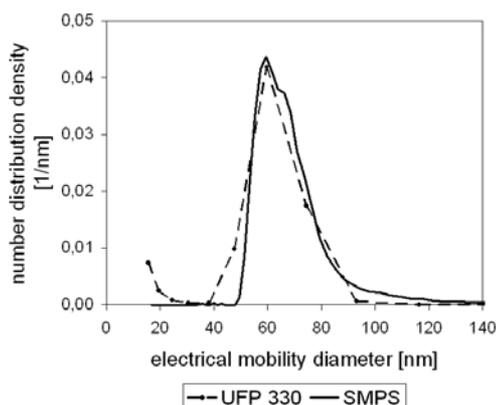


Figure 3. Comparison of the UFP 330 to SMPS

UFIPOLNET ([www.ufipolnet.eu](http://www.ufipolnet.eu)) is financed by the LIFE financial instrument of the European Community under No. LIFE04 ENV/D/000054.

L. Hillemann, A. Zschoppe and R. Caldw (2007), Aerosol mobility spectrometry based on diffusion charging, European Aerosol Conference 2007, Salzburg, Abstract T05A008.

## Ultrafine particles: Comparisons UFP 330/ DMPS

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Keywords: number concentration, instrument development, comparison, network operation

Several epidemiological studies have shown a relationship between high number concentrations of ultrafine particles (< 100 nm) and adverse health effects. However, most routine measurements of particulate matter are limited to the mass concentration, e.g. PM<sub>10</sub> or PM<sub>2.5</sub>. One major reason for this is that commercially available measurement technique is relatively expensive and needs more maintenance than in the routine network operation can be provided. Within the frame of the project UFIPOLNET a new instrument to measure ultrafine particle number concentrations has been developed which is easy to handle and needs less maintenance than e.g. available SMPS systems.

The new instrument (Ultrafine Particle Monitor, UFP330) consists of a Corona Charger, a DMA, and an electrometer. The measured current is online transferred to a number size distribution (20 – 500 nm) and locally stored. Within routine networks the number of measured parameters which might be saved continuously is limited. Thus, number size distributions are usually replaced by integral concentrations within certain size classes. For the UFP330 the size classes have been defined as follows:

name	range
N1	20 - 30 nm
N2	30 – 50 nm
N3	50 – 70 nm
N4	70 – 100 nm
N5	100 – 200 nm
N6	> 200 nm

Within the frame of UFIPOLNET 4 prototypes of the instrument have been built and are operated at 4 stations in Europe. To ensure the data quality and comparability the UFPs have been operated in parallel to a DMPS system in a street canyon, representing a typical measurement site within an urban network. Number size distributions obtained by DMPS have been converted to size classes according to N1 – N6.

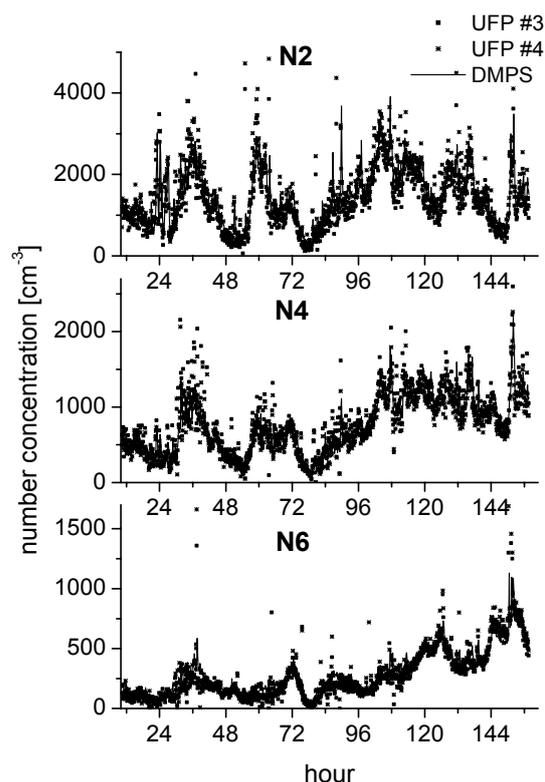


Figure 1: Number concentrations measured in parallel with two new UFP and one DMPS in the street canyon site

Figure 1 shows the results of three selected size channels for parallel measurements between two UFP prototypes and one IfT-operated DMPS. In the first view they show a good correlation, only a few outliers have been registered. In general, the correlation for the size classes N2 – N5 is higher than for the largest and smallest one. The result is nearly independent on the concentration at this site.

In February 2007 the instruments have been set up at four measurement sites within Europe. Three of them measure there parallel to another size spectrometer such as SMPS or DMPS. These results will give more information about the data quality at differently polluted sites and also over a longer period.

This project ([www.ufipolnet.eu](http://www.ufipolnet.eu)) is financed by the LIFE financial instrument of the European Community under No. LIFE04 ENV/D/000054.

# Measurements of ultra-fine particles in Europe: differences and similarities

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Keywords: PM, Europe, size distributions

Epidemiological studies have shown a close relationship between particulate matter (PM) mass concentrations and mortality and / or morbidity (e.g. Dockery et al., 1993). However, they have also pointed out spatial differences in the health impact of PM mass concentrations or mass concentration increments (e.g. Le Tertre et al., 2005). One of the reasons for this might be that PM characteristics (chemical composition, size distribution) do vary from place to place, both at a large scale (across a continent) and at a more regional/local scale (from rural background to kerbside sites). To test this hypothesis, the results of the epidemiological studies should be confronted to PM characterisation data, beyond the only PM mass concentrations reported by the air quality monitoring networks.

Such parameters (PM mass concentration, chemical composition and particle number concentration and / or size distribution) have been measured at various locations in Europe for many years. However, most of them rarely reach possible users such as epidemiologists, modellers, and policy makers. Here we present a compilation of European PM data sets that are more comprehensive than the traditional PM-mass oriented monitoring network sites (EMEP, AIRBASE).

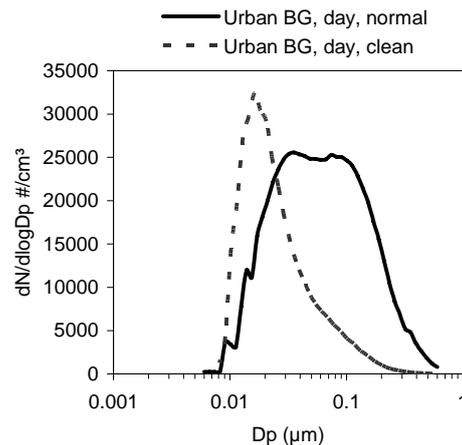
These data are complemented with a dedicated winter time study in the Milan area, where the urban and regional background particles were characterized under extremely clean conditions, i.e. in absence of a regional background (see Figure 1). This allows for an in-situ characterisation of locally emitted particles in the urban environment.

Following observations have been made from the available data sets:

- At all sites, most of the particles (70 – 80% of the number) have a diameter < 100 nm. At polluted sites, those particles consist mainly of carbonaceous material (BC + organic matter).
- Volatility measurements indicate that freshly emitted elemental carbon is not internally mixed with the volatile (organic carbon and sulfate) particles. Particles originating in the urban area come mainly from combustion processes,

especially direct traffic emissions, domestic heating and industrial activities, whereas the regionally emitted particles are different with much less traffic contribution. Nitrate, after POM the major secondary component during stagnant conditions, can be entirely apportioned to the regional background aerosol.

- Particle number (> 10 nm) increases more than proportionally to PM mass, due to an increasing contribution of small (< 100 nm) particles in polluted sites.
- At polluted sites, PM<sub>2.5</sub>, PM<sub>10</sub> as well as number concentration are highest during winter.
- At polluted sites, the relative contribution of nitrate is highest during winter due to semi-volatile nature of ammonium nitrate.
- Physical and chemical characterisation of the urban background aerosol indicate consistently that local urban emissions account for 50 – 70% of the PM mass in the Milan urban background during winter time, whereas the regional background accounts for 30 – 50% of the mass.



**Figure 1:** Urban background daytime aerosol number size distribution under normal (stagnant) winter time conditions and under extremely clean conditions. The former is a superposition of locally emitted + long range transported particulate matter. In the latter case, only locally emitted particles are observed.

## Origin and features of ultrafine particles in Barcelona

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Keywords: ultrafine particles, road traffic, air quality, number size distribution, aerosol chemistry

Concentrations of the aerosol number (N), PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub>, and the chemical composition of PM<sub>10</sub> and PM<sub>2.5</sub> are being monitored in Barcelona since 2003. The main features of the ultrafine particles number size distributions are summarised in this presentation.

In this Mediterranean city, the aerosol N<sub>10</sub>, N<sub>5</sub> (N of particles >10nm and >5nm, respectively) and PM<sub>2.5</sub> concentrations exhibits mean values of 15400cm<sup>-3</sup> (2003-2005), 22800cm<sup>-3</sup> (2005-2007) and 27µg/m<sup>3</sup>, respectively. The size distribution of N<sub>10-800</sub> (N of particles with size into the range 10-800nm) presents a maximum in the particle diameter 34nm (D<sub>pN</sub>: diameter where the dN/dlogD reaches the maximum). Ultrafine particles (<100nm) accounts for 86% of the number concentration with the range 10-800nm.

The aerosol N concentrations typically exhibit strongly marked weekly cycles. During weekdays, the number concentration reaches the highest values during the morning and evening rush hours, in such a way that the ultrafine particles concentration exhibits a morning-to-night concentration ratio of 2.5, being the highest morning-to-night concentration ratio (~3.0) observed in the 30-50nm size bin. During weekends these cycles are not observed owing to the strong change in the daily pattern of the road traffic intensity. The D<sub>pN</sub> diameter exhibits lower values during daylight (~37nm) than at night (~50nm) during working days and a relatively 'flat' daily patten during weekends (~40nm). The results obtained points that the 'road traffic intensity' (number of vehicles/hour) and the processes affecting the 'new particle formation during the dilution and cooling of the vehicle exhaust emissions' are the main factors inducing the observed mean daily patterns in the ultrafine particles concentrations.

The processes affecting the day-to-day variations in the 'number concentration and number size distribution' and their relationship with fine particle mass concentration, has also been studied. The main results are summarised as follow:

**1.** The time series of the aerosol D<sub>pN</sub> diameter and fine particle mass concentrations (PM<sub>2.5</sub> and PM<sub>1</sub>) tend to exhibit: i) correlated increases at night, and ii) day-to-day correlated variations. Moreover, during events of strong increases in the D<sub>pN</sub> diameter, ammonium-nitrate is

the aerosol specie exhibiting the highest increase. These observations points that condensation of ammonium-nitrate onto pre-existing particles is highly involved in the growth events associated with increases in the fine particle mass. These correlations between D<sub>pN</sub> and 'fine particle mass' are more clearly observed in winter because of the enhanced condensation rates at low temperatures.

**2.** The number concentration in all size ranges is negatively correlated with wind speed. However, the correlation coefficient between wind speed and the number size distribution dN/dlogD decreases when decreasing the particle size (e.g. r= -0.32 for 90nm particles and r= -0.16 for 13nm particles; Figure 1). This is caused by the fact that very small ultrafine particles are formed under 'windy clean air conditions' favoured by the low surface area available for condensation. The occurrence of these events has also been documented in other European cities (Charron and Harrison, 2003). The influence of wind speed in the particle size >20nm is higher in winter than in summer owing to the high background aerosol concentrations in this season.

**3.** The time series of the N and PM<sub>2.5</sub> concentrations tend to exhibit frequent day-to-day correlated variations. However, events of high total number concentration (N) associated with high N<sub>10-20</sub> and low PM<sub>2.5</sub> concentrations are frequently recorded. These high N<sub>10-20</sub> (>6000cm<sup>-3</sup>) and low PM<sub>2.5</sub> (<20µg/m<sup>3</sup>) episodes were mostly recorded during the morning and evening rush hours in summer and autumn being these events associated with an enhancement of the new particle formation during the dilution and cooling of the vehicle exhausts.

**4.** Events of new particle formation in ambient air (so-called 'nucleation events', as described by Kulmala *et al.*, 2004 or Hamed *et al.*, 2007) are rather infrequent. About 2 to 4 events per year have been observed, being these episodes recorded in summer. These events also contributed to the above cited high N<sub>10-20</sub> (>6000cm<sup>-3</sup>) and low PM<sub>2.5</sub> (<20µg/m<sup>3</sup>) episodes.

An analysis of the PM<sub>2.5</sub> chemistry and the aerosol size distribution dN/dlogD shows that organic matter and black carbon are the only two PM<sub>2.5</sub> chemical components showing a significant correlation with the ultrafine particles. The other

aerosol species included in the fine fraction, such as sulphate, nitrate, ammonium, sea salt and mineral dust, only shows a significant correlation with the number of particles  $>0.1\mu\text{m}$ .

The number concentration and the DpN diameter exhibit a higher day-to-day variability and reaches higher concentrations in the cold season. This is caused by the more frequent day-to-day changes in the synoptic scale meteorology and by the lower temperatures which favour both condensation and nucleation processes during the dilution and cooling of the vehicle exhaust emissions.

Some of the features observed in the ultrafine particles in Barcelona are common to other cities of the European Union (see as examples Charron & Harrison, 2003; Rodríguez *et al.*, 2007; Rodríguez & Cuevas, 2007).

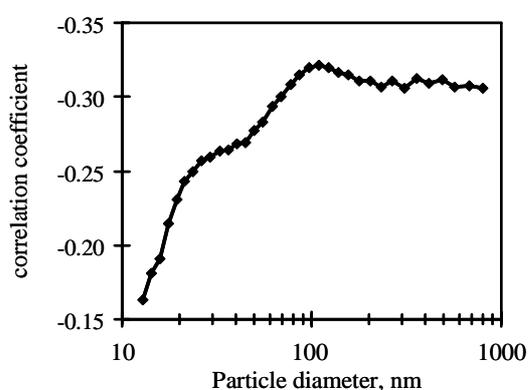


Figure 1. Correlation coefficient between wind speed and aerosol size distribution at Barcelona.

Part of this work was supported by the Ministry of Environment of Spain (project “Evaluación integral del impacto de las emisiones de partículas de los automóviles en la calidad del aire urbano”).

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# Ultrafine particles in the UK

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Keywords: particles, ultrafine, wind, emission

Particle number has been counted at eight predominately urban sites in the UK (Figure 1) since 2000 using TSI 3022A condensation particle counters (50% detection at 7 nm). Measurements of particle size spectra have also been made in the 12 – 450 nm at three sites since 1998 using scanning mobility particle sizers (TSI 3071A and TSI 3022A) size range. Annual data capture efficiencies were up to 92.7% for particle number, and up to 79.3% for particle size spectra.

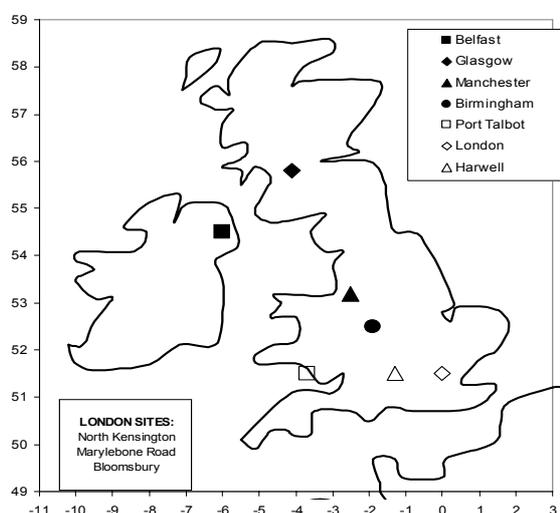


Figure 1: Site locations

These data are now available from [www.airquality.co.uk/archive/particle\\_data.php](http://www.airquality.co.uk/archive/particle_data.php) while information about the locations of the sites is available from [www.bv-aurnsiteinfo.co.uk](http://www.bv-aurnsiteinfo.co.uk).

Data has been updated since first reported (Harrison and Jones, 2005). Except at Glasgow, where data was affected by a small number of days with particularly high values, particle number concentrations were lowest in summer months (July, August) and highest in the winter (December, January) at all sites. Concentrations were similar at the urban centre and urban background sites (typically around 20,000 cm<sup>-3</sup>), with lower concentrations at a (coastal) industrial site, and substantially higher concentrations at an urban kerbside site.

On weekdays at urban sites there was a clear diurnal profile in the particle number concentration with the change in time of the peak concentration between

winter and summer time indicating an anthropogenic source for the particulate.

Directional profiles of particle number, PM<sub>10</sub> and NO<sub>x</sub> at Birmingham show higher concentrations when the wind is from the south east, mainly related to the lower wind speeds associated with this wind direction. Mean particle number concentrations were generally related to wind speed by the relationship;

$$X = c.(-\ln(U/2) + 3),$$

where U is the wind speed in knots. This relationship is similar to the results obtained for airborne bacteria at inland sites and with a standard Gaussian dispersion model (ADMS3) with a large area source and a neutral atmosphere over a wind speed range of 2 to 20 knots (Harrison et al, 2003). At Port Talbot, higher concentrations of particle number and PM<sub>10</sub> occur when the wind is from a local steel works, and when wind speed increases, and not from the direction of a nearby motorway which was associated with the highest concentrations of NO<sub>x</sub>. In contrast, at the kerb of a heavily trafficked road in an urban street canyon (Marylebone Road), the directional concentration profile of particle number was more closely related to that of NO<sub>x</sub>, than to those of PM<sub>10</sub> or PM<sub>2.5</sub>.

Emission factors for particle number in three size ranges (11-30; 30-100 and >100 nm) were estimated separately for heavy and light-duty vehicles in a heavily trafficked street canyon where the traffic speed vary considerably over short distances (Table 1) (Jones and

Particle size range [nm]	Emission factor [number veh <sup>-1</sup> hr <sup>-1</sup> ]	
	Heavy duty	Light duty
11-30	2.14 x 10 <sup>14</sup>	2.30 x 10 <sup>13</sup>
30-100	3.19 x 10 <sup>14</sup>	2.84 x 10 <sup>13</sup>
>100	1.03 x 10 <sup>14</sup>	0.70 x 10 <sup>13</sup>

Table 1: Calculated emission factors

Harrison, 2006). The calculation of emission factors assumes that particulate disperses from the on-road source in a similar way to the dispersion of NO<sub>x</sub>.

This work was funded by the UK Department for the Environment, Food and Rural Affairs under contracts EPG 1/3/184 and CPEA 28. Particulate data was collected by Bureau Veritas (formerly Casella) and the National Physical Laboratory and King's College, London. Meteorological data is from the UK Meteorological Office via the British Atmospheric Data Centre.

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# Ultrafine particles in Stockholm

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Keywords: vehicle exhaust, NOx, road traffic, PM10

## Introduction

In Stockholm, most measurements of particulate matter are for PM10 and the levels exceed the EU-directives along many streets in the city centre (Norman and Johansson, 2006). But substantial differences in the temporal and spatial distribution of PM10 and particle number concentrations (referred to as PNC) are observed due to differences in emissions, background concentrations and meteorology (Johansson et al., 2007). While high PM10 levels are mainly due to non-exhaust emissions of road wear, consisting of soil mineral, ultrafine particles (UFP, diameter < 100 nm) are due to vehicle exhaust particles, consisting of soot and organic compounds. UFP contribute with less than 10% to PM10 and is suspected to be more important for mortality than the coarse particles. Nevertheless, the EU directive regulates the total mass of all particles less than 10  $\mu\text{m}$  irrespective of size, morphology and chemistry and also irrespective of their health effects. Measures aimed at reducing the negative health impact of particles must necessarily build on an understanding of the controlling factors not only for PM10 concentrations but also for other particle size fractions. Routine monitoring of particle size distribution has now started at Hornsgatan in Stockholm in the frame of the UFIPOLNET project.

## Measurement sites & methods

Ultrafine particles have been measured using the instrument developed within the UFIPOLNET project (hereafter called UFIPOL). During a period of 2 months parallel measurements were made using a commercial SMPS (TSI 3071A) and CPC (TSI 3010) (called SMPS). Total number of particles (> 7 nm aerodynamic diameter) was measured using a CPC3022 (TSI Inc.). Other measurements include NOx, CO, BC, PM2.5 and PM10. The site, Hornsgatan, is a densely trafficked site in central Stockholm (see Johansson et al., 2007). In this paper we also present data from a rural background site, Aspveten, 70 km south of Stockholm. At this site PM10, BC and particle size distribution is measured.

## Comparison of UFIPOL and SMPS

The comparison of total particle number concentrations from UFIPOL and SMPS show good agreement with a correlation of 0.88. The median number concentrations were 9620 and 10100  $\text{cm}^{-3}$  for UFIPOL and SMPS, respectively (Table 1). However, for the smallest size fraction (20 – 30 nm) the UFIPOL showed 34% lower concentrations and

the correlation was only 0.70, indicating that the UFIPOL gives to low concentrations. Other size fractions did not differ significantly (Figure 1).

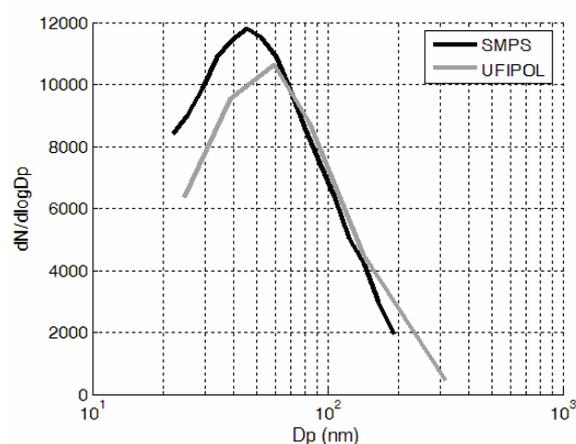


Figure 1. Mean size distributions measured with SMPS and UFIPOL.

Table 1. Comparison of UFIPOL and SMPS measurements during May - August, 2007 at Hornsgatan.

Size fraction UFIPOL (nm)	Size fraction SMPS (nm)	UFIPOL ( $\text{N}/\text{cm}^3$ )	SMPS ( $\text{N}/\text{cm}^3$ )	Slope <sup>1</sup>	Intercept <sup>1</sup>	Correlation
>20	>20.5	9620	10100	0.98 $\pm 0.01$	-370 $\pm 150$	0.88
20 - 30	20.5- 31.6	1510	2280	0.77 $\pm 0.02$	-230 $\pm 40$	0.70
30 - 50	31.6- 48.8	2870	2710	1.17 $\pm 0.02$	-320 $\pm 70$	0.75
50 - 70	48.8- 74.8	1930	2430	0.84 $\pm 0.01$	-120 $\pm 40$	0.80
70 - 100	74.8- 99.8	1570	1200	1.35 $\pm 0.02$	-70 $\pm 30$	0.82
100 - 200	99.8- 205.4	1530	1470	1.17 $\pm 0.02$	-170 $\pm 33$	0.80

<sup>1</sup>UFIPOL= $k$ \*SMPS+m, orthogonal regression, where  $k$ =slope and  $m$ =intercept.

## Comparison with PNC, NOx, BC and PM10

Figure 2 shows the correlation coefficients of particle concentrations of different sizes with total number concentrations >7 nm (PNC), NOx, BC and PM10 as measured at Hornsgatan 2007. Particle concentrations of all sizes except the largest size (100 – 200) show rather high correlations for PNC, BC and NOx, between 0.65 and 0.82. For all there is slightly lower correlation with the smallest size range (20 – 30 nm). For PM10 there is very low correlation with all particle sizes (<0.5).

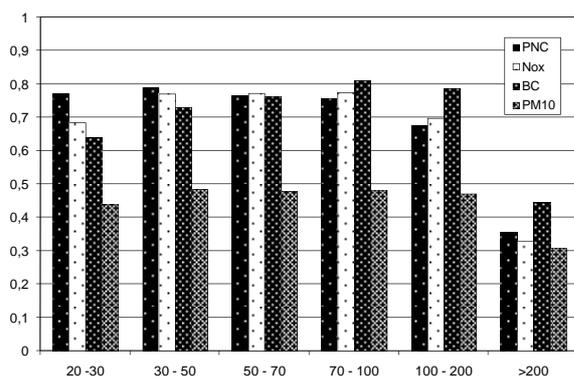


Figure 2. Correlation between different particle size fractions and PNC (total particle number, > 7nm), NOx, BC and PM10 (Hornsgatan, Feb. – Jul., 2007).

A large fraction of the total number of particles at Hornsgatan is smaller than 20 nm. Comparison with the total number as measured by the UFIPOL, with a cut-off around 20 nm with the total number of particles measured with a CPC3022 shows that 60% of all particles < 7 nm are between 7 and 20 nm (Hornsgatan during February-July, 2007). A scatter plot of total number of particles >7 nm and the sum of all measured by the UFIPOL (>20 nm) is shown in Figure 3.

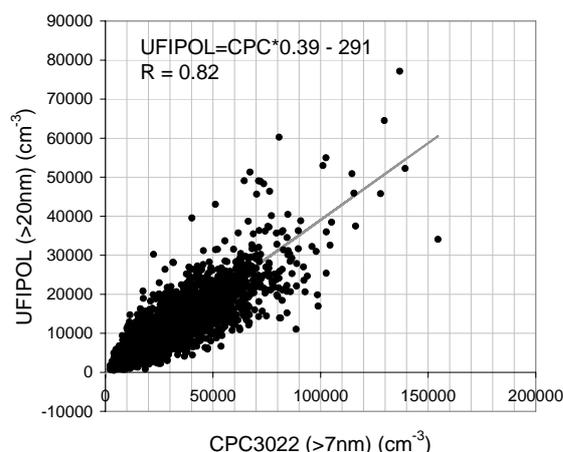


Figure 3. Orthogonal regression of total particle number concentrations (>7 nm, CPC3022) versus total particle number as measured with the UFIPOL (>20 nm).

#### Contribution from non-local sources

Most of the particles larger than 200 nm originate from sources outside of Stockholm. Total number

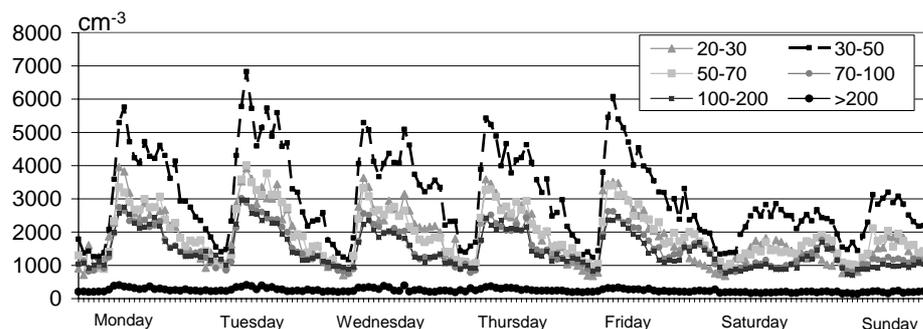


Figure 4. Mean diurnal variations of the different particle sizes based on UFIPOL.

concentrations (> 10 nm) at a rural background site 70 km south of Stockholm, is around  $2000 \text{ cm}^{-3}$ , as compared to around  $30\,000 \text{ cm}^{-3}$  at Hornsgatan. At Hornsgatan the maximum number is seen for 20 nm sized particles, whereas at Aspveten particles are around 100 nm during most of the time due to the aged aerosol (Tunved et al., 2004).

This aerosol is clearly seen to have a major influence on the variability of accumulation mode particles also at a densely traffic site in central Stockholm. This is illustrated by comparing the variability of the particle number concentrations of different sizes at Hornsgatan with that observed at a rural background site (Aspveten 70 km south of Stockholm) (Figure 5). Very high correlation is seen between particles at Hornsgatan and Aspveten of sizes larger than 200 nm, whereas the correlation is low for particles smaller than 70 nm (<0.3). The diurnal variations of particles with diameters larger than 200 nm is totally different as compared to the variation of particles <100 nm (Figure 4).

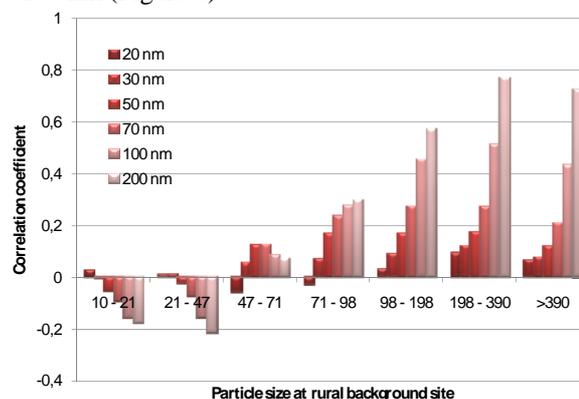


Figure 5. Correlation between particle number concentrations of different sizes at Hornsgatan in Stockholm (UFIPOL) with concentrations of different sizes at a rural background site, Aspveten, 70 km south of Stockholm.

This work was supported by the EU/LIFE project UFIPOLNET.

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# Ultrafine Particles in Urban Air of Prague

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Keywords: particle, particulate matter, gases, transport, local furnaces, OC, EC

The question of air quality and the respective problems have been recently more and more connected with particulate matter and the size fractions of these particles in connection with the gaseous components emitted mainly by passenger and freight transport and by local furnaces. The dominant role is played by transport, both in the Czech Republic and in Europe, as concerns its impact on human health.

Although the EU adopted the limit for PM<sub>10</sub> and the limits for PM<sub>2.5</sub> are being discussed, the WHO requires, with regard to research outputs, not only PM<sub>2.5</sub> and smaller PM<sub>1</sub> particles measurements but also monitoring of even smaller particles with size fractions of tens and hundreds of nanometers. Therefore the Project EU-UFIPOLNET was launched within which new instruments for monitoring the number of ultrafine particles were developed and tested in the measuring networks in the Czech Republic, Germany and Sweden.

For the Czech Republic the measuring station Praha–Smíchov Tunnel was selected, where air quality is influenced mainly by traffic – ca 30 000–50 000 cars per day (see Figure 1).

To demonstrate the results of the six-month measurements the months August and September 2007 were selected, and to show the relations between individual measured components correlations were carried out (see Table 1 and Table 2).

The presented tables show high correlation between fractions of particles CL<sub>2</sub>–CL<sub>4</sub> both in NO and NO<sub>2</sub> and CO, and in fact also with MPXY (meta-para xylene), which clearly confirms the influence of traffic on creation of ultrafine particles. Almost zero correlations for SO<sub>2</sub>, PM<sub>10</sub> and O<sub>3</sub>, which is

decomposed by reaction with NO at traffic stations, are not surprising.

In the future, after obtaining further data from the measurements, statistical processing for individual fractions will surely be useful, connected with the analysis of particles for the contents of OC/EC.



Figure 1 a, b: Smichov – Tunnel, Praha.

Table 1 Smichov – Tunnel, Praha Correlation by CORREL/MSEXCEL August 2007

	PM10	NO	NO <sub>2</sub>	NO <sub>x</sub>	CO	O <sub>3</sub>	SO <sub>2</sub>	BZN	TLN	EBZN	MPXY	OXY
CL0	0.17	0.42	0.33	0.42	0.42	0.12	0.21	0.06	0.19	0.22	0.18	0.18
CL1	0.29	0.65	0.61	0.69	0.65	-0.10	0.11	0.21	0.21	0.44	0.47	0.34
CL2	0.35	0.75	0.74	0.81	0.75	-0.29	0.01	0.31	0.20	0.57	0.67	0.46
CL3	0.38	0.80	0.78	0.86	0.79	-0.32	-0.03	0.33	0.20	0.58	0.71	0.49
CL4	0.44	0.84	0.74	0.88	0.78	-0.18	0.01	0.29	0.18	0.49	0.57	0.42
CL5	0.57	0.36	0.43	0.42	0.44	-0.16	-0.04	0.54	0.26	0.37	0.41	0.33

Table 2 Smichov – Tunnel, Praha Correlation by CORREL/MSEXCEL September 2007

	PM10	NO	NO <sub>2</sub>	NO <sub>x</sub>	CO	O <sub>3</sub>	SO <sub>2</sub>	BZN	TLN	EBZN	MPXY	OXY
CL0	0.46	0.86	0.77	0.87	0.73	-0.39	0.46	0.64	0.47	0.59	0.58	0.57
CL1	0.53	0.92	0.84	0.93	0.81	-0.39	0.44	0.72	0.53	0.67	0.65	0.64
CL2	0.54	0.92	0.87	0.94	0.84	-0.39	0.42	0.76	0.55	0.71	0.70	0.68
CL3	0.57	0.91	0.88	0.94	0.87	-0.40	0.41	0.80	0.56	0.75	0.73	0.72
CL4	0.62	0.90	0.88	0.93	0.86	-0.34	0.42	0.80	0.53	0.74	0.73	0.72
CL5	0.61	0.71	0.68	0.73	0.74	-0.46	0.33	0.78	0.49	0.72	0.71	0.67

## **Health and Particles: The epidemiological view**

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Keywords: Air Pollution, Fine Particulate Matter, Ultrafine Particles, Environmental Medicine, Public Health.

Ambient air pollution in general has improved substantially over the past decades due to innovative changes in combustion processes and overall pollution mitigation. However, environmental research has implicated over the last decade that the mass of ambient fine particles with a diameter smaller than 2.5  $\mu\text{m}$  which penetrate into the deep lung and ultrafine particles defined as particles smaller than 100 nm persist to cause significant disease burden to European populations. Specifically, systemic effects of ambient particles outside the lung have called attention and lead to exacerbation of cardiovascular diseases. Research in current years has established the biological plausibility of the health effects observed and provides evidence on several mechanisms contributing to the exacerbation of cardiovascular disease by ambient particles. No evidence for a threshold of the health effects of ambient particles has been detected. In urban areas, traffic related pollution as well as regionally transported particles persist to be associated with adverse health effects, and further research establishing the best ways to protect vulnerable populations such as children, the elderly and the chronically ill is needed.

# Updated WHO Air Quality Guidelines

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Keywords: Air pollution – adverse effects, Risk assessment.

The results of research conducted in the last decade have significantly increased the evidence on health impacts of the most common air pollutants, including particulate matter and ozone. This newly accumulated evidence was used to review and update the WHO Air Quality Guidelines, completed in 2006 and resulting in a set of guidelines for PM<sub>10</sub>, PM<sub>2.5</sub>, O<sub>3</sub>, NO<sub>2</sub> and SO<sub>2</sub> (WHO 2006). The research data for O<sub>3</sub> and particulate matter PM indicate that there are risks to health at concentrations currently found in many cities in developing and developed countries. Moreover, the research has not identified thresholds below which adverse effects do not occur. The guideline values summarized in Table 1 indicate the air quality required for a significant reduction of health risks.

Table 1. Summary of air quality guideline values

Pollutant	Averaging time	AQG value
<b>Particulate matter</b> PM <sub>2.5</sub>	1 year	10 µg/m <sup>3</sup>
	24 hour <sup>1</sup>	25 µg/m <sup>3</sup>
PM <sub>10</sub>	1 year	20 µg/m <sup>3</sup>
	24 hour <sup>1</sup>	50 µg/m <sup>3</sup>
<b>Ozone, O<sub>3</sub></b>	8 hour, daily maximum	100 µg/m <sup>3</sup>
<b>Nitrogen dioxide, NO<sub>2</sub></b>	1 year	40 µg/m <sup>3</sup>
	1 hour	200 µg/m <sup>3</sup>
<b>Sulfur dioxide, SO<sub>2</sub></b>	24 hour	20 µg/m <sup>3</sup>
	10 minute	500 µg/m <sup>3</sup>

<sup>1</sup> 99<sup>th</sup> percentile (3 days / year)

In addition to guideline values, interim targets are given for each pollutant. These are proposed as incremental steps in a progressive reduction of air pollution and are intended for use in areas where pollution is high. These targets aim to promote a shift from high air pollutant concentrations, which have acute and serious health consequences, to lower air pollutant concentrations. If these targets were to be achieved, one could expect significant reductions in risks for acute and chronic health effects from air pollution. Progress towards the guideline values should, however, be the ultimate objective of air

quality management and health risk reduction in all areas.

In addition to PM<sub>2.5</sub> and PM<sub>10</sub>, ultrafine particles have recently attracted significant scientific and medical attention. The Guidelines conclude that, while there is considerable toxicological evidence of potential detrimental effects of ultrafine particles on human health, the existing body of epidemiological evidence is insufficient to reach a conclusion on the exposure–response relationship to ultrafine particles. Therefore no recommendations can be provided at present as to guideline concentrations of ultrafine particles.

Air Quality Guidelines, Global update 2005. World Health Organization 2006.

<http://www.euro.who.int/Document/E90038.pdf>

# Can we use fixed ambient air monitors to estimate exposure to ultrafine particles?

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Keywords: ultrafine particles, outdoor, indoor, personal exposure

Nearly all epidemiological ambient air-pollution studies have used ambient air monitoring data as surrogate for the exposure of the population of interest. The reasons for the use of fixed ambient air monitors to estimate short- and long-term exposures to population are that this approach has been successful, and most epidemiological studies require large populations and long exposure/effect follow-up times. Monitoring of individual exposures is too inconvenient and expensive to be applied for such purposes. However, since people spend up to 90% of their time indoors, the validity of ambient concentrations as an accurate estimate of exposure has raised concerns because exposure misclassification could bias epidemiologic results.

Ultrafine particles (smaller than 0.1  $\mu\text{m}$  in diameter) are more variable in space and time than fine particles ( $\text{PM}_{2.5}$  or  $\text{PM}_{10}$ ) as they have a higher dependence on the particle sources and their variability due to faster removal from the atmosphere. Consequently, the exposure assessment of ultrafine particles is believed to be more error-prone than the exposure assessment for  $\text{PM}_{2.5}$  or  $\text{PM}_{10}$  (Pekkanen and Kulmala, 2004). In the present review, we provide a discussion of three issues: (a) the representativeness of an ambient monitoring station for an urban area, (b) the relationship between outdoor and indoor and (c) between outdoor and personal ultrafine particle concentrations.

## Representativeness of an ambient monitoring station for an urban area

In the framework of our study on spatial and temporal variation of particle number concentration (NC) in Augsburg, Germany we measured NC at four locations during December 2003 (winter period), and at three locations from April to May 2004 (spring period). One monitor (MON) was located at an urban background site, two monitors (FH and BOU) were located at traffic influenced background sites, approximately 100 m away from the nearest major road, and the fourth monitor (UNI) was located in the outskirts of the city.

Figures 1 and 2 present the time series of hourly NC averages for the winter and the summer period, respectively. Although the particle number concentrations at all sites followed the same pattern, the range of the day-to-day variation differs for different monitoring sites.

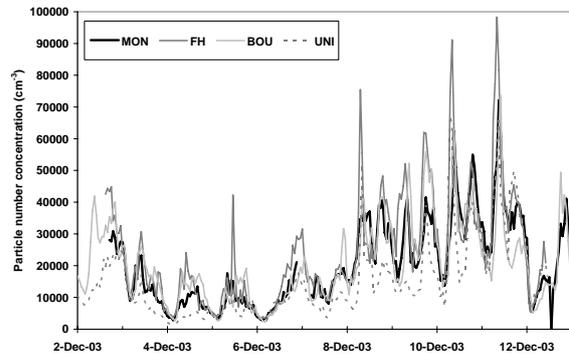


Figure 1: NC at different monitoring sites during the winter measurement period in Augsburg, Germany.

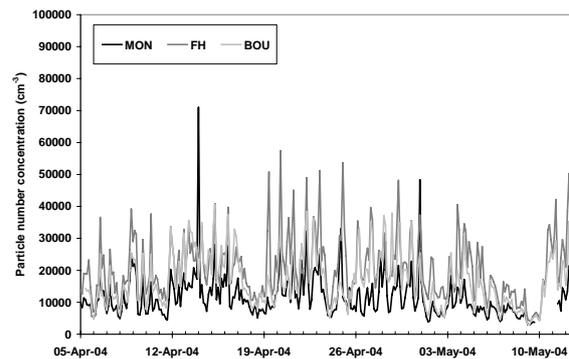


Figure 2: NC at different monitoring sites during the spring measurement period in Augsburg, Germany.

In the spring period the average NC levels at the two background sites with traffic impact were 16,943  $\text{cm}^{-3}$  and 20,702  $\text{cm}^{-3}$  respectively, compared to 11,656  $\text{cm}^{-3}$  at the background site. The inter-site correlations between the monitoring sites were high for both monitoring periods ( $r > 0.80$ ).

The absolute NC levels differed significantly implying that cross-sectional studies should enhance the spatial resolution of exposure estimates for NC to attribute more accurate exposure levels to study subjects. The high temporal correlations of NC implicate that in epidemiological time-series studies the use of one single ambient monitoring station is an adequate approach for characterizing exposure to ultrafine particles.

## Relationship between outdoor and indoor ultrafine particle concentrations

People spend typically 80-90% of their time indoors. Therefore, particle concentrations indoors

rather than those outdoors can have a large effect on personal exposure. Particles generated indoors can confound the association of health with ambient particles only if the particles from the two origins have similar toxicity and their concentrations correlate. As there are indoor sources of ultrafine particles, such as smoking, cooking and cleaning, it can be assumed that the concentration of ultrafine particles generated indoors are not correlated with the ones that penetrate into indoor environments from outdoors.

Only few studies have evaluated the correlation between outdoor and indoor concentration of ultrafine particles. When major sources indoor sources were excluded, a high correlation ( $r=0.92$ ) was observed in a room with closed windows (air exchange rate  $a=0.91h^{-1}$ ). The indoor/outdoor ratio was 0.33 for closed windows and increased to 0.78 for tilted windows ( $a=3.44h^{-1}$ ) (Cyrus et al., 2004).

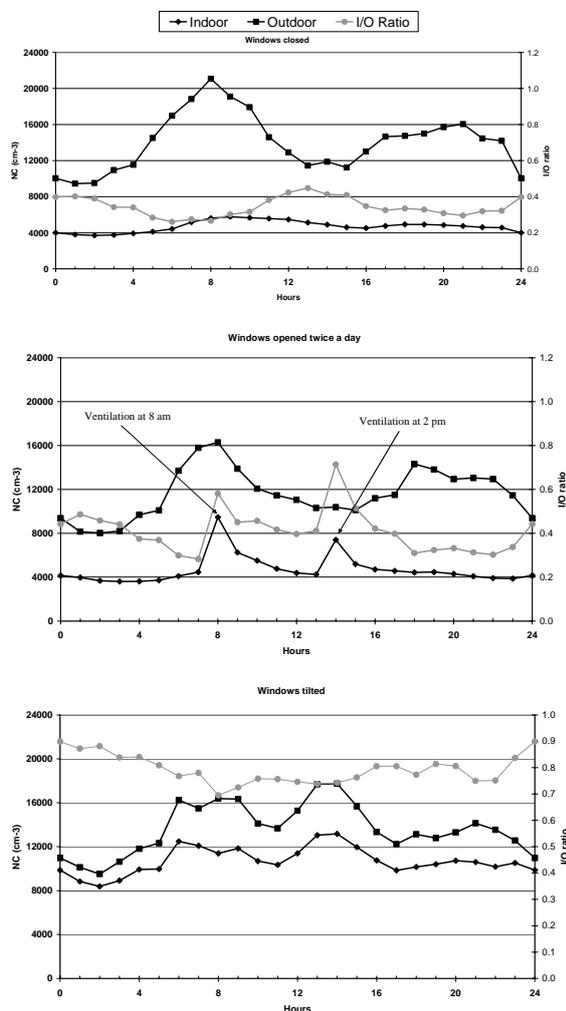


Figure 3: Diurnal pattern of indoor and outdoor average particle NC as well as indoor/outdoor ratios for different ventilation modes

Figure 3 shows the diurnal patterns of 1-hour averages of indoor and outdoor NC for different ventilation conditions: closed windows, opened

windows twice a day and tilted windows all day long. It could be seen that rapid changes of the air exchange rates during the day may lead to lower correlations between indoor and outdoor NC concentrations.

### Relationship between outdoor and personal ultrafine particle concentrations

The main requirement for a time-series study of health effects of ambient particulate matter (PM) is that the daily variation in the PM concentration measured at a central monitoring site correlates with the variation in average personal exposure (Zeger et al., 2000). Many studies have demonstrated that individual personal exposures to PM are poorly correlated with ambient concentrations. Wilson and Suh (1997) and Mage et al. (1999) argued that the composition and properties of ambient particles differ substantially from those generated in other microenvironments and that epidemiological studies use central-site ambient PM as a surrogate for exposure to “PM of ambient origin”, not as a surrogate for total PM exposure.

Nowadays a few studies have reported the attempt to estimate the contribution to personal exposures from particles originating outdoors, to quantify exposure errors arising from the use of a central site surrogate, and to understand the effect of such errors on epidemiological conclusions. These results demonstrate the usefulness of separating total personal particle exposures into their ambient and non-ambient components. The results support previous epidemiologic findings using ambient concentrations by demonstrating an association between health outcomes and ambient (outdoor origin) particle exposures but not with non-ambient (indoor origin) particle exposures.

So far, there are no studies on the correlation between the ambient-air and personal-exposure concentrations of ultrafine particles. In our presentation we will show the first results of a pilot study on the relationship between the personal and outdoor ultrafine particle concentrations.

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# Cardiovascular Emergency Calls Associated to Urban Submicron Aerosol Fractions

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Keywords: Health effects of aerosols, submicron particles, size distribution, urban areas

**Introduction:** It is well known that high concentrations of airborne particles are associated with the development of various environment related diseases and the exacerbation of various diseases. Since middle of the nineties epidemiologists found more and more indications that in addition to respiratory illnesses cardiovascular illnesses seem to be associated to airborne particulates. Recently, ultrafine particles have come under special scientific scrutiny. Usually, these particles do not contribute significantly to the mass concentration PM10 but they dominate particle number concentration. Especially, cardiovascular diseases are under suspicion to be evoked and exacerbated by particles significantly smaller than one micrometer.

This study is primarily aiming at two questions:

One aim is to investigate the influence of “rather low” common urban concentrations on the health state of city dwellers, who are not occupationally exposed.

Secondly, this study is quantifying risk differences for selected cardiovascular diseases associated with different size fractions of submicrometer particles, PM2.5 and PM10.

**Material and Methods:** The study was carried out in the City of Leipzig. This city is located in the Leipzig basin with no significant elevations in and around the city. Leipzig has approx. 500,000 inhabitants. There is no significant pollution by industry. In the City of Leipzig urban traffic is a very important source of airborne particles.

Aerosol measurements were carried out at the Leibniz Institute for Tropospheric Research using TDMPMS (twin differential mobility sizer system) working detecting particles with diameters from 3 to 800 nm. The measuring site can be regarded as urban background. Additionally, PM10 and PM2.5 data of public authorities were used for comparison of the health effects of these coarser particles with the effects of the smaller ones.

Cardiovascular emergency calls were selected from the total number of emergency calls for a time period of 12 month within the City of Leipzig.

Therefore, there is no bias produced by the selection of areas within the city.

**Results:** Table 1 lists the urban particle concentrations of different size fractions found during the measuring period.

Size Fractions (Dp nm; PM µg/m <sup>3</sup> )	Mean	Median	Min	Max
Dp<100	12094	10893	1487	34650
100<Dp<500	1919	1723	334	18668
Dp>500	28.54	17.91	2.107	280
Dp<800	14043	13111	2450	35338
PM10	32.48	28.56	6.829	109.7
PM2.5	20.61	18.18	1.375	84.06

Table 1. Statistics of daily averages of particle number concentrations during the measuring period.

In total 5326 cardiovascular emergency calls were used for epidemiologic analysis. Generally, there is no significant difference in incidence between the weekdays and weekends. There are 22.74% and 22.44 % of cardiovascular emergency calls, respectively.

We found:

- a significant positive correlation between the risk for cardiovascular emergency calls and the particle number concentrations,
- a time lag of 1 to 8 days for the health effect of the particles,
- differences in effect for different particle size fractions,
- differences in effect on different cardiovascular diseases.

Significant effects could be found despite of the great inner urban differences in the concentrations of some particle fractions (Tuch *et al.* 2006).

Tuch, Th., Herbarth, O., Franck, U., Peters, A., Wehner, B., Wiedensohler, W., Heintzenberg, J. (2006). *J Expo Sci Environ Epidemiol*, 16, 486-490.

## Health and Particles - Regulatory Aspects

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Keywords: Air Quality Framework Directive, Daughter Directives, national legislation

Science and regulation have different approaches to problems. The scientist finds an open question and will try to find out the facts and the mechanisms behind them in full detail. To fill gaps in knowledge is a welcome task.

The regulator finds a possible problem with many uncertainties. Gaps in knowledge are unwelcome. The questions to be answered are:

- Is there a problem, and do I have enough time to find out whether or not?
- Is there a need for (immediate) action?
- If yes, which kind of action?
- Does this action solve the problem, or does it cause more problems (for me or for others)?

With respect to particles and health, the answer to the first two questions is YES. Time-series studies and cohort studies could demonstrate health effects of particles: An increase in total mortality and hospital admissions, respiratory diseases and symptoms. The data indicate possible carcinogenicity, too. Thus, there is a problem and action is necessary.

Which kind of action is adequate to reach the aim of lowering immission concentrations of particles? Which particles?

The *COUNCIL DIRECTIVE of 15 July 1980 on air quality limit values and guide values for sulphur dioxide and suspended particulates (80/779/EEC)* refers to the determination of black smoke and its conversion into gravimetric units in Annex I. Alternatively, Annex IV refers to gravimetric measurements without specifying the size fraction to be sampled.

During the following years, more data on health effects emerged suggesting smaller particles to be more important. The *COUNCIL DIRECTIVE 96/62/EC of 27 September 1996 on ambient air quality assessment and management (Framework Directive)* listed:

- Fine particulate matter such as soot (including PM<sub>10</sub>) and
- Suspended particulate matter

among the atmospheric pollutants to be taken into consideration.

The *COUNCIL DIRECTIVE 1999/30/EC of 22 April 1999 relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air (First*

*Daughter Directive)* defined limit values for PM<sub>10</sub> with a *Stage 1* to be met by January 2005 and indicative limit values in *Stage 2* foreseen to be met by 2010, but to be reviewed in the light of further information on health and environmental effects, technical feasibility and experience in the application of *Stage 1* limit values in the Member States.

The scientific background was described in the Position Paper *Ambient Air Pollution by Particulate Matter (1997)*. It stated:

“There is increasing evidence that health effects occur at very low levels of PM, and without an apparent threshold. This evidence arises from studies, initially in the US, but which have more recently been carried out in Europe and elsewhere with similar conclusions. These studies have, in general, used different measures of particles, but the majority have used PM<sub>10</sub> and the Group felt that this was the most appropriate measure for a limit value for particles at the present time.”

“Good reasons might be given for considering other fractions rather than just PM<sub>10</sub>, e.g., PM<sub>2.5</sub>, and there might be an increasing need of such kind of measurements in the future. At present knowledge on health effects of particle fractions is insufficient, and sufficiently standardised measurement methods are not available to provide a sound basis for limit values for particle fractions smaller than PM<sub>10</sub>. As this will probably change in the future, limit values set for PM<sub>10</sub> now may have to undergo revision at a later stage.”

It remains difficult to decide if PM<sub>10</sub> or PM<sub>2.5</sub> is the more adequate fraction because only few studies with parallel measurement of both fractions exist (Englert, 2004). WHO (2004) summarises the results of the *Systematic Review of Health Aspects of Air Pollution in Europe* as follows:

“Nevertheless, there is sufficient concern to consider reducing exposure to coarse particles as well as to fine particles. Up to now, coarse and fine particles have been evaluated and regulated together, as the focus has been on PM<sub>10</sub>. However, the two types have different sources and may have different effects, and tend to be poorly correlated in the air. The systematic review therefore recommended that consideration be given to assessing and controlling coarse as well as fine PM. Similarly, ultrafine particles are different in composition, and probably to some extent in effect, from fine and coarse

particles. Nevertheless, their effect on human health has been insufficiently studied to permit a quantitative evaluation of the risks to health of exposure to such particles.”

Regulations should be simple and cost-effective. They have to be proportional and not excessive. It is necessary that compliance can be checked.

A system with limit values for PM<sub>2.5</sub> and PM<sub>10</sub> or PM<sub>2.5</sub> and coarse particles (PM<sub>10-2.5</sub>, CP) in parallel would not be really simple, and a change from PM<sub>10</sub> to another metric only a few years after implementation of PM<sub>10</sub> measurements might not be cost-effective. Another aspect is that the smaller fractions are part of the larger ones, i.e., ultrafine particles (UF) are part of PM<sub>2.5</sub>, and PM<sub>2.5</sub> is part of PM<sub>10</sub> (see Figure 1). Under scientific aspects, a change from PM<sub>10</sub> to PM<sub>2.5</sub> might be justified, but for regulatory purposes this is not necessarily the case considering the great contribution PM<sub>2.5</sub> is making to PM<sub>10</sub>.

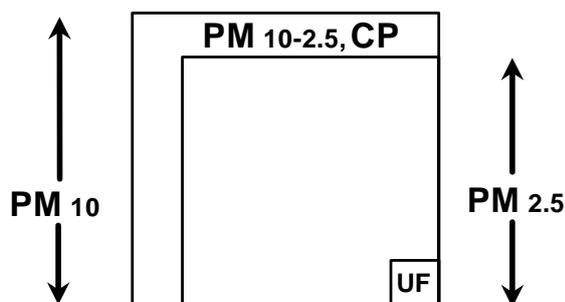


Figure 1. Particulate matter fractions. The ratios of the areas of the squares roughly correspond to the ratios of the concentrations in European cities (adapted from Englert (2004)).

The scientific basis for defining limit values is sufficient for PM<sub>10</sub> and for PM<sub>2.5</sub>, too, but at present not for CP and even less for ultrafine particles. The situation with respect to measurement equipment and intercomparisons is similar.

However, a special problem for regulators is the fact that no “safe” PM level can be defined. Which size of remaining risk has to be tolerated is difficult to decide, and cost-benefit analyses can only shed light on some of the aspects but cannot provide really “objective” criteria, one reason being the comparison of real costs and monetised values of statistical life.

Scientists usually favour differentiation, but a system based on several limit values in parallel is not easily understood by the public (and maybe not only by the public). The proposal made by the 1997 Position Paper Group was a 24-hour PM<sub>10</sub> limit value of 50 µg/m<sup>3</sup> as a 98-Percentile (i.e. a maximum of 7 exceedances) combined with an annual PM<sub>10</sub> mean of 20 µg/m<sup>3</sup>. In this case, short-term and long-term values are roughly equivalent. This

proposal was adopted as *Stage 2*. For *Stage 1*, however, the 24-hour PM<sub>10</sub> limit value of 50 µg/m<sup>3</sup> with a maximum of 35 exceedances, which would be equivalent to an annual mean of 30 µg/m<sup>3</sup>, was linked with an annual mean of 40 µg/m<sup>3</sup>. The result was as could be expected: The annual mean can be met much easier than the 24-hour mean. Unfortunately, the noncompliance with the 24-hour limit value may give the misleading message that health problems are mainly due to short-term changes in air pollution.

Keeping in mind future developments with the possible inclusion of ultrafine particles, the way how to make the system simple and cost-effective is far from being found. A Directive combining Framework Directive and Daughter Directives is currently in preparation and will probably supplement PM<sub>10</sub> *Stage 1* values with PM<sub>2.5</sub> targets. Inclusion of ultrafine particles (number concentration) will be reserved to future revisions.

And future revisions will come. Current air pollution legislation is based on European Directives. It is a long way from a first draft to the final adoption of a Directive, and the process of transfer to national legislation also needs time, usually at least two years. Meanwhile, new scientific results emerge, inducing a call for revision. This process probably will continue.

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## Health effects of inhaled ultrafine particles in the lungs and other secondary target organs like brain and heart.

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Keywords: ultrafine particle, nanoparticle, translocation into circulation, nanoparticle biokinetics, nanoparticle accumulation in secondary target organs.

Nanoparticles are increasingly used in a wide range of applications in science, technology and medicine. Since they are produced for specific purposes which cannot be met by larger particles and bulk material they are likely to be highly reactive, in particular, with biological systems. On the other hand a large body of know-how in environmental sciences is available from toxicological effects of ultrafine particles after inhalation. Since nanoparticles feature similar reactivity as ultrafine particles a sustainable development of new emerging nanoparticles is required.

Cardio-vascular effects observed in epidemiological studies triggered the discussion on enhanced translocation of ultrafine particles from the respiratory epithelium towards circulation and subsequent target organs, like heart, liver, spleen and brain, eventually causing adverse effects on cardiac function and blood coagulation, as well as on functions of the central nervous system. There is clear evidence that nanoparticles can cross body membranes and reach and accumulate in the above mentioned secondary target organs.

To quantitatively determine accumulated fractions in such organs the ultimate aim is to quantitatively balance the fractions of nanoparticles in all interesting organs and tissues of the body including the remainder body and total excretion collected between application and autopsy. Substantial uncertainty remains if only selected organs are analyzed. Furthermore, in case of analysis based on a label (radioactive, fluorescent, magnetic, etc.) firm fixation of the label to the nanoparticles need to be demonstrated. Since these gross determinations of nanoparticle contents in organs and tissues do not provide microscopic information on the anatomical and cellular location of nanoparticles such studies are recommended to be complemented by electron microscopy analysis.

In addition, the role of particle parameters determining this translocation dynamics remains to be not fully understood. Nanoparticle parameters such as size, hydro- / lipophilicity, surface charge, surface ligands and their possible exchange in various body fluids need to be considered. The current knowledge on systemic translocation of ultrafine particles in man and animal models and an estimate of accumulating particle number, surface area and mass in secondary target organs during short-term and chronic exposure will be discussed in order to demonstrate the relevance of translocated fractions of nanoparticles.

This work was supported by the CEC (FIGD-CT-2000-00053), EU FP6 PARTICLE\_RISK 012912 (NEST), and U.S. NIH Grant HL070542.

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# Ultrafine Particle (UFP) Measurements and Modelling

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Session “Modelling Fine and Ultrafine Particles” on October 24, 2007.

Keywords: UFP measurement, UFP modelling, transport, aerosol dynamical processes

During the past years, there has been a growing interest in ultrafine particles (UFP) that are encountered in our environment. According to the most used convention, the class of UFPs includes airborne particles with diameters less than 100 nm.

Several properties of environmental UFPs have attracted the concern of environmental health research: Their small size and therefore deep entrance into the human lung; their capability to travel from the respiratory system into practically all organs of the body including the brain; their increased fraction of relatively insoluble material and therefore increased resistance against clearance; their relatively high abundance in terms of particle number concentration especially in urban areas; their specific surface area compared to a mass-equivalent of larger particles; their increased fraction of soot and its associated toxic micropollutants; their relatively long life-time in the atmosphere due to inefficient wash-out and sedimentation.

It is of a natural consequence that the sources, the sinks as well as the entire lifecycle of environmental UFPs need to be explored in order to better assess the human exposure to these particles and to evaluate the associated risks.

Important contributions of environmental UFPs originate from primary sources, such as high temperature combustion, as well as secondary sources, such as gas-to-particle conversion. It appears that within cities and larger conurbations, the background of primary emissions, such as from industry and traffic, dominates the average concentrations of UFPs while in the continental background atmosphere, their mean concentrations are overwhelmingly controlled by secondary particle formation. In the background atmosphere, UFPs have been shown to originate at large numbers at diameters < 10 nm from gas-to-particle formation. Stable particles at sizes between 1 and 2 nm can be formed from charged or neutral molecular clusters (Kulmala et al. 2007).

UFP measurements in Europe have been summarised for rural and urban locations by van Dingenen et al. (2004), whereas for Germany a survey has been given by Birmili et al. (2006).

“Modelling ultrafine particles” can refer to, either the simulation of particle transport near their sources (Vardoulakis et al., 2003), the evolution of the aerosol population as a result of dynamic processes, or the combination of both.

Aerosol dynamical processes are relevant for the evolution of new particles resulting from secondary particle formation, but also for the primary emission processes. The most important atmospheric processes include the nucleation (formation) of new particles, condensation of vapours onto pre-existing particles, coagulation between particles, and particle deposition (e.g., Friedlander 2000). Downstream primary particle sources such as traffic, particles may undergo coagulation and also chemical processes.

This presentation will summarize some main findings during UFP observations, and outline parallels to the achievements of model simulations:

- UFP observations
- Primary UFPs, mainly from motor traffic
- Secondary UFPs
- Aerosol dynamical processes in the atmosphere
- The lifetime of UFPs in the atmosphere
- Indoor UFPs

Birmili, W. and 11 others (2006) *Räumlich-zeitliche Verteilung, Eigenschaften und Verhalten ultrafeiner Aerosolpartikel (< 100 nm) in der Atmosphäre, sowie die Entwicklung von Empfehlungen zu ihrer systematischen Überwachung in Deutschland*. 93 p. UBA-Texte, 26, on-line at <http://www.umwelt-daten.de>.

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# Modelling ultrafine particles in urban environments

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Keywords: ultrafine particles, traffic emissions, aerosol modelling

## INTRODUCTION

Ultrafine particles (UFP) and their sources and fate in the atmosphere continue to be key subjects in the atmospheric research due to concerns about the effects on human health and global climate. Over the recent years intensive measuring activities were conducted to characterise the particle size distribution and composition at different ambient and indoor locations, including kerbside, urban background, near-city and rural level. Traffic emissions could be identified as a dominating source of UFP (aerodynamic diameter less than  $0.1\mu\text{m}$ ). This can be seen by the large gradient in particle concentration between rural and kerbside locations shown in figure 1. Particle number emission factors and size distribution could be estimated for real world driving conditions (Ketzel et al., 2003) and for soot particles (Rose et al. 2006).

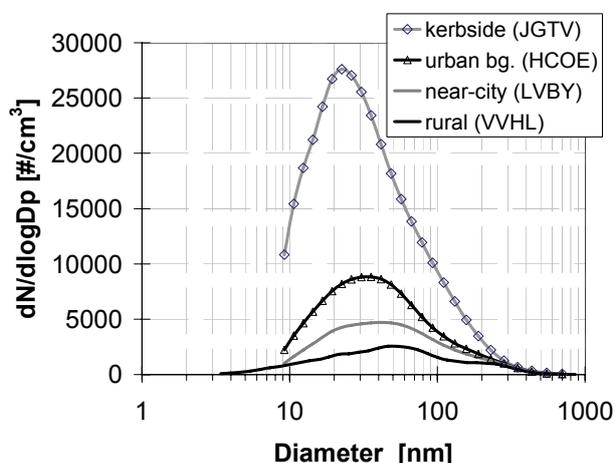


Figure 1. Measured particle size distribution at several locations in Denmark and Southern Sweden. (Based on Ketzel et al. 2004)

## METHODS

Here we apply these emission factors together with a street pollution model OSPM (Berkowicz, 2000) to predict time series of total particle number and NOx at street level. Moreover we identify the relevant particle dynamic processes acting at

different time scales. We describe a coupling of a plume dispersion model and a sectional aerosol dynamic model that is able to describe the evolution of the particle size distribution on the way from the exhaust to the urban background (Ketzel et al. 2005).

## CONCLUSIONS

The time scales for particle dynamic processes (e.g. coagulation, deposition, dilution, condensation) are estimated for urban street and background environment and discussed in their relevance for influencing the particle size distribution. In agreement with the literature it can be concluded that coagulation is too slow to alter the size distribution in the exhaust plume and dilution is the dominating process. A similar conclusion can be drawn for the street scale. In a more confined environment as e.g. a road tunnel the removal processes coagulation and deposition might play an important role.

We show that for model predictions at street level the total particle number can be treated as ‘inert tracer’, i.e. particle transformation has not to be modelled in detail and rather considered as part of the emission process. Using OSPM we obtain high correlation ( $R=0.8$ ) between measured and modelled total particle number assuming constant emission factors (Figure 2). If we account for a temperature dependence of the particle emissions the model results improve slightly ( $R=0.82$ ).

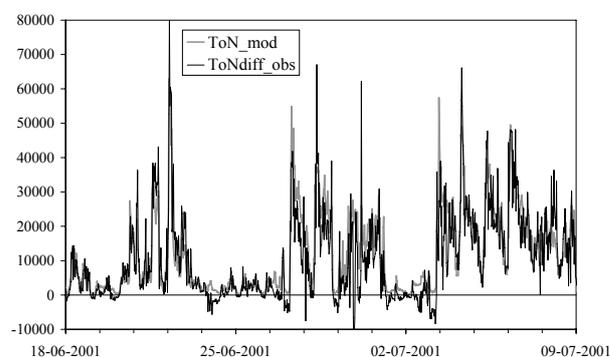


Figure 2. Time series of OSPM model predictions (grey) and measurements (black line) for total number concentration ToN [ $\text{cm}^{-3}$ ] at a kerbside (Jagtvej) for a period of 3 weeks.

The modelling of the full particle size distribution at urban scale shows that 1) the coagulation and deposition account for ca. 25% loss of particles in the urban background compared to street level and 2) the size dependent removal lead to a slight increase in 'peak' - diameter of the particle size distribution from ca. 25nm to 30nm, see figure 2. Both effects are in agreement with the observation.

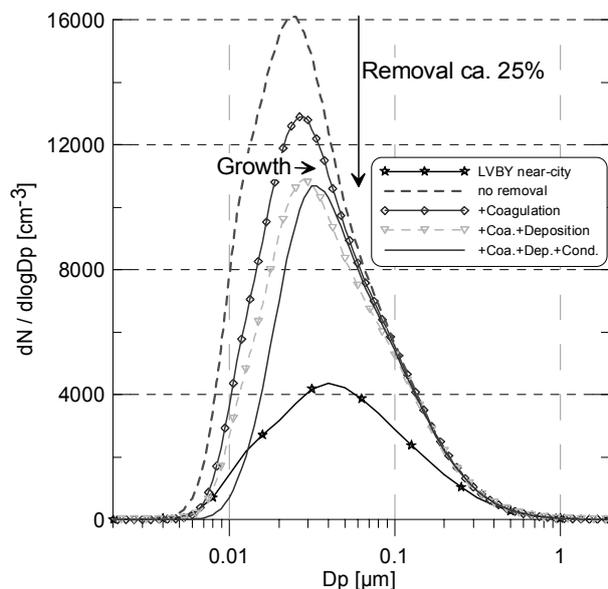


Figure 3. Modelled particle size distribution using the measured near-city distribution as background. (Based on Ketzel et al. 2005)

#### ACKNOWLEDGEMENTS

This work was supported by the Danish Research Agency and the Danish EPA under the Danish Ministry of Environment. The authors thank Prof. Erik Swietlicki and Dr. Adam Kristensson for sharing their experimental data with us.

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## Modelling PM<sub>x</sub>-Emissions and –Concentrations of streets for environmental impact assessments and action plans

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Keywords: Particular matter, vehicle emission, action plan, re-suspension.

The field measurements in the vicinity of roads and in streets canyons in Germany show at some locations an exceedance of the limit values for PM<sub>10</sub> of the EU Directive 1999/30/EU respective 22. BImSchV (2007). Therefore action plans and PM<sub>10</sub>-modelling for environmental impact assessments are needed.

There exist a large number of modelling tools for the determination of PM concentrations. In the internet a user specific selection on the basis of a catalogue of criteria is possible for example using the "Model Documentation System (MDS)" of the European Topic Centre on Air and Climate Change<sup>1</sup>. Usually, in the frame of action plans and environmental impact assessments for the determination of PM<sub>x</sub> concentrations, models are used, which do not directly account for the formation or interaction of particles. In the near field of roads, this is acceptable because of the large time scales of these effects and the restriction to PM<sub>10</sub>. These simplified and cost efficient procedures use the information about the large- and regional scale background concentration from the results of appropriate monitoring stations in the open countryside. The concentrations caused by the emissions of the city are either also taken from monitoring stations or are calculated on the basis of the cities emissions (traffic, industrial and domestic sources). Traffic induced hot-spot concentrations can be detected on the basis of screening or more sophisticated models, in Germany frequently applied are LASAT, PROKAS, IMMIS, MISKAM, AUSTAL.

PM emissions from traffic are a main source of ambient concentrations especially at hot spots in urban environment. Traffic PM emissions can be subdivided into three main groups:

- A) direct exhaust emissions,
- B) direct emissions other than exhaust as e.g. from brakes and clutches, and
- C) indirect or re-suspended PM emissions from the tyre/road interface.

A) Direct exhaust emissions are predominantly found in the fine fraction (PM<sub>2.5</sub>) and are basically a function of the vehicle engine and driving patterns, but not weather or road conditions (EMEP/CORINAIR, 2003, UBA 2004). They are measured in laboratories and documented in different emission databases (e.g. COPERT 2006, UBA 2004,

EMEP/CORINAIR 2003). In Germany is used the emission database of UBA (2004).

B) Emissions from wear of brakes and clutches consist to about equal parts of the fine and coarse (PM<sub>10</sub>-PM<sub>2.5</sub>) fraction (Garg et al., 2000, EMEP/CORINAIR, 2003) and correlate well with the direct emissions and other vehicle emissions e.g. NO<sub>x</sub> (Wählin et al., 2006). For these emissions there are less measurements available and they are for example not included in the UBA emission databases (UBA, 2004).

C) Emissions from road abrasion, tyre wear and road dust re-suspension are found partly in the fine but mostly in the coarse fraction. This PM source is often less correlated with the exhaust emission due to an influence from 'external factors' such as tyre type, vehicle induced turbulence, road and weather condition (Gustafsson et al., 2005; Kupiainen et al., 2005; Norman and Johansson, 2006; Johansson et al., 2006). These external factors provide a major challenge for the estimation of this type of emissions and presently much research is undertaken to elucidate this PM source.

The findings of some research projects (e.g. Lohmeyer et al., 2004) together with results of Gehrig et al. (2003) led to the procedure suggested in Table 1. This method to display the results follows the general procedure of the INFRAS emission factor handbook (UBA, 2004) for all exhaust emissions and the procedure of Gehrig et al. (2003), which is to provide the emission factors as a function of the so called "traffic situations", e.g. motorway, city main roads, city slow traffic etc.. For a description of these traffic situations see Table 1 or UBA (2004).

The data indicate that non-exhaust emissions are also increased for low average travelling speed due to more unsteady driving conditions and the more frequent acceleration / deceleration cycles.

At most of the locations in the studies of Lohmeyer et al. (2004) and Gehrig et al. (2003) the vehicle speed is 40-50 km/h only one location with slightly higher and one with slightly lower speed. Due to the limited range of vehicle speeds in our data

<sup>1</sup> <http://air-climate.eionet.eu.int/databases/MDS/index.html>

and the many other influencing factors this issue is not discussed any further within this studies.

Finally it can be concluded, that in fact the presently available tools for the quantification of emission and dispersion allow a determination of

PM-concentrations for assessments and action plans but the uncertainties are still large and important physical dependencies are insufficiently determined or absolutely unknown. In this respect, a large need of research and development exists.

Table 1: Simplified version of the proposed German PM<sub>10</sub> emission factors for non-exhaust emissions (last two columns) in dependence of the traffic situation. Values for a fleet mix containing 4% heavy duty traffic and exhaust emissions according to (UBA, 2004) are given. Table adapted from Ketzel et al. (2007).

Traffic situation	average Speed [km/h]	Share of constant speed driving[%]	exhaust emiss. factor (fleet-mix) [mg/km veh]	non-exhaust emission factor (fleet-mix) [mg/km veh]	non exhaust emission factor*	
					cars / vans	trucks
motorways or outside cities	60-130				22	200
Tunnel	60-100				10	200
City main road (HVS1)**	56	46	19	29	22	200
City main road (HVS2)**	44	52	20	41	30	300
City main road (HVS3)**	34	44	22	54	40	380
City main road (HVS4)**	28	37	26	66	50	450
City traffic lights (LSA2)**	24	32	28	82	60	600
City slow traffic (IO_Kern)**	17	23	32	118	90	800

\* Values for good quality of the road surface, flat terrain and conditions of rain as usual in Germany.

\*\* Speed limit = 50 km/h;

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## Five years ultrafine and fine ambient particles number concentrations measurements at a traffic-orientated site in Dresden

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Keywords: Ultrafine particles, number concentration, air quality, monitoring, traffic

The European Union decided a series of directives to improve the monitoring and evaluation of the air quality and inform the public. In Saxony, the concentration of most pollutants could be reduced successfully in ambient air for the last 15 years. The authorities tasks of monitoring could be reduced (SO<sub>2</sub>) or completely stopped (TSP).

Findings of epidemiology studies refer to the health-relevant meaning of ultrafine aerosol particles (<100 nm). The State Agency for Environment and Geology decided to document the air quality situation for such ultrafine particles at a single observation site in Saxony. This measurement is optional without legal base according to precautionary principle. A traffic-orientated measuring site was chosen to collect air quality data for many years, while amongst others the fraction of low emission vehicles will increase.

The intention from the beginning was to integrate the measurement into the air quality monitoring network in Saxony. A project with the Leibniz Institute for Tropospheric Research in Leipzig covers not only the setup and installation of the measuring technique, but also the training of the personnel and the quality assurance of the data. The measurements are performed with a TDMPS (Twin Differential Mobility Particle Sizer). In two separate ranges, particle number size distributions are measured from 3 to 800 nm. The measured data are reduced to 8 particle size classes and ½-h-average values. The data are validated and administered like other air pollutants in the data base and wide-area evaluations can be made.

The measuring container "Dresden-Nord" is situated on an open square near the railway station

Dresden Neustadt. The container is placed 5 m from a road and 50 m to a busy crossing (about 50.000 vehicles per day with 5% heavy traffic). The sampling point height is 3.5 m. Table 1 contains characteristic values of the pollution levels from this point for the last 5 years. The average number concentration of particles in the range from 3 to 800 nm amounted to 22.923 cm<sup>-3</sup>. The concentration of the ultrafine particles (3 - 100 nm) was determined to 20.055 cm<sup>-3</sup>. The ultrafine particles represent 87% of all particles. Results for particle classes are listed in table 2.

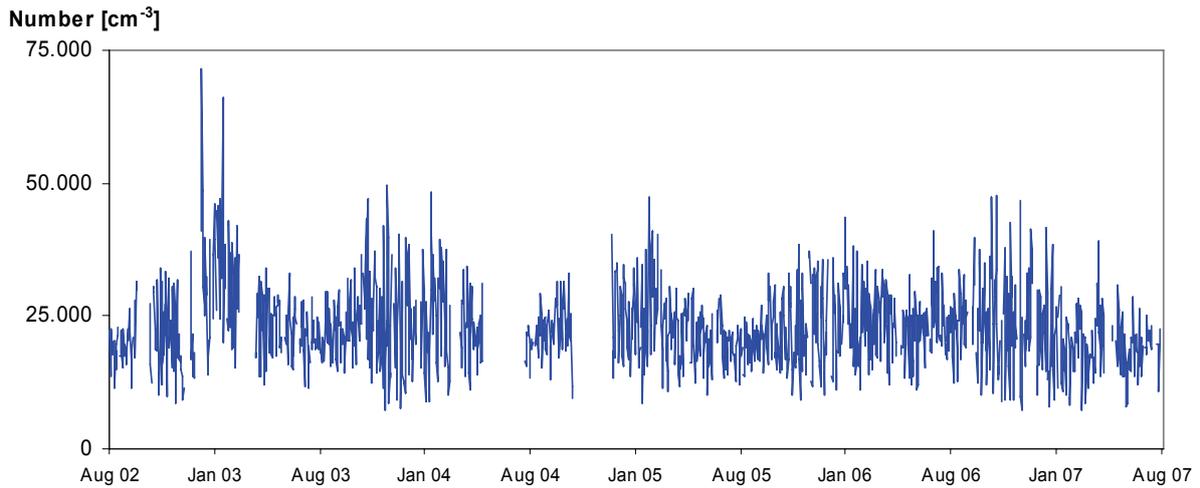
The evolution of daily averages of particle number concentrations from 3 to 800 nm is shown in fig. 1. A large variation of the daily values is demonstrated. The data availability is 80%.

Safe statistic data can be made over 5 years. Fig. 2 shows an example. The middle reduction of the load can be illustrated with the mean value of all Sundays in relation to the mean value of all Mondays until Fridays. It is shown, how the average load can be reduced, if the economic life is reduced to the Sunday-level.

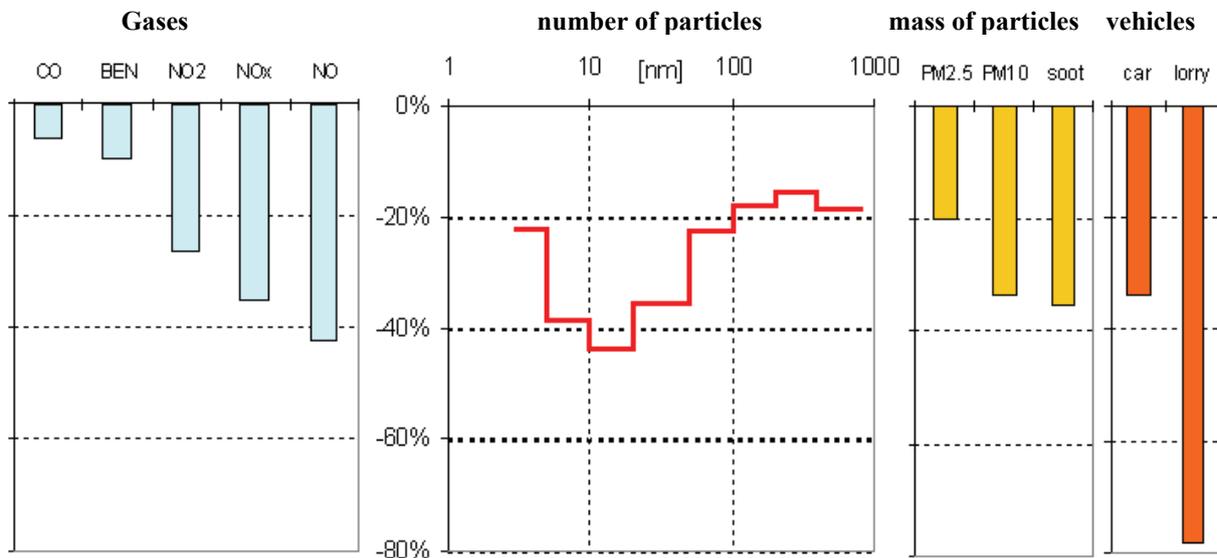
The presented data set provides the first long term measurements of number size distributions within a regular monitoring network. The data are available for epidemiological investigations now.

A need for a mobile aerosol standard for examination in the measuring station is seen for the further improvement of the quality assurance.

Thanks are given to the staffs of the Environmental Operating Company (UBG) in Radebeul for the special measurements.



**Fig. 1:** The variation of the daily averages for the number concentration of particles from 3 to 800 nm at a traffic-orientated site in Dresden over 5 years.



**Fig. 2:** Mean percentile reduction on Sundays opposite on mean from Mondays to Fridays over 5 years for gases (left), number of particles for 8 particle classes from 3 to 800 nm (middle), particle mass of different pollutants (middle-right) at traffic-orientated measuring station “Dresden-Nord” and number of vehicles (quite right) on a near vehicle counting point.

**Table 1:** Pollutant concentration at the measuring point from 1<sup>st</sup> August 2002 to 31<sup>st</sup> July 2007

Pollutant	PM <sub>10</sub> [µg/m <sup>3</sup> ]	PM <sub>2.5</sub> [µg/m <sup>3</sup> ]	Soot [µg/m <sup>3</sup> ]	NO <sub>2</sub> [µg/m <sup>3</sup> ]	Benzene [µg/m <sup>3</sup> ]	CO [mg/m <sup>3</sup> ]
5 year average	33	21	4,4	46	2,4	0,6
98-percentil <sup>1</sup>	80	56	9,3	72	6,2	1,3

**Table 2:** Particles concentration in particles/cm<sup>3</sup> at the measuring point from 1<sup>st</sup> August 2002 to 31<sup>st</sup> July 2007

Particles sizes class	3 – 5 nm	5 – 10 nm	10 – 20 nm	20 – 50 nm	50 – 100 nm	100 – 200 nm	200 – 400 nm	400 – 800 nm
5 year average	603	3.179	6.427	6.209	3.685	2.162	672	68
98-percentil <sup>1</sup>	1.421	6.590	13.791	12.587	6.544	3.016	1.382	214

<sup>1</sup> related to daily averages

# Ultrafine particles in NRW – case studies in urban background and at an „Autobahn“

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Keywords: SMPS measurements, weekly profiles, traffic emissions

The potential impact of ultrafine particles (UFP) on human health is increasingly discussed in recent years. This paper reports about UFP measurements made at two background sites (urban and suburban, resp., distance approx. 8 km, Duisburg area, (Quass et al. 2004)) and upwind/downwind of a highly trafficked motorway (A61 near Meckenheim, (Quass et al 2007)). All measurements were made using SMPS/CNC (TSI). PM<sub>10</sub> mass concentrations were measured by TEOM systems.

## Urban/suburban background measurements

At urban background, the particle number concentration (PNC) of UFP (particle diameter < 100 nm) show distinct dependence on the day of week for rush-hour conditions (see fig. 1), while being less variable at midnight.

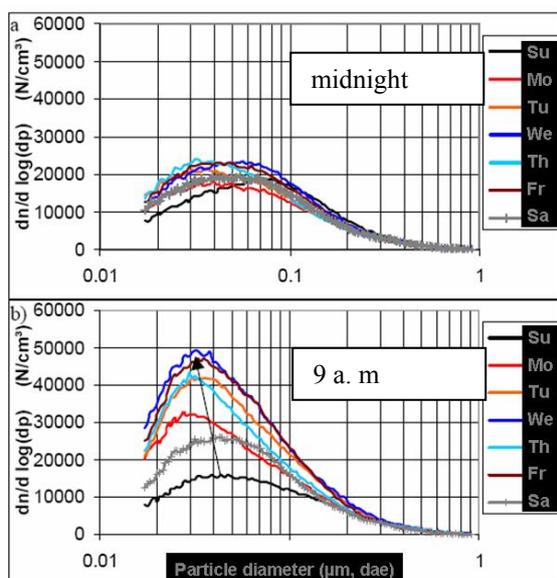


Figure 1: Size-resolved number concentration of the urban background aerosol for different weekdays and two daytimes (Averages of a 3-month period)

During rush hours, a slight shift of the modal diameter was observed with increasing concentrations indicating a higher fraction of freshly produced combustion particles. Peak concentration increased during the week with highest values on Wednesday and Friday. For that daytime (09:00) a.m. this weekly profile could be found also for PM<sub>10</sub> concentrations and the correlation between PM<sub>10</sub> and the modal peak concentration was significant

( $r^2 \approx 0.7$ ). However, at other times, this correlation was much lower (midnight:  $r^2 \approx 0.1$ ). This clearly shows that PM<sub>10</sub> concentrations temporarily may significantly correlate with UFP concentrations, e.g. during situations with high traffic emission rates. With respect to the question of spatial representativeness of UFP measurements a comparison of UFP concentration data measured at the suburban and urban site was made. Increasing correlations were obtained for longer averaging times, but even the highest  $r^2$  values did not reach those usually obtained for PM<sub>10</sub> or PM<sub>2.5</sub>. Clearly, UFP concentrations are influenced more by local processes (sources, short atmospheric lifetime) than PM<sub>10</sub>, which has to be accounted for in e.g. epidemiologic study designs.

Data averaging time	30 min	6 h	24 h
$r^2$	0.35	0.50	0.54

Table 1: inter-site correlation coefficients for different data averaging times (total UFP <100nm).

## Highway measurements

Within a measurement program mainly intended to derive emission factors for non-exhaust emissions a campaign with upwind-downwind SMPS measurements was carried out. During this period the average traffic volume was ca. 2.200 vehicles/h with 16% high duty vehicles.

Figure 2 shows the obtained number size distribution and the downwind/upwind ratios. The traffic emissions affect the particle concentrations throughout the entire particle size spectrum. Changes of the size distribution (new modes), indicated by a change of the downwind/upwind ratio, could be observed for the particle size ranges of about 100 nm and for particles below 30 nm. While the first new mode most probably consisted of soot particles, the latter may be interpreted as fresh nucleation particles originating from unburned or only partially combusted fuel.

This was further underlined by a correlation analysis of downwind-upwind concentration differences for particles numbers and nitrogen oxides.  $R^2$  values were calculated for the sum of PNC in each size range given in table 2. In the size range below 50 nm higher correlation of PNC vs. NO and NO<sub>x</sub> was found than PNC vs. NO<sub>2</sub>. On the contrary, NO<sub>2</sub> correlated better with PNC for larger particles (50-200 nm).

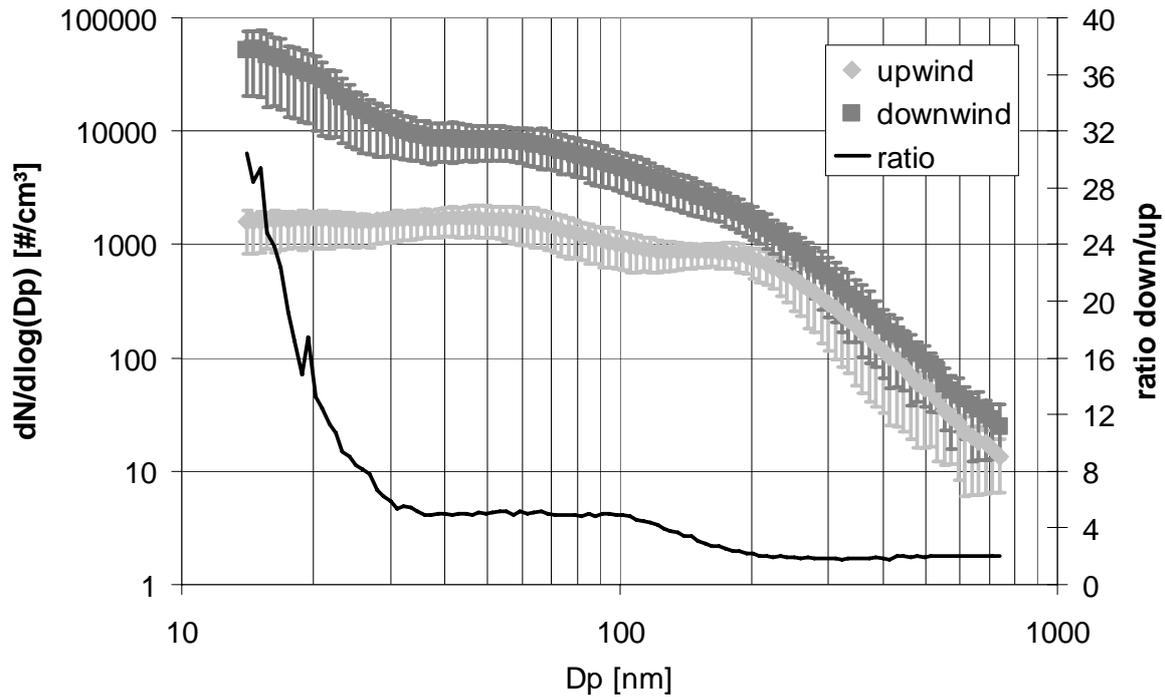


Figure 2: Upwind and downwind PNC size distribution (mean and 25/75 percentiles) and downwind/upwind ratio (N=371 measurements, 0.5 h averages)

	<50 nm	UFP	50-200 nm	200-500 nm	Total Number
NO <sub>2</sub>	0.42	0.46	0.69	0.23	0.47
NO <sub>x</sub>	0.62	0.66	0.67	0.32	0.67
NO	0.63	0.66	0.60	0.32	0.67
PM <sub>1</sub>	0.51	0.54	0.52	0.13	0.55
PM <sub>10</sub>	0.25	0.25	0.16	0.02	0.25
PM <sub>1-10</sub>	0.06	0.06	0.06	0.00	0.06

Table 2:  $r^2$  correlation coefficients between downwind-upwind differences of PNCs (for 3 size ranges) and of various pollutant concentrations

$R^2$ -coefficients with PM concentrations are lower but still indicate correlation in case of PM<sub>1</sub>, whereas PM<sub>10</sub> and PM<sub>1-10</sub> do not correlate at all with ultrafine and fine PNCs.

Comparing the size distributions and PNCs in the urban background with those obtained at the highway considerable differences are obvious:

- The pronounced size mode around 30-50 nm observed in the urban aerosol is not found near the highway. There, smaller particles <30 nm have a relatively higher concentration
- Above 30 nm, PNCs even downwind the highway do not reach the concentrations found in urban environment. This is probably due to the low background contribution (1 order of magnitude lower than midnight concentration in the city) and good dispersion conditions at the investigated highway site.

### Summary and conclusions

Particle numbers and number size distributions measured at urban background and near a highway in rural environment are presented. The urban aerosol shows a distinct daytime dependence and a weekly variation partially correlating to that of PM<sub>10</sub>. Correlation with PM concentrations is significant only in high emission situations (rush hour). The spatial representativeness is weaker than for PM due to higher importance of local sources and lower particle lifetime. The modal diameter is usually found between 30 and 60 nm. This mode was also present in the aerosol downwind the highway, however a strong mode of finer particles could be observed additionally. Number concentrations of this mode better correlate with NO than with NO<sub>2</sub>, indicating fresh particle production.

### Acknowledgements

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## Ultrafine and fine particle measurements in Switzerland at various stations and on different roads

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Keywords: mobile measurements, aerosol mass spectrometry, <sup>14</sup>C-analysis, FMPS (fast mobility particle sizer)

In the last 2-3 years several field campaigns have been performed in Switzerland to study the aerosol composition their sources and the aerosol number size distributions (Figure 1). Some campaigns were performed at fixed locations in urban, rural, and Alpine areas. In addition, mobile measurements allowed the characterisation of the spatial distribution of the chemical composition and the aerosol size distribution.

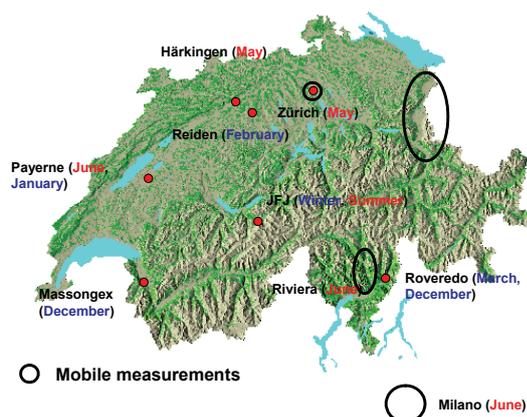


Figure 1. Locations of the Aerodyne aerosol mass spectrometer measurements from 2005-2007 in Switzerland.

The main instrumentation discussed in this presentation were the Aerodyne aerosol mass spectrometer to detect the volatile PM1 components, the <sup>14</sup>C analysis for the quantification of the fossil and non-fossil EC and OC fractions and the TSI fast mobility particle sizer to characterize the aerosol number size distribution from 5.6 to 560 nanometers.

Measurements in Roveredo (Alpine valley along the Transalpine San Bernardino Route) showed that the particulate matter, especially the organic mass is strongly dominated by wood burning particles. This could be shown both by the aerosol mass spectrometer measurements and the <sup>14</sup>C analyses (Alfarra *et al.*, 2007, Szidat *et al.*, 2007).

The results of the studies in the Alpine valley are however not representative for the rest of Switzerland. On the Swiss Plateau, secondary

inorganics ammonium nitrate (especially in winter) and ammonium sulfate are the main fraction of PM1. The elemental carbon is mostly due to fossil sources (especially traffic) (Szidat *et al.*, 2006). The organics in Zürich summer were mostly secondary (2/3) and wood burning, charbroiling and wood burning were similarly important (Lanz *et al.*, 2007a). In winter, surprisingly also half of the organics are secondary, and wood burning is about 3 times more important than traffic (Lanz *et al.*, 2007b). The secondary organics in winter and summer are non-fossil, probably coming mostly from terpene and isoprene emissions in summer and volatile wood burning emissions in winter.

Mobile measurements in various locations have shown that the aerosol volume ( $D < 560$  nm) is often highest in villages (especially in winter) whereas the nanoparticles ( $D < 50$  nm) are by far highest on the highway due to the high load and high fractions of diesel vehicles.

In summary, wood burning is very important in Switzerland during winter for the particulate mass. Traffic contributes significantly aerosol number (especially nanoparticles), to elemental carbon also to ammonium nitrate (due to the NO<sub>x</sub> emissions).

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## Temporal and spatial variability of sub- $\mu\text{m}$ aerosol concentrations in the urban atmosphere of Leipzig

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Session "Measurement of UFPs in ambient air II" on October 24, 2007.

Keywords: particle exposure, spatial variability, traffic aerosols, UFP exposure

### INTRODUCTION

Scientific evidence has consolidated associations between environmental aerosols (also termed "PM", particulate matter) and negative health effects, such as asthma and cardiopulmonary disease (WHO, 2004). While there is little debate about whether these effects are real, numerous questions have arisen which particular types (i.e., sub-fractions) of environmental particles are responsible for the health risk, and how the exposure risks for the population may be reduced in a most efficient way (HEL, 2002).

In urban environments, fine ( $< 1 \mu\text{m}$ ) and ultrafine ( $< 0.1 \mu\text{m}$ ) particles, as well as number concentration of particles, have moved into the focus of public health interests. The rationale is that ultrafine particles are inhalable into the deep lung, contain increased amounts of soot, aromatic organics, and trace metals, and account for the majority of particle number in urban atmospheres.

Current estimates of exposure to fine and ultrafine particles, however, are particularly hampered by the lack of knowledge when characterising the spatial and temporal variability of fine and ultrafine aerosols within urban landscapes. Several studies have indeed suggested that especially ultrafine particles are highly variable with location (Hussein et al., 2005, Tuch et al., 2006). Before particle metrics including fine and ultrafine particles can be considered for a systematic monitoring, a broader scientific understanding of the behaviour of these aerosol properties in an urban atmosphere is required.

### FIELD EXPERIMENT

A series of field experiments ("PURAT" – Particles in the urban atmosphere: Behaviour of fine and ultrafine particles, their spatial variation and relationships with local policy action) was conceived to investigate the spatial and temporal distribution of urban aerosols on different urban length scales.

Long-term measurements between 2004 and 2005 in a street canyon and at an urban background site in

Leipzig, Germany (population ca. 500,000) were used to evaluate the general impact of traffic density and meteorological processes on urban aerosol concentrations (PURAT-0).

A second experiment (PURAT-1) involved simultaneous particle size distribution measurements at 6 fixed observation sites in Leipzig: Five out of the six fixed measurement sites were within a distance of 3 km in the downwind plume of Leipzig's city centre. The character of the observation sites spanned a range between two "roadside" sites (i.e., distance to a major road  $< 10 \text{ m}$ ), and four urban background sites. In addition, a mobile aerosol laboratory was deployed to measure number size distributions at additional 7<sup>th</sup> site.

Particle size distributions were determined using various scanning mobility analysers (SMPS). In order to avoid systematic bias due to instrumental differences regular intercomparisons were made between individual SMPS systems before and during the experiment. Individual instruments showed systematic differences in their readings, and this needed to be taken into account when evaluating the final size distribution data set.

### RESULTS

An example for the observed variations in particle size distributions during PURAT-1 is given in Figure 1, showing simultaneously measured size distributions at 7 measurement sites. As expected, particle concentrations were generally increased near sources of motorised traffic. This concerned especially the size range of ultrafine particles ( $< 100 \text{ nm}$ ). However, significant differences were also observed between urban background sites.

We next examined the correlation of particle number size distributions between different locations by evaluating linear correlations of particle number concentrations. Figure 2 shows the measures of determination ( $R^2$ ) obtained for the particle size interval 40-120 nm (Aitken particles).

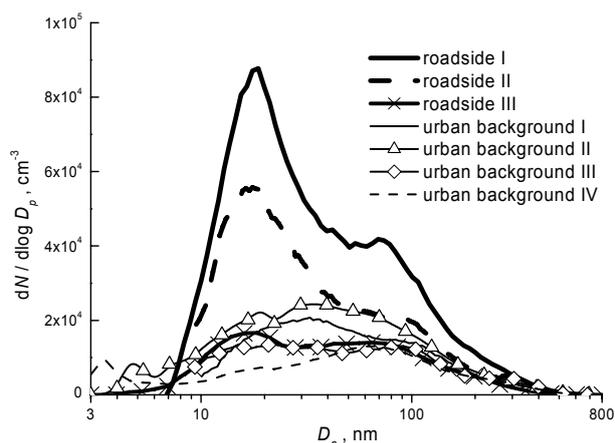


Figure 1: Particle size distributions measured simultaneously at seven urban sampling locations. Exemplary data from a morning rush hour episode in Leipzig, April 13, 2005, 0530-0700 h.

	RS I	RS III	UB I	UB II	UB III	UB IV
RS I	1	0.42	0.61	0.53	0.53	0.43
RS III	0.42	1	0.67	0.52	0.66	0.57
UB I	0.61	0.67	1	0.87	0.90	0.72
UB II	0.53	0.52	0.87	1	0.87	0.66
UB III	0.53	0.66	0.90	0.87	1	0.72
UB IV	0.43	0.57	0.72	0.66	0.72	1

Figure 2: Measure of determination ( $R^2$ ) of linear correlations between particle number concentrations (size interval 40-120 nm) at 6 different urban observation sites. “RS” denotes the roadside, “UB” the urban background sites referred to in Figure 1. Data coverage is April 7, to May 8, 2005.

The general conclusions from the correlation matrixes were:

- Aitken particle concentrations (40-120 nm) correlate well between urban background sites. (The best correlation was found between two neighbouring urban background sites.)
- Particle concentrations at roadside sites correlate less with urban background concentrations, and eventually even less between different roadside sites.
- Correlations increase for accumulation mode particles (> 120 nm), but decrease for nucleation mode particles (10-40 nm).
- Concentrations at all particle sizes correlate positively with the density of traffic in the proximity of the measurement sites.

Besides the results obtained from the experiment PURAT-1, this presentation will also give a summary of the long-term dual point measurements (PURAT-0) as well as of a subsequent experiment PURAT-3, which concentrated on an even smaller transport length scale 1-100 m.

For the region around the Eisenbahnstrasse street canyon in Leipzig, microscale transport simulations of traffic aerosols will be presented.

## CONCLUSIONS

During multiple site experiments in Leipzig, the spatial and temporal variations of aerosol particle number distributions were characterised across a range of several observation sites. The results demonstrate that UPFs tend to be much more variable within the complex terrain of a city than, for instance particle mass.

The impact of localised and time-dependent traffic sources in an urban landscape causes poor correlations between roadside and urban background observations. The best correlations in Aitken mode particles were found between neighbouring background sites ( $R^2=0.90$ ). These results indicate that the effects of particle emission and transport are complex in an urban atmosphere, and that a single point measurement of ultrafine aerosol concentration in a city may be representative within a limited spatial radius only. This experimental data set will be used to validate numerical emission and transport models in the future.

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## Chemical composition of aerosol particles including UFPs in Saxony

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Keywords: particulate mass, ions, OC/EC, organics, mass closure, source apportionment

Size-segregated particle sampling using impactors in different seasons and at differently polluted sites in Saxony in combination with trajectory analysis for the air mass origin estimation can be used to find out particle sources, fractions of different sources and long-range transport effects.

During the last decade a set of projects was accomplished by the IfT in Saxony including MINT 97 (Melpitz Intensive Measurement Campaign), Feinstaub 1999/2000 (a source study of PM in Saxony (I)), Schwebstaub 2003/04 (a study of size-segregated chemical composition in Saxony (II)), Feineintrag 2006/07 (a study of PM long-range transport in Saxony (III)) or the FAT project 2004/05 (examination of PM in a street canyon). Detailed descriptions of the measurement campaigns were published elsewhere (Plewka et al., 2004; Herrmann et al., 2006; Gerwig et al., 2006; Brüggemann et al., 2007). Several interesting results will be presented here:

- Significant differences in chemical composition of particles in different size ranges depending of air mass origin (east-west contrast)

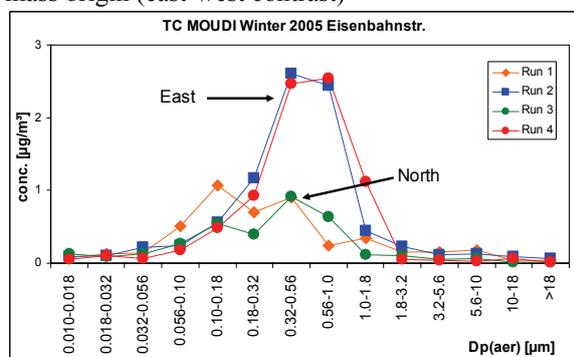


Fig.1: TC long range transport from East in Winter

- Seasonal differences of particle composition by size-segregated measurements at a roadside in Dresden (traffic and domestic heating influence)
  - Source apportionment by size-segregated particle component concentration measurements in summer and winter at a traffic, urban background, and regional background station
  - Correlation of particulate OC/EC in different size ranges with traffic density in a busy street canyon
- Some concluding remarks are given about the limited possibilities of local authorities to avoid exceedances of the PM<sub>10</sub> limit value of 50 µg m<sup>-3</sup> in the streets. Mass distribution shows a maximum

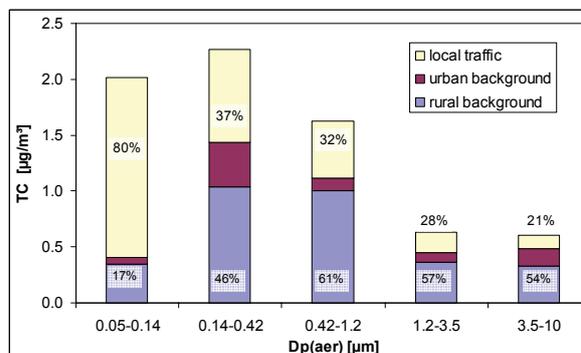


Fig.2: Contribution of local traffic to TC in a street canyon depending of particle size range

(about 50%) in the size range of  $D_{p, aer} = 0.42-1.2 \mu\text{m}$  originating mostly from long-range transport processes. Small particles from local emissions account for a high fraction of the number concentration, but only for a small fraction of the mass concentration.

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## PM abatement from a European perspective – current legislation and the CAFE thematic strategy

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Keywords: particle metrics, upcoming Air Quality Directive, European legislation.

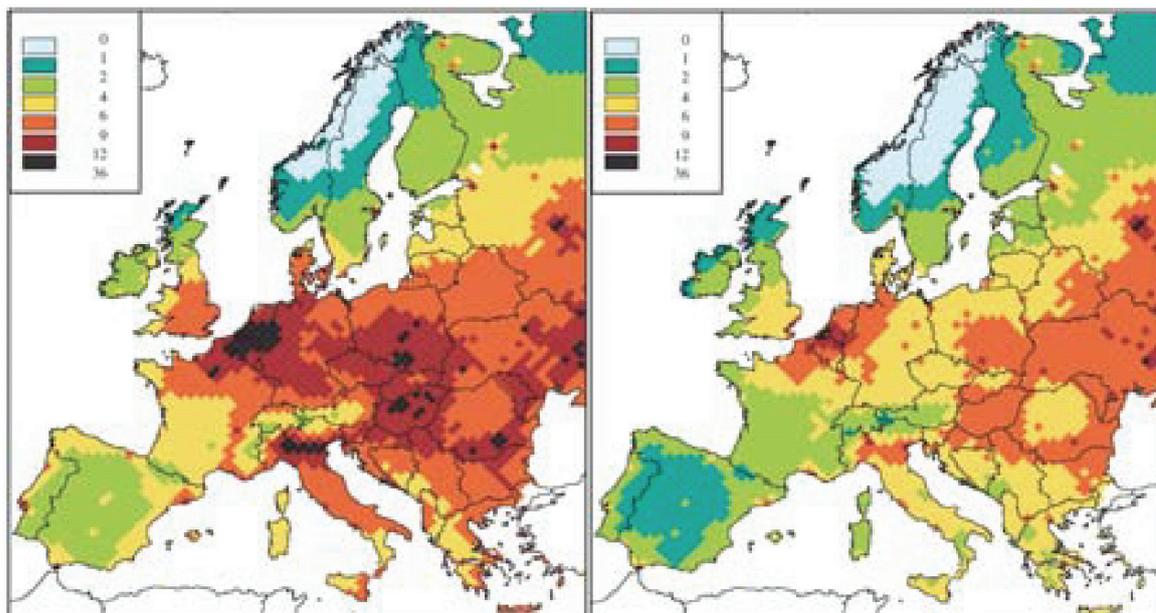


Figure 1. Estimated losses in life expectancy (in months) attributable to exposure to fine particulates 2000 and 2020; Source: IIASA, 2005

Attainment of the current PM<sub>10</sub> limit values laid down in the EC legislation poses big problems in the majority of the Member States. European PM<sub>2.5</sub> data are still incomplete. Until the end of 2006, about 140 plans and programmes to abate air pollutions have been set up.

With a view to attain an European air quality which has no significant negative impacts on health until 2020, the Commission has launched the thematic strategy “Clean Air For Europe” (CAFE) in 2005. As a political paper, this strategy lists important legislative projects such as the revision of the air quality legislation or the NEC directive. The CAFE strategy was underpinned by a comprehensive set of scientific reports, inter alia the 2nd PM Position Paper, an updated assessment of health effects by WHO, or an integrated assessment modelling of abatement options. PM turns out to be the pollutant with the most severe impact on public health, leading to a loss of life expectancy of more than 12 months in parts of Europe (compare Fig. 1). According to the scientific judgement, PM<sub>2.5</sub> is strongly associated with mortality and cardiopulmonary disease and was therefore chosen as principal PM metric for the upcoming regulation. It was felt too early to regulate UFP or particle numbers, but more research such as

determining and linking exposure to UFP with health effects was recommended.

The key elements of the Common position of Council and EC Commission for a new Air Quality directive as well as the state of proceedings are outlined in the paper. In respect to PM, the existing PM<sub>10</sub> limit values (annual and daily means) shall remain unchanged, but the attainment period may be extended by 3 years under certain conditions. The PM<sub>2.5</sub> burden shall be regulated by a target value of 25 µg/m<sup>3</sup> which applies everywhere, which will become legally binding in 2015. In addition, the average national exposure of the general public in big cities shall be reduced by 20 % going from 2010 to 2020. A revision of the directive is foreseen in 2013 with a view of making this reduction target mandatory. Negotiations between Council, Commission and European Parliament before the 2nd reading in Parliament, which will probably take place in December 2007, are still running. Main remaining points of discussion are the regulations for PM<sub>2.5</sub>, the applicability of the limit values and the prolongation of attainment dates.

# LIFE-ENVIRONMENT Project KAPA GS

## Klagenfurts Anti PM10 Action Programme with Graz and the South Tyrol

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Keywords: PM10 mitigation options, dispersion model, traffic bans, resuspension, domestic fuels, CMA

In many European cities the high pollution of particulate matter is an enormous environmental and health problem. Within the project KAPA GS several measures have been simulated and demonstrated from 2004 – 2007 in order to tackle the rising particle pollution during the winter period.

A dispersion model considering all known emissions has been developed by the Technical University of Graz and validated by monitoring stations for PM10 / PM2,5 / PM1.0 and NOx. Additionally a statistical forecast model enables to forecast the PM10 concentration (daily mean) for the next day in Klagenfurt, Graz and Bozen. A specific innovation has been done in Klagenfurt by linking the dispersion model with current wind and PM10-data: the result is an hourly updated nowcasting model.

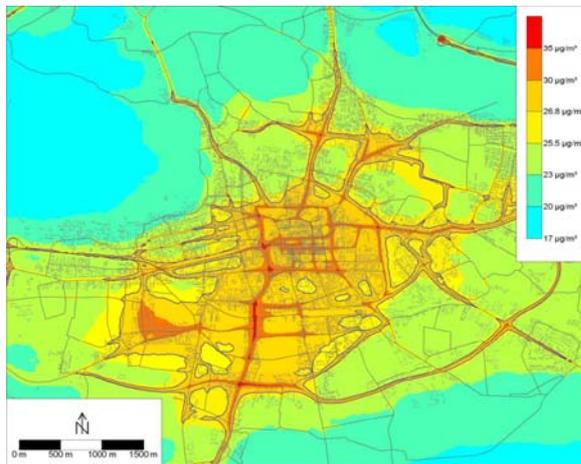


Figure 1. Dispersion model: annual Mean of PM10 in Klagenfurt, base case 2005

The dispersion model figures out the sources of pollution within a grid of 10 X 10 m.

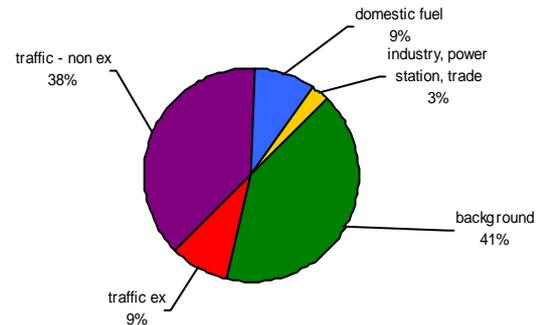


Figure 2. Sources apportionment of a hot spot in Klagenfurt, Völkermarkter Strasse, 23.000 ADT winter mean

The model is the main tool for the simulation of measures to reduce PM10 and calculate szenarios for mitigation strategies.

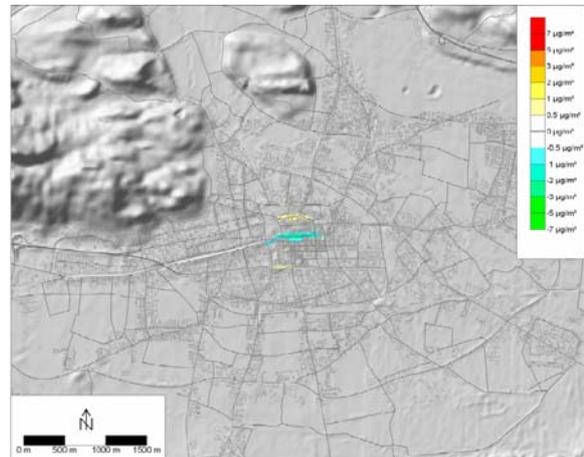


Figure 3. The traffic ban in the inner city of Klagenfurt (Neuer Platz/ Burggasse) reduces the PM10-level up to 3 µg/m<sup>3</sup> (annual mean).

Due to the remarkable contribution of re-suspension (38% of PM10 winter mean value) new road sweeping machines and CMA as dust-binder were tested.

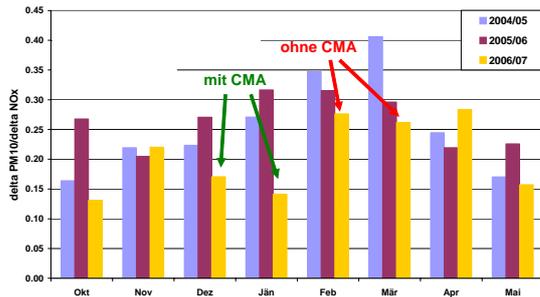


Figure 4. Delta PM10/ delta NOx ratio in winter 2004/2005, 2005/2006, 2006/2007(monthly mean, Völkermarkter Strasse). During periods with CMA-application a significant reduction of the PM10-level (up to -17% in January) has been monitored.

Moreover, to reduce traffic exhaust emissions the following solutions were tested and implemented: Park and Ride System with special anti-PM10-shuttle-bus, car free days, traffic bans and environmental zones, retrofitting of the city bus fleet in Graz und Klagenfurt by PM-catalytic converters and PM-filter systems.

PM-pollution from domestic fuel was tackled by intensive promotion campaign aiming at substitution of individual heating systems by natural gas or district heating connections.

8 electronic indicator boards (video walls) inform the population about the current PM 10-levels, park&ride possibilities and traffic bans, when paasing by on the main roads in Klagenfurt.

Many actions were initiated in the field of public awareness and dissemination to motivate people participating actively in setting activities against the PM10 pollution. Information campaigns in schools, folder, advertising campaigns, placements in newspapers, press conferences etc. have informed the public about the problem of the PM10 pollution and the EU-Project KAPA GS.

The two international project conferences (November 2005 in Graz, March 2007 in Klagenfurt) have been very important for the exchange of experiences to find a common solution.

The project KAPA GS demonstrates best practice for other cities in Europe.

More information and downloads:  
[www.kapags.at](http://www.kapags.at)  
[www.feinstaubfrei.at](http://www.feinstaubfrei.at)

Many measures tested and implemented within the project KAPA GS were very effective.

Klagenfurt has to save 50 days exceeding the limit value for PM10. Without the prognosticated traffic increase this target could be met in 2020 even on a hot spot in Klagenfurt by combining technical and planning measures.

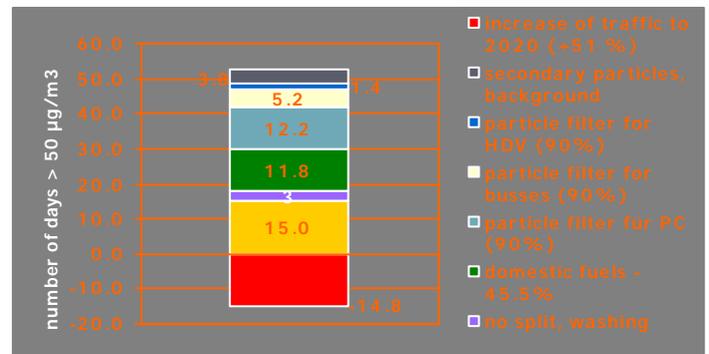


Figure 5. Reduction potential of measures in Klagenfurt, calculated in number of days exceeding the daily limit value of PM10.

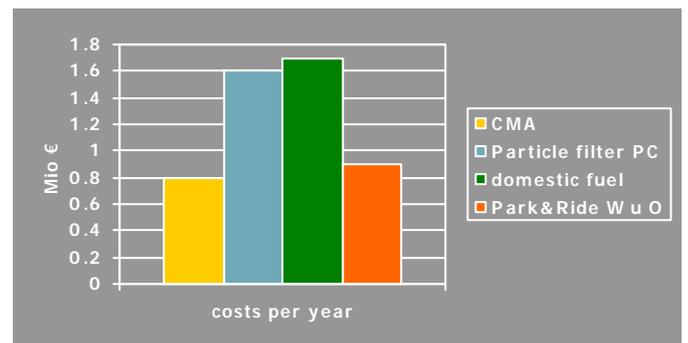


Figure 6. For some measures cost-benefit-analyses have been carried out.

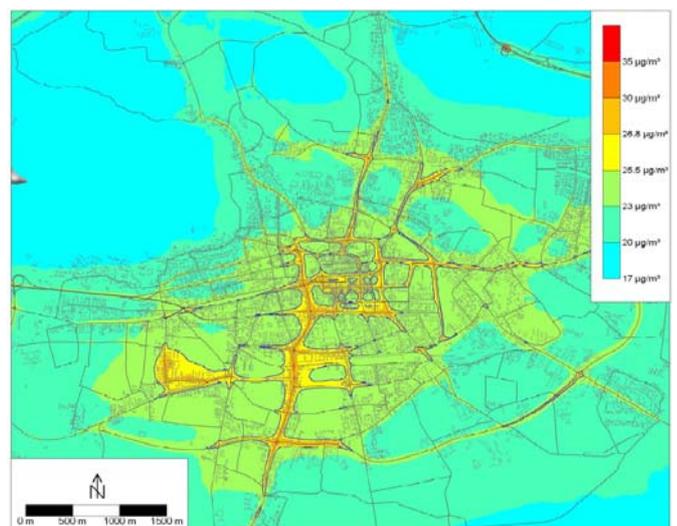


Figure 7. Best case Scenario for Klagenfurt 2020: Without traffic increase the PM10-levels comply with limit values in living areas.

# The PM and NO<sub>x</sub> air pollution in Copenhagen and assessment of possible measures to reduce the air pollution

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Keywords: Traffic, scenarios, environmental zones, ultra fine particles

Particulate matter (PM) and NO<sub>x</sub>/NO<sub>2</sub> are the most serious air pollution problem, in Copenhagen and in many other large cities in Europe. The main sources in Copenhagen to these pollutants are the local road traffic and the long range transported pollutants from Europe. Regulations of PM and NO<sub>x</sub>/NO<sub>2</sub> are closely related, because the road traffic is an important source to both.

NO<sub>x</sub>, NO<sub>2</sub> and PM<sub>10</sub>/TSP have been measured in the Danish Urban Air Pollution Monitoring program for many years. The monitoring strategy is based on simultaneous measurements at rural background, urban background and at "hot spots" (streets) in combination with air quality modelling (Kemp et al., 2007).

In addition, we made measurements of ultrafine particles in size fractions 10-700 nm by DMPS (Differential Mobility Particle Sizer).

Receptor modelling has been used for source apportionment of the different pollutants (Wählén, 2003), and source-receptor modelling for analysis of the measurement data and for scenario calculations. Both types of modelling are used for assessment of the air pollution in relation to the EU air quality directives now and in the future (Brandt and Palmgren, 2005).

NO<sub>x</sub> shows a clear decreasing trend in busy streets due to the stricter emissions standards for motor vehicles. NO<sub>2</sub> has been nearly constant in streets, because ozone is the limiting factor for formation of NO<sub>2</sub>, until recently where we observe a weak increase. This was probably due to more diesel cars, which emit NO<sub>2</sub> directly. This means that the limit value for annual average of NO<sub>2</sub> is exceeded now at busy streets and scenario calculations show that the limit value will be exceeded many years from now, if no other measures will be taken.

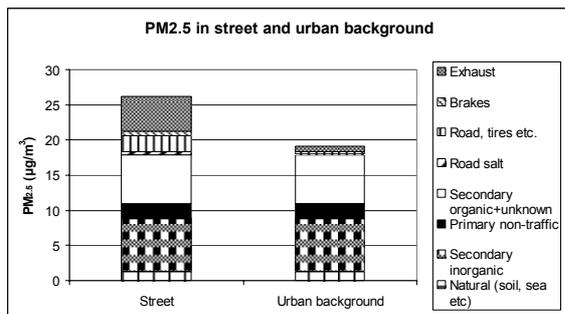


Figure 1. The different PM<sub>2.5</sub> contributions in streets and urban background.

PM<sub>10</sub> shows generally a decreasing trend, but the limit value of annual averaged will be exceeded at busy streets. However, the local contribution is rather low compared to the regional contribution (figure 1), which makes it difficult to reduce the PM pollution locally.

The ultrafine particles in a street can roughly be divided in 4 main parts, long range transport, soot, and 2 condensation parts from local traffic, figure 2 (Wählén, Berkowicz and Palmgren, 2006)

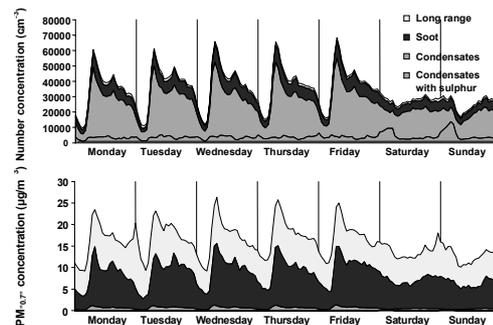


Figure 2. Ultrafine particles in a busy street in Copenhagen. Upper graph is the number concentration, and the lower graph is the volume concentration, both of the average week.

Investigations have been made of the effect of different types of environmental zones in central Copenhagen, focussing on retrofit of particles filters on heavy duty vehicles. Scenario calculations were performed for 3 different scenarios.

The results of one scenario, where all heavy duty vehicles should be equipped with particles filters, if they have EURO III engines or older. Results are shown in figure 3. The reduction of PM<sub>2.5</sub> in urban background will be rather small, i.e. below 0.4 µg/m<sup>3</sup>, which should be compared with the annual average, approx. 20 µg/m<sup>3</sup>.

Similar scenario calculations were performed for ultrafine particles. The reductions were up to 2,000 (N/cm<sup>3</sup>), which should be compared to the annual average 8,000 (N/cm<sup>3</sup>).

All scenario calculations were performed for urban background, because it was assumed to be most representative for the exposure of the population. The relative reductions at busy streets will be much larger.

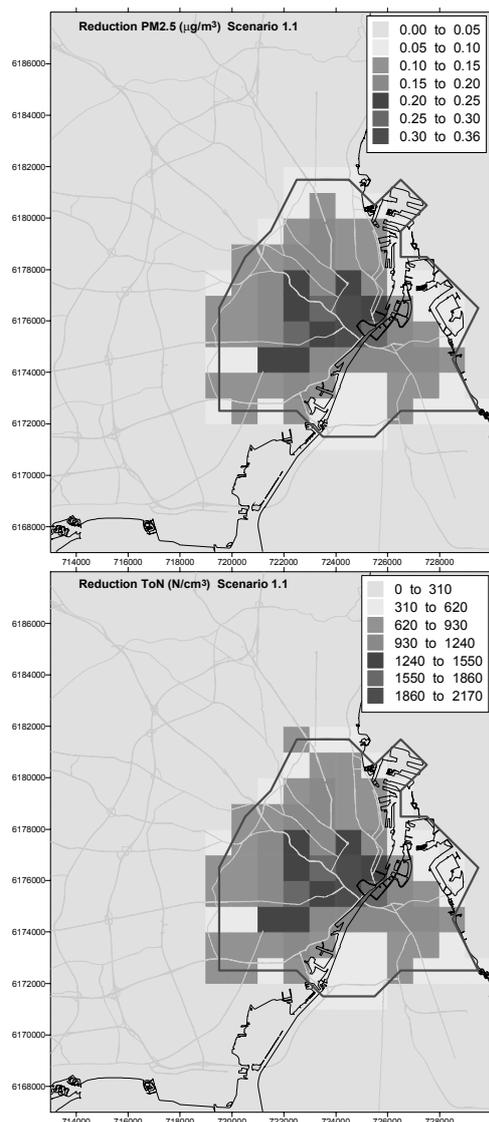


Figure 3. Expected reduction of PM<sub>2,5</sub> (upper graph) and ultrafine particles (lower graph) in urban background in Copenhagen, if all heavy duty EURO III vehicles or older were equipped with particles filters.

In addition to traffic wood combustion in wood stoves is an important source to PM in Denmark, especially in the residential areas around the bigger cities (Glasius et al. 2006).

Similar methods have been used to assess the effect in Denmark of the EU Thematic Strategy for air pollution.

This work was supported by the Danish Environmental Protection Agency.

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## Implementation of air quality directives in a candidate state



A Turkish - German Twinning Project under the project leadership of the German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety 

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Keywords: Twinning Project, Air Quality Directives, Transposition and Implementation..

The Twinning Project “Air Quality” started in October 2004 and ended in December 2006. The Turkish Ministry of Environment and Forestry (MoEF) and the German Federal Ministry for Environment, Nature Conservation and Nuclear Safety (BMU) were partners in this project. It had to fulfill four main tasks:

- Transposition of the Air Quality Framework Directive 96/62/EC and the Large Combustion Plants Directive 2001/80/EC into Turkish (Draft) Regulation
- Draft Agreed Framework Regulation on Air Quality which defines the Roles and the Responsibilities of the involved ministries (considering both directives)
- Strengthening of the qualification of the administration (Know-How-Transfer) – Strengthening of the Quality management and preparation of the accreditation of the two laboratories – Refik Saydam Hygienic Center (RSHC) and Gölbaşı
- Agreed strategic Action Plans on further implementation steps of the two directives

During the first year legal gap analysis, a structure analysis concerning the administration and assessments were carried out. Two drafts have been developed, one for a Turkish Air Quality Framework by-law and one for a Large Combustion Plant by-law. Both drafts consider for the transition time the old regulation. These by-laws are supposed to fulfill the requirements of the European Air Quality Framework and Large Combustion Directives. In addition, trainings were arranged for strengthening the Quality Management in the two above mentioned laboratories. Additional trainings and working groups informed about topics like zones and agglomerations, preliminary assessment and assessment of air quality, emission inventories, air pollution modeling, data validation, reporting, permission and inspection procedures etc. One result of these activities was a proposal concerning zones and agglomerations in Turkey. Another important result was the

implementation of the EURAD model (Rhenish Institute in Cologne, University of Cologne, Germany) at the Turkish Meteorological Service which is part of the MoEF.

In connection with the Twinning project the equipment for an ambient air pollution network for Ankara was ordered and was delivered shortly before the end of the project. Trainings about maintenance, repairs, quality assurance and accreditation procedures were given.

The second year of the Twinning project was generally concerned with the preparation for the implementation of the above mentioned Directives. Cities like Istanbul, Izmir, Bursa, Erzurum were visited in a campaign to check the automatic instruments which were already in place in different local networks. The station in Erzurum is part of the national network. The MoEF built up this network during the time of the project from its own resources. Therefore some of the experts who were employed in the project were also involved in these activities. Each one of the 81 provinces received one station which is equipped with one SO<sub>2</sub> monitor and one PM10 monitor. The MoEF plans to equip these stations with additional components.

Calibration and maintenance tasks are normally carried out by companies or in the case of Izmir by the local university. In general, gas cylinders with certificates are used for calibration.

In addition to practical trainings two strategy papers were prepared for the future implementation of the prepared by-laws. These strategy papers recommended new administration structures for the implementation of the directives and gave estimates for the costs concerning the implementation of the directives.

The following proposal concerning future ambient air pollution network stations and regional network centres for Turkey was made: see Figure 1.

## Distribution of station types in the regional networks

(inclusive additional ozone monitoring stations in agglomerations)

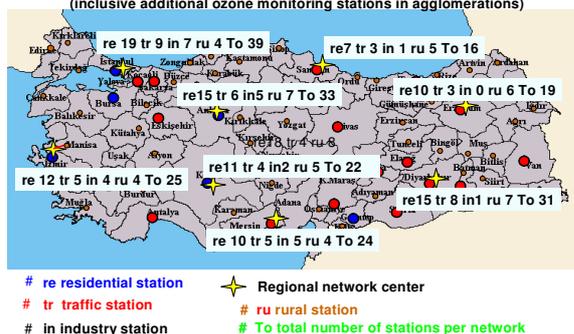


Figure 1: Proposal for future ambient air pollution networks and regional network centres

Main future tasks are:

- The two by – law drafts need to become effective in order to provide legal certainty for future investors
- Further trainings on running an air pollution network are needed
- Build up of a nationwide quality management system to ensure the equal standard of the measured emission and air quality data in all parts of Turkey (introducing a primary standard, round robin tests, cooperation with international institutions etc.)
- Build up of emission data bases (national and regional); ensuring the quality of the data
- Further implementation of models (national and regional); validation of the model results
- Build up of action and clean air plans for regional areas and agglomerations where the limit values have been exceeded

This work was supported under Project Title “Air Quality, Chemicals, Waste; Component 1: Air Quality”, TR 03-IB-EN-01 by the European Commission



The project leadership was carried out by the German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety

Reports and strategy papers: unpublished information

# Street-Detailed Calculation Of The Air Quality In Saxony

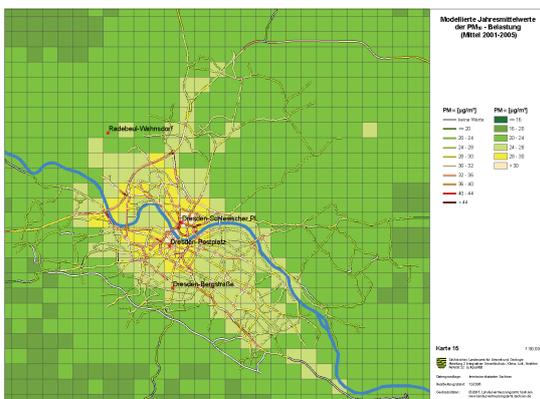
Uwe Wolf

Sächsisches Landesamt für Umwelt und Geologie

Keywords: air quality, immission register, spread calculation

The Saxony immission register is an outstanding tool to calculate and display the air quality from NO<sub>2</sub> and PM<sub>10</sub> in Saxony

Through the combination from different dispersion models is an area-wide calculation of the immission situation in rural areas as well as street-detailed calculation in urban areas possible.



Picture 1, modelled yearly average values of PM<sub>10</sub>-pollution (average 2001-2005)

Following methods were used:

- an interpolation with inverse distance weighting to calculate the background pollution
- the Lagrange spread model LASAT to calculate the additional pollution from the area
- the Gauss model PROKAS to calculate the net entry through transport in urban areas
- the diagnostic spread model PROKAS B to calculate induced transport additional pollution in the streets

For Practical applications (e.g. clean air plans for Saxony communes) the variation between measured and calculated values are mostly under 20%.

Table 1, comparison / modelling

	Measured		Calculated	
	PM <sub>10</sub>	NO <sub>2</sub>	PM <sub>10</sub>	NO <sub>2</sub>
Dresden - Bergstraße	33	58	31 (31)	47 (49)
Dresden - Mitte	31	31	30	31
Dresden - Nord	33	47	35	49

With this procedure it is possible to calculate not only current but also future situations. With these results it is possible to review the success of the actions taken. Therefore it is used in five cities of Saxony to create clean air plans.



UFIPOLNET LIFE04 ENV/DE/000054





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# POSTER

## Particle number concentration in the urban area of Rome

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Keywords: Ultrafine particles, Polycyclic Aromatic Hydrocarbons, Nitrogen oxides

Atmospheric particulate matter (PM) pollution is presently regulated on mass basis through PM<sub>10</sub> convention that measures the mass of particles collected with a 50% efficiency for particles with an aerodynamic diameter of 10  $\mu\text{m}$ , in that way including, coarse particles down to ultrafine particles (UFPs),  $<0.1 \mu\text{m}$ . UFPs deposit very efficiently in the lungs. Their small size, high number concentration and surface area allow them increased ability of absorption of organic molecules and penetration into cellular targets in the lung and systemic circulation (Li et al., 2003). Such characteristics account for their importance in explaining the health effects of PM.

This work describes the first results of a study started in April 2007 to investigate the UFP pollution in the urban area of Rome. Particle number concentration and size distribution have been measured by means of TSI 3936 Scanning Mobility Particle Sizer, configured with 3080 Electrostatic Classifier, 3081 Differential Mobility Analyzer and 3786 water-based ultrafine condensation particle counter. Number concentration data have been compared with nitrogen oxides (NO<sub>x</sub>), measured by chemiluminescent analysis and particle-phase Polycyclic Aromatic Hydrocarbons (PAHs), measured by Ecochem PAS 2000 PAH Monitor. Positively charged UFPs are being sampled using the TSI 3089 electrostatic nanoparticle precipitator, after passing the aerosol through a TSI 3080 Electrostatic Classifier.

Transmission Electron Microscopy (TEM) is a technique widely used to study the size, morphology, composition, microstructure and crystallinity of individual particles (Chen et al., 2005). Such technique is being used in the ongoing UFP characterization study, by means of an Energy-Filtered Transmission Electron Microscopy (EFTEM) FEI TECNAI 12.

Figures 1 and 2 show the daily trend of number particle concentration in the range 20-880 nm compared respectively with NO<sub>x</sub> and particle-phase PAH. The pattern of variation of such pollutants are very similar, suggesting a common automotive origin. Number particle concentrations show a typical daily modulation with minimum values measured during nocturnal hours when the effect due to the reduction of the automotive traffic emission overcomes the decrease of the atmospheric mixing height.

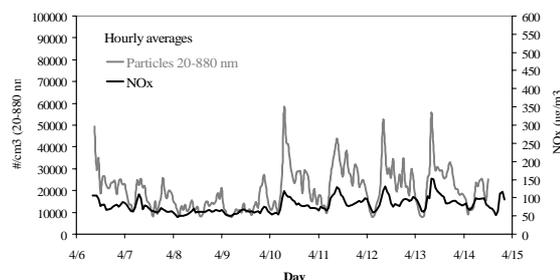


Figure 1. Daily trends of particle number concentration (20-880 nm) and NO<sub>x</sub>.

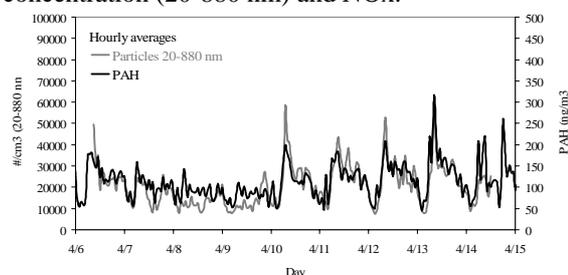


Figure 2. Daily trends of particle number concentration (20-880 nm) and particle-phase PAH.

The first results of TEM study indicates the presence of single (figure 3), and aggregates carbonaceous UFPs.

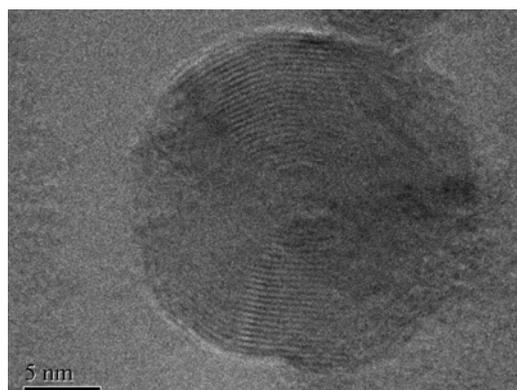


Figure 3. EFTEM micrograph of a carbonaceous UFP.

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## Using a sampling and monitoring device, a solution for PM10-PM2,5 assessment ? Experience with the Swam 5-A

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Keywords: PM10, PM2,5, Equivalence

Present European PM regulation (directive) is from 1999, measurement standards from 1998 (PM10) and 2005 (PM2,5).

However, reconciliation, through “Equivalence trials” automatic analysers actually used by member states the gravimetric reference method is still an on-going process in 2007.

Presently the Walloon network for PM relies on the beta attenuation analyser MP101M.C. It should be run with a 24h cycle to satisfy to Equivalence criteria. But that time resolution is not acceptable for the Competent Authority. Accordingly, it is foreseen to supplement and eventually replace that analyser by others, which means more Equivalence trials ahead.

The correction factor (PM10) currently used for the MP101M.C is 1,37.

Besides, from our experience, reference PM samplers available up to now do not completely meet requirements of the gravimetric standard. This they bring an initial uncertainty, using part of the budget, when candidate analysers are processed through gravimetric equivalence trials.

ISSeP ran and tested two dual channel sampler and beta attenuation analysers of FAI-Instruments (Fonte-Nuova, Italy) between October 2006 and August 2007. Although more sophisticated uses are put forward, ISSeP ran exclusively the devices as a PM10 and PM2,5 sampler and analyser, operated at the reference flow of 2,3 m<sup>3</sup>/h. Cycle was always 24H (hence two filters sampled - one PM10 and one PM2,5) and two beta attenuation results (one PM10 and one PM2,5) for each device and each day.

In April 2007, initial devices were exchanged against a more transportable version. One of these exchange Swams was with an embarked optical analyser delivering results for two sizes or classes.

Remedies or improvements of the manufacturer in reaction to ISSeP's feedback were an upgrade of the filter cartridge (to avoid filter adhesions observed with some brands of quartz filters) and a subsequent mechanical adjustment to maintain perfect shape of dust spots (as mandatory for the beta attenuation analysis).

Experienced achievements of the Swam as an analyser include actual permanent measurement of

flow, temperature conditioning for filters to ensure respect of the maximum allowable 5°C (ambient vs. filters) temperature difference and recording of that difference.

Table 1. Precision of Swam 5β analyser

Filter material	PM10	PM2.5
	u.bs (µg/m <sup>3</sup> )	u.bs (µg/m <sup>3</sup> )
fiberglas whatman	0.70	0,67
quartz macherey-nagel	0.99	0.99
quartz what. winter	0.70	0.67
quartz what. summer	0.76	0.72

Table 2. Equivalence : β analyser result vs. gravimetric result (PM10, quartz Macherey-Nagel)

equation	Y=1.03x-0.66
u%	7,4
factor from ratio the means	1.00
factor from orthogonal regression through origin o.R.o.	1.01

Table 3. MP101.C-pm10-(2h cycle) versus Swam β result considered as reference

swam filter material	u%	derived corr. factor	
		from ratio	From o.R.o.
fiberglass whatman		1.15	
quartz macherey	7,5	1,20	1,21
quartz what. winter		1,17	
quartz what summer	25	1,25	1,26

This confirms MP101M.C is not appropriate for compliance monitoring when run with a cycle shorter than 24H. However, as long as it is still used, the appropriate correction is presumably in the range 1,15-1,25 rather than the currently used 1,37.

Support for this work of DGRNE (Cellule Air), Walloon Ministerial competent authority, is gratefully acknowledged.

EN 12341 (1998) Air quality, determination of the PM10 fraction of suspended particulate matter

EN 14907 (2005) Standard gravimetric measurement method for the determination of the PM2,5 mass fraction of suspended particulate matter.

Demonstration of equivalence of ambient air monitoring methods (report of EC working group) (<http://ec.europa.eu/environment/air/cafe/index.htm>)

# Measurement and analysis in space and time of ultrafine particle number concentration in ambient air. The case of Parma

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Keywords: ultrafine particles, number concentration, urban ambient air.

Several studies have analysed the influence of ultrafine particles (UFP) on human health in atmosphere (e.g., Pope et al., 1995). Even though preliminarily, such works suggest that motor vehicles are a significant UFP source (Kittelson et al., 2006). Additionally, this source could be even more important than for PM<sub>10</sub> (Harrison et al., 1999).

In this work, the major objective was to collect a data-set of UFP number concentration in a middle-size urban area in Italy (Parma) with the aim to evaluate connections with the local traffic sources.

Measurements were taken every 2 seconds at two urban sites, 700 meters far each other: a traffic site (less than 1 meter from the road) and a urban background site (around 100 meters from the closest road). UFPs were measured during winter and summer time of 2007 (both week-days and week-ends) by using two tandem Water CPCs (TSI, mod 3781).

UFPs concentrations were relatively low (fig.1-4) when compared to similar works (e.g., Harrison et al., 1999).

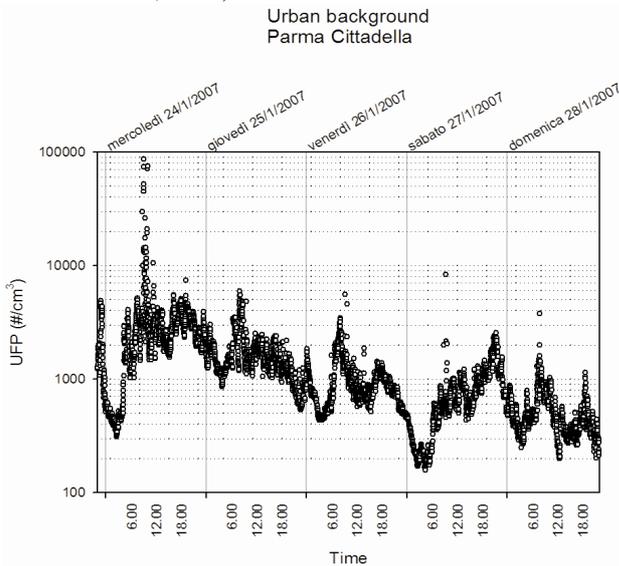


Figure 1. UFP number concentration during the winter campaign at an urban background station in Parma.

As found in previous studies (e.g., Zhang et al., 2004), UFP concentrations measured at the two sites showed significantly different tendencies. Differences related mainly to two factors (fig.1-4): at the traffic site (i) higher concentrations and (ii) faster

and more intensive dilution processes were measured.

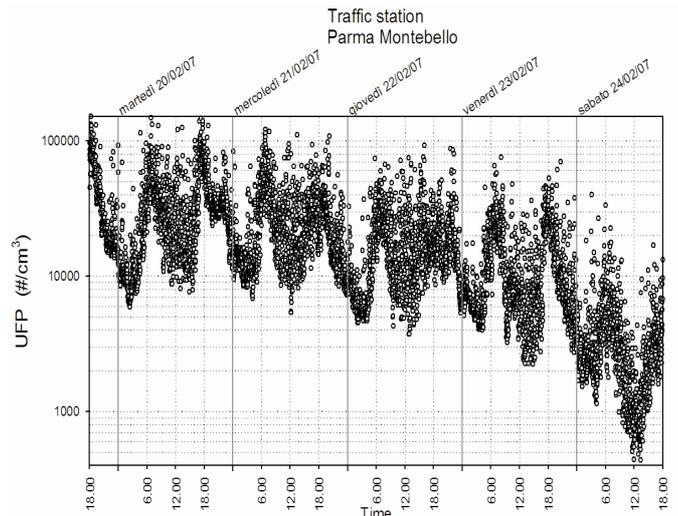


Figure 2. UFP number concentration during the winter campaign at a traffic station in Parma.

Beyond a more rapid reduction of UFP number, the faster dilution processes at the traffic site could also trigger other physical processes (such as nucleation) in connection to other parameters (such as solar radiation).

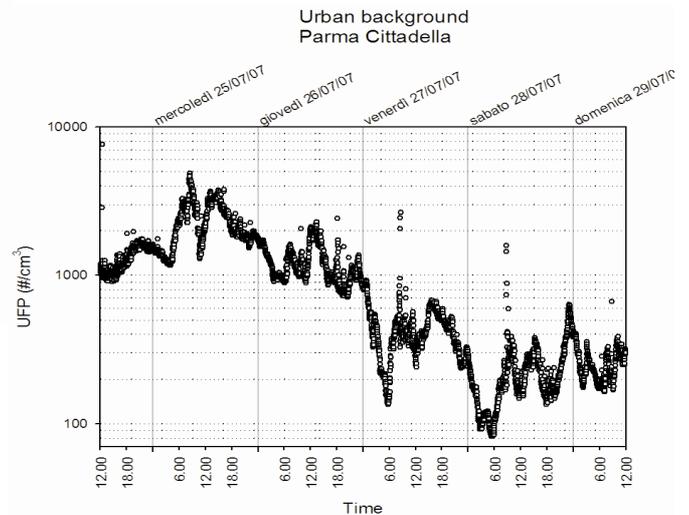


Figure 3. UFP number concentration during the summer campaign at an urban background station in Parma.

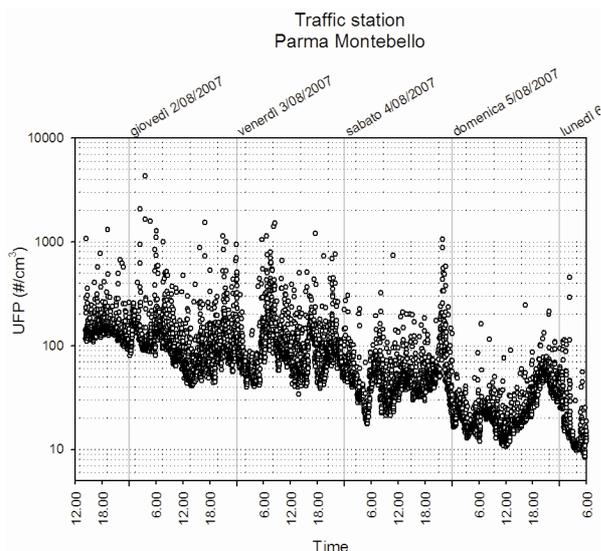


Figure 4. UFP number concentration during the summer campaign at a traffic station in Parma.

UFPs measured at the traffic site clearly shows (fig.2 and 4) as expected (e.g., Shi et al., 1999) the two peaks related to the traffic rush hours indicating traffic emissions to be the prevailing source.

The influence of the traffic flows can also be recognised at the urban background site likely in connection to transport phenomena (e.g., due to wind speed). At this site, the influence of meteorology can easier be analysed. This is particularly true for a clear correlation with the total solar radiation ( Harrison et al., 1999). Its influence can be seen at both the sites: in effect, beyond the rush hours peaks, the traffic site shows clearly a third peak at midday (fig.1-4).

The analysis of the all measurements and the comparison between winter and summer time, week-days and week-end showed, in summary, three major contributions. Firstly, a very low urban background UFP concentration (lower than 1000 #/cm<sup>3</sup>) particularly visible in summer. Secondly, a significant contribution due to local traffic sources, up to 100000 #/cm<sup>3</sup> (traffic site in winter time). Finally, a significant contribution due to secondary processes closely linked to meteorology, particularly solar radiation, and evident at midday.

This work was supported by the Italian Ministry of Environment, Land and Sea in the framework of the project “Air Pollution Emission Monitoring in the city of Shanghai”.

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## Experiences with ultra fine particle monitoring in air quality monitoring networks in Europe

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Keywords: Combustion particles, Health effects of aerosols, Outdoor aerosols, PAH(s), SMPS

In air monitoring networks particles in the range of a few hundred nm are not measured. For mass related measurements the ultra fine particles are negligible. Nevertheless these small particles (below 500nm) contribute with 80% to the particle number concentration in ambient aerosols. Special in urban regions the total particle concentration is determined by very small particles like diesel soot, generated by combustion processes. In epidemiological discussions on air born particles the focus of interest has shifted from mass to number concentration in the recent years. Therefore it a very interesting task to measure the ultra fine particles additionally to the coarse fraction and compare the data with the meteorological parameters and the values of important volatiles.

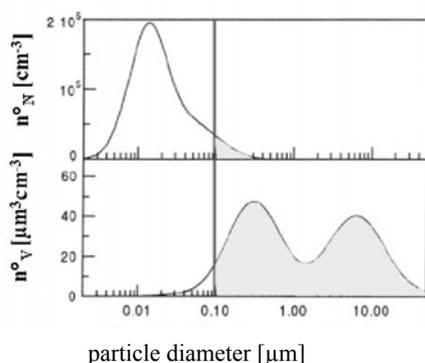


Figure 1. A typical volume and number distribution in an urban aerosol (Seinfeld and Pandis, 1997)

To compare the measurements of ultra fine particles with the data, obtained from the well proofed instrumentation of the air quality monitoring networks, it is essential to be sure that the devices for measuring nano particles are working stable and comparable. Such investigations were carried out in the last years for the GRIMM SMPS+C instruments with good results (for example in December 2006 in Leipzig at the Leibniz Institute for Tropospheric Research).

There are also strong efforts at the moment in Germany to work out the details of measuring ultra fine particles for a VDI DIN directive. A first

draft defines already the use of a CPC (Condensation Particle Counter) and a DMA (Differential Mobility Analyser).

In the following, some examples of measuring campaigns are mentioned in which GRIMM instruments were used in air quality monitoring networks to obtain additional information about the aerosol particles. Where “SMPS+C” means a combination of a CPC with a DMA for the size range between 5 and 1100 nm and “WRAS” (Wide Range Aerosol Spectrometr) the combination of a SMPS+C with an OPC (Optical Particle Counter) for particle diameters up to 30 µm.

In April 2006 such measurements with a WRAS were done in Graz (Austria). The measured particle concentrations related strongly to the traffic and folkloric events (Easter fires).

In May 2006 during eight days a measurement in Salzburg (Austria) was carried out. Here additionally to the WRAS a PAH sensor was installed. The concentrations of the ultra fine particles and the values of the PAH sensor correlated strongly with the values for NOx and CO<sub>2</sub>, which were measured by the network.

In winter 2007 in Graubünden (Switzerland) a long-term measurement (over six weeks) was done. Here the particle concentration, measured with a SMPS+C, in rural and urban regions were compared.

Special thanks to the involved networks.

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## UFIPOLNET: Concentration of Particle Number Distributions at 4 Stations in Europe

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Keywords: atmospheric aerosols, instrument development, number concentration, number size distribution

Several studies show a decline of particle mass concentrations in Central Europe of TSP and PM10 1990 - 99. In contrast, particle number concentrations of ultrafine particles (< 100 nm = UFP) were not changed during winter periods 1991 – 1999 in Erfurt/Germany (Cyrus et al. 2002). There are however only a limited number of long-term UFP measurements in Europe. Epidemiological studies showed a relationship between high number concentrations of UFP and adverse health effects.

The European Commission needs therefore more information about UFP concentrations for evaluation processes within the CAFE process and the Thematic Strategy on Air Pollution.

The project UFIPOLNET (Ultrafine Particle Size Distributions in Air Pollution Monitoring Networks) intends to demonstrate that the newly developed Ultrafine Particle Monitor UFP 330 is able to perform adequately in routine network operation.

The instrument produces a number size distribution (20 – 800 nm). Only 6 size classes >20, >30, >50, >70, >100, >200 (N1 – N6) are transferred to the central measurement network stations to reduce the amount of data collected in the databases.

First comparisons with a DMPS for ambient aerosols (Wehner et al. this issue) show a good correlation with a DMPS measuring in parallel at a street canyon site.

Since December 2006 in Dresden and February 2007 in Augsburg, Stockholm and Prague, the UFP 330 will run continuously until October. It is planned to run the instruments on a permanent basis for a longer period. All sites are near busy roads; Augsburg is an urban background site. The number concentrations will be correlated with nitrogen oxides, benzene and other continuously measured parameters in a routine measuring network. In some places, traffic numbers will be correlated with the measurements.

At three stations, SMPS/DMPS size spectrometers have been monitoring for several years. Figure 1 compares the annual mean concentrations of total number concentrations per

station (2003 – 2005). Augsburg shows about half, Stockholm twice as many particles as Dresden. Prague and Dresden show almost the same concentration of NOx in 2005, while the street canyon of Stockholm shows almost twice the concentration. The correlation with NOx indicates the traffic influence (Birmili, 2006).

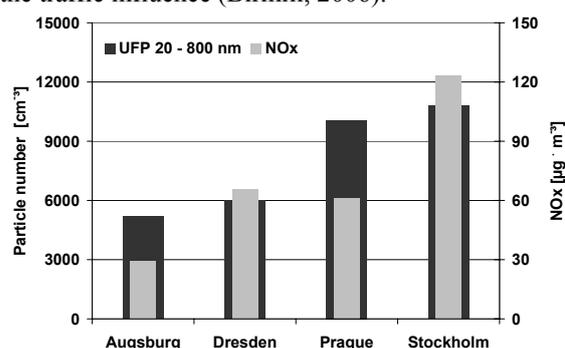


Figure 1: Number concentrations at the 3 stations with reference instruments

One aim of UFIPOLNET is to harmonise the sampling conditions (particle pre-impaction and humidity) as well as the evaluation of identical size classes. In this way, interpretations of particle number concentrations and size distributions will be facilitated. Comparable results will permit analysis of absolute differences between ultrafine aerosol size distributions at many polluted sites over long periods.

UFIPOLNET ([www.ufipolnet.eu](http://www.ufipolnet.eu)) is financed by the LIFE financial instrument of the European Community under No. LIFE04 ENV/D/000054. The authors wish to thank Heinz Ott for providing NOx-data in Augsburg from the routine air measuring network of the Bavarian State Agency of Environment (LfU).

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## Determination of particle emission factors of individual vehicles under real-life conditions

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Keywords: ultrafine particles, traffic, urban pollution, vehicle emissions.

Road traffic constitutes an important source of particulate matter and trace gases. Most particles in vehicle exhaust are in the ultrafine size range. In contrast to large particles which are a result of wear of road pavement, tyres and brakes, ultrafine particles are respirable and penetrate deep into the lungs, posing a threat to health. Especially in the densely populated urban areas, road traffic can lead to severe pollution of the ambient air. Since today half of the global population lives in urban areas, it is a matter of public and scientific concern to examine the emissions under real-life conditions. Particularly the measurement of the particle emissions from individual vehicles that form a car fleet delivers insight into the variability of particle emissions among cars and might allow the assignment of emission properties according to vehicle type, age, fuel technology etc.

Here we present an experimental setup for the measurement of particle emission factors (EF) from individual vehicles, which was designed for continuous on-road sampling. The system has been applied in two test experiments conducted in the Göteborg area. The measurement sites were at a rather busy major street in Göteborg (Western Sweden) and a sparsely frequented two-lane country road 25 km from Göteborg, respectively. The measurements were each performed monitoring the traffic on one of the two lanes. The inlet of a sampling line was installed in the centre of the lane and attached to the street surface to extract air directly (*in-situ*) from the plumes of passing vehicles. To measure CO<sub>2</sub> and number density of particles > 10 nm simultaneously, the collected air was distributed to a CO<sub>2</sub> monitor and a condensation particle counter (CPC, TSI 3010), respectively. The additional registration of licence plate numbers from the vehicles driving past gives information about the vehicle type, engine power and further technical data provided by the National Road Administration.

While during the first experiment everyday traffic was observed to prove the applicability of the described approach, the second experiment served to study the intra-vehicle variability of particle emissions under predetermined driving conditions (specified speed and gear). A set of four selected vehicles, consisting of a diesel car (car I), an older medium-sized petrol car (car II) and two compact cars (car III, car IV) was applied.

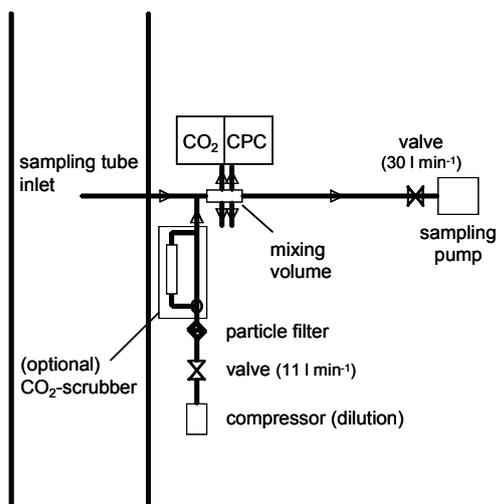


Figure 1. Measurement setup for simultaneous sampling of CO<sub>2</sub> and particle concentration.

Although the sampled air volume was diluted with a known amount of particle-free background air, the very high particle numbers within the vehicle exhaust plumes gave concentrations exceeding the upper limit of the particle counter. A solution to nevertheless quantify the excess particle numbers in the plume is presented and the particle EF's are derived from the enhancement ratios of particle number to CO<sub>2</sub> mixing ratio in the exhaust plumes of passing vehicles. When assuming a CO<sub>2</sub> emission of 164 g km<sup>-1</sup> veh<sup>-1</sup>, which is an average value for petrol-fuelled vehicles (cp. [www.starterre.fr/voiture-auto/emission\\_co2](http://www.starterre.fr/voiture-auto/emission_co2)), particle emission factors of the order  $2 \times 10^{13}$  part km<sup>-1</sup> veh<sup>-1</sup> were derived from the observed car passages (cp. Figure 2). This value agrees well with those published in the literature for petrol-driven vehicles (e.g. Jones & Harrison, 2006; see also lower side bar in Figure 2), which constitute more than 90% of the Swedish fleet (Ahlvik, 2002). Emission factors for diesel driven passenger cars are one order of magnitude higher (cp. upper side bar in Figure 2). Given the obtained results, the used setup proved appropriate to quantify particle emission factors from road traffic.

The measurements and observations of this study demonstrated that the simultaneous measurement of CO<sub>2</sub> and particle number for the purpose of characterising individual vehicles'

emissions is a feasible approach. Furthermore, by using the described method the particle emission can be linked to other indicators for traffic exhaust such as  $\text{NO}_x$ , VOC or CO in order to better understand real-world vehicle to vehicle variation in particle production.

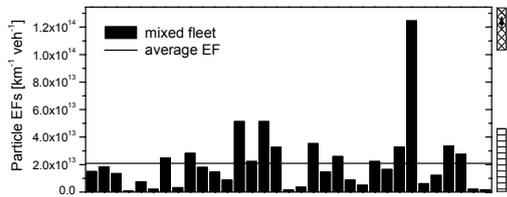


Figure 2. Single vehicle number particle emission factors derived from measurements in Göteborg.

This work was supported by the Swedish Foundation for Strategic Environmental Research MISTRA and the National Swedish Road Administration. Benny Lönn, Senior Research Engineer, is acknowledged for skilful technical support.

Ahlvik, P. (2002). *Environmental and Health Impact from Modern Cars*, Report for the Swedish National Road Administration, Publication 2002:62.

Jones, A. M., & Harrison, R. M. (2006). *Atmos. Environ.*, 40, 7125-7137.

## Particle number size distributions of ambient-state and non-volatile aerosols in the city of Augsburg, Germany

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Keywords: aerosol size distribution, urban aerosols, soot particles

Fine and ultrafine (< 100 nm) aerosol particles in the environment have moved into the interest of public health research due to their presumed adverse effects upon human health, such as cardiovascular and respiratory disease. While the adverse effects of ambient particles are in general widely acknowledged, there has been only little epidemiological evidence on the role of particular sub-fractions of the aerosol.

The city of Augsburg in Southern Germany hosts a centre of environmental medical research for the quantification of air pollutants and their effects on sensitive parts of the population (KORA; Holle et al. 2005). Specialized aerosol particle measurements were started in November 2004 in order to support future epidemiological studies within KORA.

As ambient particles are a complex mixture of a myriad of chemical compounds, there is a growing need to characterise and isolate those particular sub-fractions that are relevant to human health. Volatility analysis (with thermodenuder) is a method that makes use of different volatilisation temperatures of chemical compounds, thereby separating volatile compounds, such as organic matter, sulfates and nitrates, from non-volatile compounds, such as soot and mineral dust. A volatilisation temperature of 300°C allows to identify soot particles in the ultrafine size range emitted from vehicular traffic (Wehner et al., 2004; Rose et al. 2006).

At the GSF research station Augsburg ambient state and non-volatile particle size distributions (3 - 800 nm) have been measured continuously since 11/2004 using a twin differential mobility particle sizer (TDMPS).

Figure 1 presents median particle number size distribution for ambient and non-volatile compounds during the rush hour traffic; while the ambient particle number distribution peaks in the Aitken mode (~40 nm), the curve of non-volatile residues peaks in the nucleation mode (~10 nm) and in less-volatile particle mode (~80 nm) which presents the externally mixed population of soot particles. These measurements imply that within the measurement accuracy, every ambient particle contains a non-volatile core. The chemical composition of the non-volatile residues < 20 nm is, however, not known yet.

We will present a statistical summary of the 2-year data set, including an analysis of the relationship between total and non-volatile particle size distributions, and the meteorological factors that cause high concentrations of total and non-volatile particle fractions, such as wind direction, mixed layer height, and remote transport.

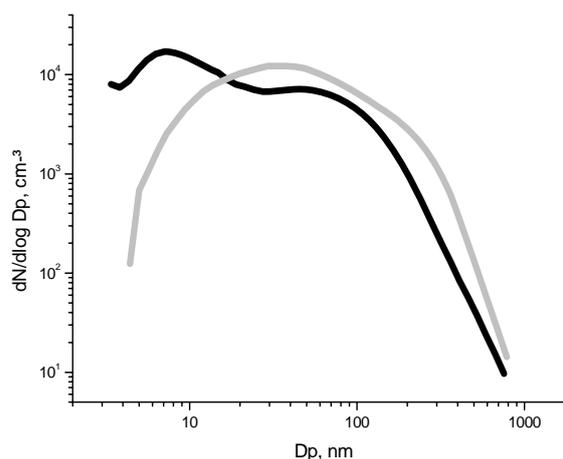


Figure 1: Median particle number size distribution of total (grey) and non-volatile (black) aerosol between 7:00 – 9:00 am.

- Holle, R., Happich, M., Löwel, H., Wichmann, H. E. (2005): KORA – A Research platform for population based health research, *Gesundheitswesen* 2005, 67 Sonderheft 1, S19-S25.
- Wehner, B., Philippin, S., Wiedensohler, A., Scheer, V., Vogt, R. (2004): Variability of non-volatile fractions of atmospheric aerosol particles with traffic influence, *Atmos. Env.*, 38, 6081-6090.
- Rose, D., Wehner, B., Ketzler, M., Engler, C., Voigtländer, J., Tuch, T., Wiedensohler, A. (2006): Atmospheric number size distributions of soot particles and estimation of emission factors, *Atmos. Chem. Phys.*, 6, 1021-1031.

# Determination of the charge distribution of highly charged aerosols

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Keywords: electrical analyzers, electrical charging, charge distribution

## INTRODUCTION

Bipolar chargers are widely used in aerosol science because their charge distribution is well understood (Wiedensohler, 1988). However, for the determination of aerosol size distributions unipolar charging may have some advantages. Using a unipolar charger in this application requires that the charge distribution is well known. Biskos et. al. (2005) examined charge distributions for soot particles with a Hewitt-type charger.

The work presented here explores the possibility of using the unipolar charger of the Electrical Aerosol Detector (Medved, 2000) for the quantification of ambient aerosols. This requires the measurement of the charge distribution applied by the charger on several particle systems varying in particle material and shape.

## EXPERIMENTAL SETUP

The charge distribution for several particle systems (latex, silica, soot) was experimentally determined in a Tandem DMA setup (Figure 1). It consists of three parts. In the left part a monodisperse fraction of the test aerosol is generated. This aerosol is subsequently diluted and positively charged in the middle part. Passing the DMA 2 the particles are classified according to their electrical mobility which depends on the number of charges carried by the particles. The combination of an electrometer and CPC 1 in the right part of the setup returns the mean charge of the particles. This allows the verification of the charge distribution.

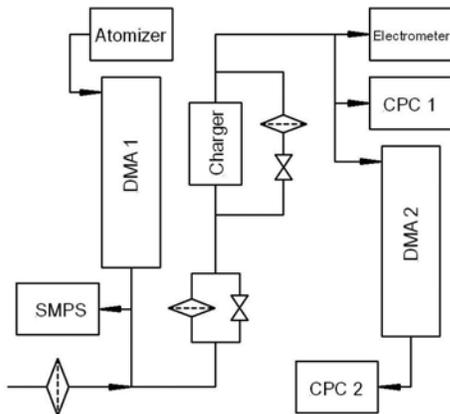


Figure 1: Experimental setup for measuring the charge distribution

## DATA CONVERSION

The high number of charges impedes the proper separation of the charge modes by the DMA. Therefore it is necessary to model the analyzing DMA using its transfer function. Fitting the output of the second CPC to the measured data determines the charge distribution of the aerosol.

## RESULTS

For small particles carrying a low number of charges a direct fit of the charge numbers is possible. When dealing with larger particles the dimension of the search space increases with the number of charges. Here, the charge distribution is described by a modified normal distribution.

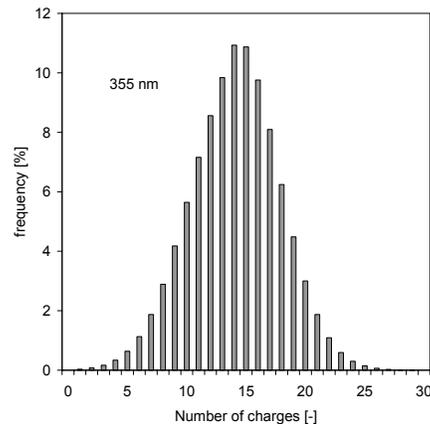


Figure 2: charge distribution, produced by the charger at 5 l/min (aerosol: ammonium chloride)

UFIPOLNET ([www.ufipolnet.eu](http://www.ufipolnet.eu)) is financed by the LIFE financial instrument of the European Community under No. LIFE04 ENV/D/000054.

Wiedensohler A. (1988). An approximation of the bipolar charge-distribution for particles in the sub-micron size range, *J. Aerosol Science*, 19, 387-389.

Medved A., Dorman F., Kaufman, S. L. (2000). A new corona-based charger for aerosol particles, *J. Aerosol Science*, 31 SUPP/1, 616-617.

Biskos G., Reavell K., Collings N. (2005). Unipolar diffusion charging of aerosol particles in the transition regime, *J. Aerosol Science*, 36, 247-265.

# Quantification of nanoparticle releases from surfaces

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Keywords: ultrafine particles, nanoparticles, release, modelling

Modelling fine and ultrafine particles in urban air requires quantification of particle releases from environmental or industrial surfaces under relevant conditions.

In many fields of application ultrafine particles or nanoparticles are employed to improve the properties of surfaces. Easy-to-clean coatings, corrosion protection and fiber reinforcing are some examples. Unfortunately, particles in this size range may be harmful to health if inhaled and deposited in the respiratory tract.

For the resuspension of particles in gas flows, the ratio of the drag force to adhesion force is the determining criterion. Larger particles easily detach from surfaces, but with decreasing particle size the decrease in drag force is larger than the decrease in adhesion force. Furthermore even when using a turbulent airflow, there is a laminar sublayer at the surface, reducing the effective drag force on the particles. Consequently, particles smaller than 10  $\mu\text{m}$  are usually not removed from a surface by air currents. For new nanoparticle-dotted products this has to be proven by the manufacturer.

In the presented project, a test device that quantifies nanoparticle releases from surfaces has been developed. It focuses on the particle reentrainment by drag force into an air flow. Furthermore it can be adapted to assess textile samples like gas filters or clothes with regard to particles released from the filter material.

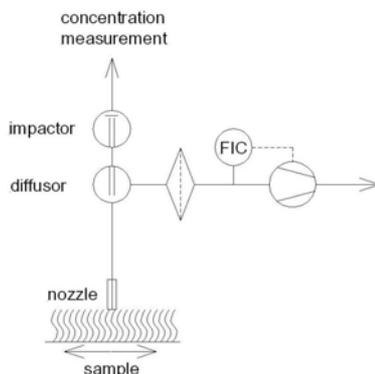


Figure 1. Scheme of the test device, a sample can be moved in two directions under the nozzle

The device consists of a nozzle and a sample carrier which can be moved in two directions. Through the nozzle, a controlled side channel blower draws a flow rate of up to 20 l/min. The nozzle has a

diameter of 5 mm. This narrow bore hole is necessary to attain large shear stresses at low flow rates. Therefore, it becomes possible to avoid unacceptably high dilution ratios. The examined sample can have a maximum area of 100 x 100 mm. It can be moved at a feed rate of 0 – 5 mm/s by a stepping motor. A controller enables the coverage of a selectable number of tracks. Consequently, a well-defined scanning of a surface sample can be achieved.

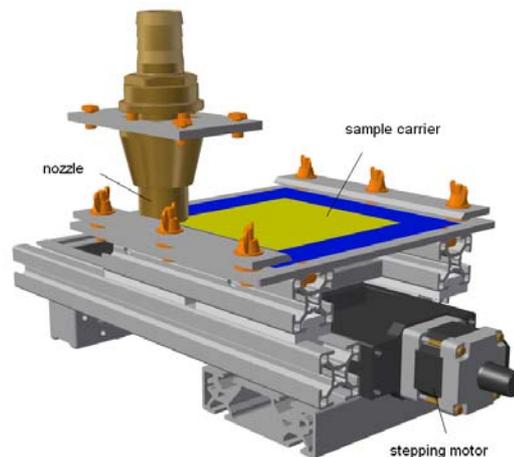


Figure 2. Test device to quantify particle releases from surfaces

The detection of the released particles is done by Condensation Particle Counter and a light scattering technique. Due to the different detection limits of the particle detectors, a distinction between particles in the nanometer and the submicron range at low concentrations is possible.

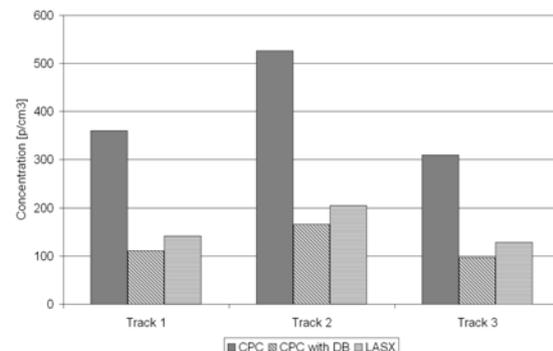


Figure 2. From the differences between the number concentration of both particle counters the release rate of particles < 100 nm can be calculated.

# Aerosol mobility spectrometry based on diffusion charging

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Keywords: electrical effects, charged particles, instrumentation, measurement

Aerosol spectrometers employed in the nanometer range base their measurement on the classification of charged particles in an electric field. Accurate measurement requires a well-defined charge status for the aerosol which is usually achieved by ionization of gas in the presence of radioactive sources such as Kr<sup>85</sup> (Liu, 1974). Diffusion charging has also been used for this purpose, but it generates a much higher charge level of the particles making the data inversion more difficult.

The aim of this paper is to model a particular diffusion charger and to evaluate the feasibility of using this charger in an aerosol spectrometer. The applicability of the device for environmental aerosols will be shown by comparing parallel measurements with an SMPS-system.

Charging of particles by means of an electrical field or corona-generated ions has been known for many years. Whitby and Clark introduced the diffusion charging of an aerosol by preventing the particles from undergoing strong field charging. This mechanism was used to produce a monotonic decreasing relationship between size and electrical mobility of the aerosol particles (Whitby, 1966). More recently this principle inspired the design of the so-called "corona-jet-charger" which attaches ions on particles by diffusion and forced convection in a mixing chamber (Medved, 2000).

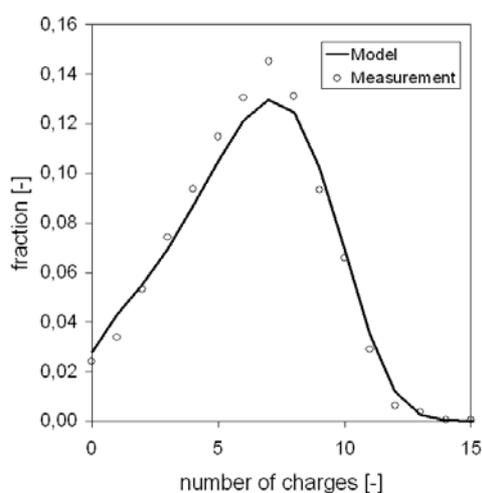


Figure 1. Calculated charge distribution compared to measured data (particle size 227 nm)

The charge distribution generated by this device can be calculated by applying Fuchs' limiting sphere

theory to the flow regime in the mixing chamber (taken from Biskos et. al., 2005). It is also possible to measure the charge distribution of particles leaving the charger by classifying monodisperse particles after the charger according to their charge status. An example of the comparison between model and measurement is shown in Figure 1.

By combining the corona-jet-charger with a DMA and an electrometer, a new aerosol spectrometer similar to an EAA and DMPS was developed. The advantage of this device is its simple set-up and the stability and robustness of the components. More problematic is the superposition of the mobility spectra of different particle size fractions due to their broad charge distribution. This leads to a lower size resolution compared to the SMPS. However, it is sufficiently precise for environmental aerosol quantification in air pollution networks.

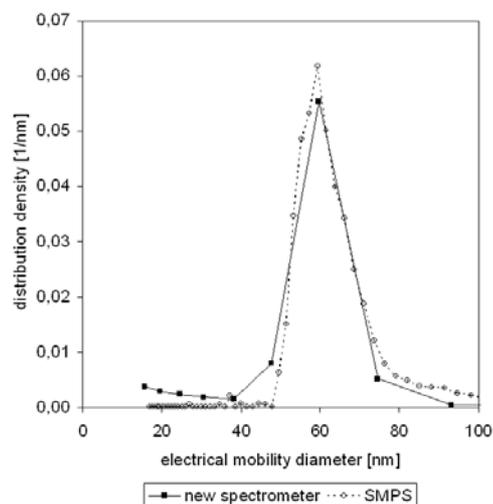


Figure 2. Distribution of 60-nm test aerosol showing SMPS and the developed spectrometer

UFIPOLNET ([www.ufipolnet.eu](http://www.ufipolnet.eu)) is financed by the LIFE financial instrument of the European Community under No. LIFE04 ENV/D/000054.

Liu, B. Y. H., Pui, D. Y. H. (1974). *J. Aerosol Science*, 5, 465-472.

Whitby, K. T., Clark, W. E. (1966). *Tellus*, 18, 573

Medved A., Dorman F., Kaufman, S. L. (2000). *J. Aerosol Science*, 31 SUPP/1, 616-617.

Biskos G., Reavell K., Collings N. (2005). *J. Aerosol Science*, 36, 247-265.

# An analysis of traffic-induced particle number emissions based on long lasting roadside and urban background measurements

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Keywords: vehicle emissions, traffic, street canyon, DMPS

In complex urban landscapes, the probability to be exposed to anthropogenic particles increases along with traffic volume and decreasing ventilation. Exposure research and urban atmospheric modelling therefore need emission factors for vehicular traffic basing on particle number rather than particle mass concentration. Because a relevant fraction of ultrafine traffic aerosols are formed by gas-to-particle conversion in the atmosphere downstream the vehicle pipes, measurements near traffic sources are required to capture the full effects of traffic on urban particulate number. Here, we report on particle size distribution measurements in an urban street canyon, and the associated inverse modelling that leads to traffic emission factors.

Long-term particle number size distribution measurements (3-800 nm) were conducted over the entire years 2005 and 2006 inside a street canyon in Leipzig, Germany (“Eisenbahnstrasse”) using a twin differential mobility particle sizer. The street features a daily traffic volume of about 10.000 vehicles at driving speeds of about 30 km h<sup>-1</sup>, which were counted in live-time by a video detection system. The share of heavy duty vehicles was about 5 % during day-time. To obtain a measure for the urban background outside the canyon, measurements were also conducted at a second station not directly affected by traffic emissions (“IFT”). By subtracting background from roadside concentrations, a measure for the true effect of traffic was obtained.

Figure 1 shows an exemplary diurnal cycle of particle number concentration on Mondays: Roadside concentrations exhibit two concentration maxima, which are associated with the morning and afternoon peak traffic times around 08:00 and 17:00.

The Operational Street Pollution Model (OSPM) was used to simulate atmospheric dispersion within the street canyon. The resulting dilution factor quantifies the degree of dilution of traffic aerosol before reaching the sampling point inside the street canyon. For each given large scale wind direction, OSPM also accounts for the turbulent transport induced by the topography as well as the moving traffic. Emission factors indicating the number of particles emitted by one vehicle per driven kilometre were calculated by dividing the concentration increment in the canyon by the dilution factor and the traffic volume (Ketzel et al., 2003).

The emission factor averaged over all vehicle types was found to vary between  $3 \cdot 10^{13}$  and  $7 \cdot 10^{14}$  veh<sup>-1</sup> km<sup>-1</sup>, depending on time of day, and also meteorological variables such as ambient temperature. We will provide a statistical summary of all observations and simulations, as well as their implications for the modelling of vehicular emissions.

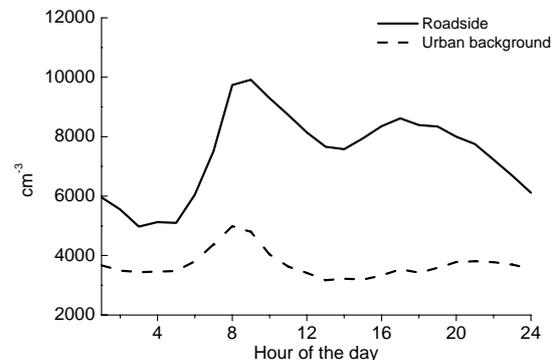


Figure 1: Mean diurnal cycle of particle number concentration (30-150 nm in diameter) on Mondays in 2005/06.

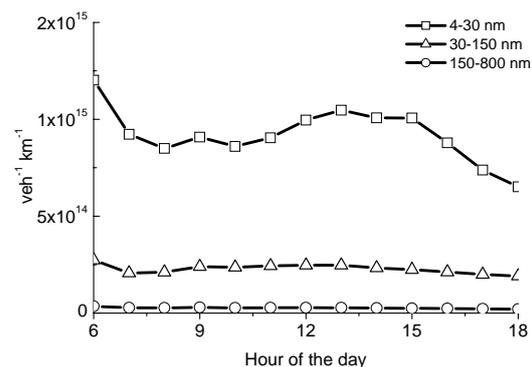


Figure 2: Mean emission factors averaged over all Mondays to Fridays in 2005/06.

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

## Microscale variations of atmospheric particle number size distributions in a densely built-up city area

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Keywords: DMPS, number size distribution, traffic, urban aerosols, exposure

The dominant source of aerosol particle number concentration in urban areas is combustion of fossil fuel in motor vehicles. Particle number size distributions near roads with high traffic are dominated by particles smaller than 100 nm in diameter. These anthropogenic submicron ( $< 1 \mu\text{m}$ ) and ultrafine ( $< 0.1 \mu\text{m}$ ) particles have moved into the focus of public health interest because they are suspected to contribute to acute and chronic disease in susceptible parts of the population.

To investigate the spatial and temporal variability of fine and ultrafine aerosol particles in the microscale environment around a street canyon, a specialised field experiment (PURAT: *Particles in the urban atmosphere*) was conducted. The measurements took place in the residential area around Eisenbahnstrasse in Leipzig, Germany. Particle number size distributions were measured continuously using differential mobility particle sizers over a time period of five months in winter 2005/2006 at the points (1)-(3) as indicated in Figure 1. Additional measurements at street level (4) between 06:00 and 10:00 were carried out to characterize microscale variations within the street canyon during a second period in July and August 2006. Regular direct intercomparisons between the instruments were made to ensure a maximum comparability of the size distributions at different points.

Our main findings are that the particle number concentrations at all four sites generally followed the general trend with concentrations backyard (3)  $<$  roof-top level (2)  $<$  street canyon (height 6 m) (1)  $<$  street canyon (ground level) (4) (see Figure 2). Particle concentrations in the backyard (3) were the lowest ( $\sim 5000 \text{ cm}^{-3}$ ), but particles on street level reached mean levels of  $25000 \text{ cm}^{-3}$ . The ratio of concentrations measured at different points did reach values of 10:1. Inside the street canyon, the measured concentrations were strongly influenced by the wind direction. The flow vortex in the street canyon (especially for northerly winds) influences the measurement points (1) and (4) directly. In this case they are downwind of the vehicular traffic as also shown by Voigtländer et al., 2006.

These first results suggest that the exposure to traffic-related particles in the outdoor environment of a densely built-up residential area exhibits substantial spatial variations. These variations need

to be considered in practical exposure assessments. The experimental results will be placed into context with simulations by a three-dimensional fluid model ASAM (All-Scale Atmospheric Model), which describes the dispersal of traffic aerosols within the domain at a spatial resolution down to 1.5 metres.

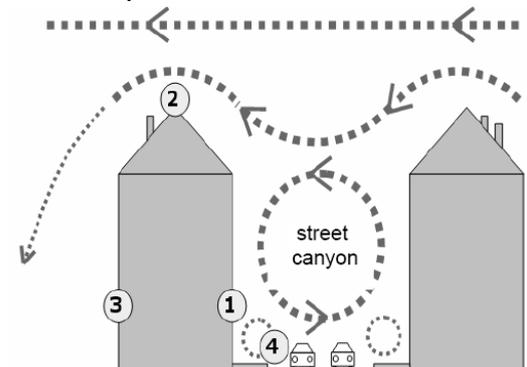


Figure 1. Sketch of the atmospheric air flow in the vicinity of the street canyon “Eisenbahnstrasse” in Leipzig, Germany. The numbers 1-4 indicate the measurement points for particle size distributions.

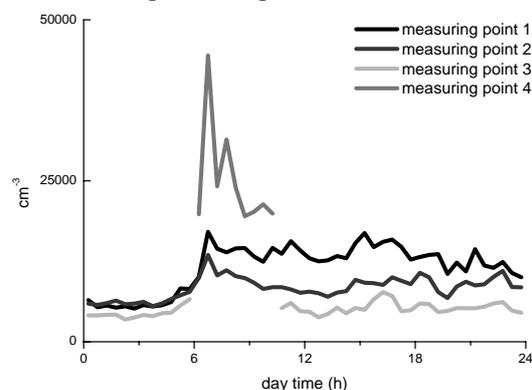


Figure 2. Mean diurnal cycle of particle number concentration (10-500 nm) at the four measurement points in the Eisenbahnstrasse, Leipzig.

Voigtländer J., T. Tuch, W. Birmili, and A. Wiedensohler. Correlation between traffic density and particle size distribution in a street canyon and the dependence on wind direction. *Atmos. Chem. Phys.*, **6**:4275-4286, 2006.

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# Continuous observations of particle size distributions (wide range) at the Frohnau Tower in Berlin with an altitude of 320 m

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Keywords: SMPS, particle size distribution, wide range, source apportionment, high altitude

## Introduction

The department of environmental engineering from the TU Berlin carried out a 1-year PM<sub>2.5</sub> measurement campaign (01.12.06-30.11.07) which is granted by the Senate of Berlin. The measurements were taken at 4 different sites to determine local, urban and regional sources of PM<sub>2.5</sub>.

One measurement site is the “Frohnau tower”, a tower at the northern outskirt in Berlin with an altitude of 345 m over ground. At the height of 320 m a measurement cabin was equipped with a gravimetric sampler (SQL 47/50, Sven Leckel Ingenieurbüro GmbH, [www.leckel.de](http://www.leckel.de)) for daily PM<sub>2.5</sub>-measurements, a continuous measurement device (SHARP 5030, Thermo Electron Inc. [www.thermo.com](http://www.thermo.com)) and a scanning mobility particle Sizer (SMPS+C type Vienna-DMA, GRIMM Aerosol Technik GmbH + Co. KG, [www.grimm-aerosol.com](http://www.grimm-aerosol.com)). The SMPS allows (in combination with the aerosol spectrometer EDM190) the detection of wide range particle size distributions (from 5 nm to 32 µm).

The objective of measurements at this altitude is the determination of transport and mixing processes of fine particles over Berlin. Figure 1 shows the measurement site “Frohnauer Turm”. The aerosol inlets are two stacked Whitby-devices. The cabin is equipped with different aerosol samplers, shown in figure 2.

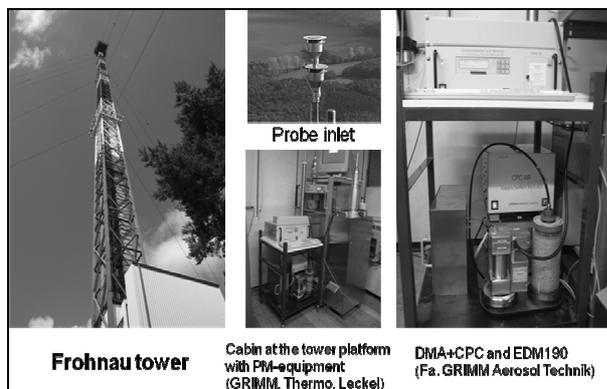


Figure 1: Measurement site “Frohnauer Turm”

## Results

Figure 2 shows an example of the wide range measurements for the 25.06.07. High numbers of ultra fine particles ( $d_p$  smaller 0,1 µm) could be detected.

This example demonstrates that even at an altitude of 320 m high densities of fine particles are transported and mixed.

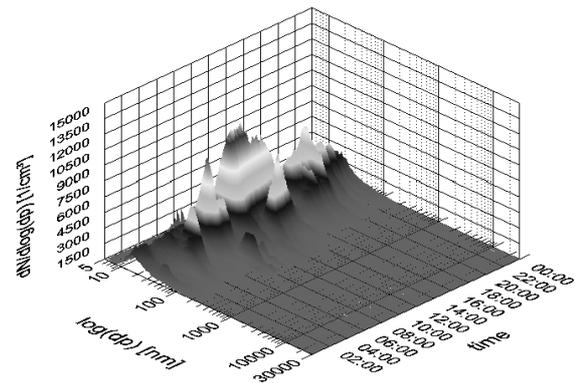


Figure 2: particle size distribution on 25.06.07

An example for the correlation of total particle counts to PM<sub>2.5</sub> measurements is given in Figure 3: The solid curve shows the diurnal concentration of PM<sub>2.5</sub> in µg/m<sup>3</sup> for the 21.06.07, measured with the SHARP 5030. The PM<sub>2.5</sub> concentration reached values of about 34 µg/m<sup>3</sup>, which is a relatively high concentration for June in Berlin. The number of total counts is illustrated by the dashed curve. The short duration of one SMPS scan allows a high time resolution of five minutes. A good agreement between the PM<sub>2.5</sub> concentration and the total particle counts can be seen. The difference between nighttime and daytime is probably caused by sources as well as meteorological parameters, e.g. the mixing layer height.

The influence of the mixing layer height is investigated by using a commercial small sized Lidar for stand alone operation as an integral part of the project (Mehnert, 2007; Pesch, 2007). The advective transports of fine particles are evaluated by calculating 2-dimensional backward trajectories using a model developed by our department.

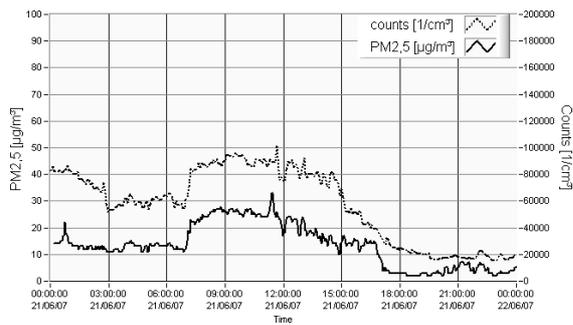


Figure 3: correlation of total counts and PM2.5  
(25.06.07, Berlin Frohnau)

The 8<sup>th</sup> of June is an example, where total counts and PM2.5 don't correlate well. The load of PM 2.5 is relatively small (between 10 and 25 µg/m<sup>3</sup>). They number concentration of particles varies between 50.000 and 140.000 counts per cm<sup>3</sup>.

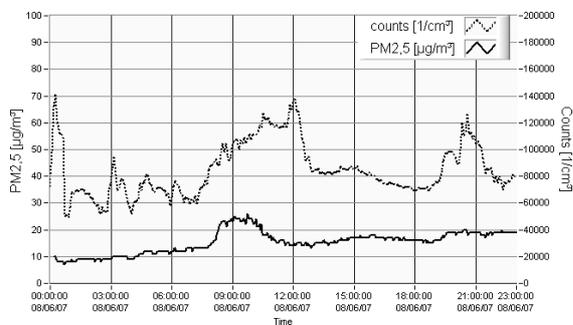


Figure 4: correlation of total counts and PM2.5  
(08.06.07, Berlin Frohnau)

### Outlook

The aim of further research is to determine the main sources and meteorological parameters leading to high particle concentrations over Berlin. The correlation of elastic backscatter Lidar data to ultra fine particle concentrations depending on relative humidity, diurnal as well as annual variations will be investigated.

Beside of the current analysis of chemical composition of PM2,5, the ultra fine particles will be analysed separately using an SMPS in combination with an electrical precipitator coupled with a GC-MS system.

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## Qualitative Characterization of Nanoparticle Emissions from Office Machines with Printing Function (Poster 2)

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Keywords: Nanoparticles, Chemical Composition, Laserprinters

This poster outlines some important aspects of the methodology for the characterization of particulate as well as gaseous emissions from office machines under controlled conditions. It also presents first results on chemical analysis of particles, emitted from laser printers.

**Methodology:** A 1 m<sup>3</sup> stainless-steel emission test chamber (ETC) is used which is especially designed for the measurement of gaseous and particulate emissions (BAM, 2003). In order to determine emissions of volatile organic compounds in small concentrations down to the lower µg/m<sup>3</sup> range and to gain good reproducibility in particle spectroscopy the test chamber must also meet a number of requirements (BAM, 2003):

- temperature, relative humidity, air exchange rate (AER) and air flow velocity must be kept constant at specific values,
- constant climate (23 ± 2 °C, 50 ± 5 % relative humidity),
- efficient air mixing with air flow between 0.1 and 0.3 m·s<sup>-1</sup> and air exchange rate between 1 h<sup>-1</sup> and 5 h<sup>-1</sup>,
- chamber walls of glass or polished high-grade stainless steel in order to minimize wall effects,
- minimized use of sealing materials because it may contribute to gaseous emissions or adsorptions and desorption effects,
- heat cleaning facility (up to 200 °C in order to tackle chamber memory effects),
- clean air and water supply with low VOC and negligible particle content.

A 13 stage (30 nm - 10 µm) low pressure cascade impactor (Dekati LPI-10) is used in order to take size resolved samples from the particles emitted. The impactor is operated at a rate of 10 liters per minute and polycarbonate foils are used as substrates. Particle samples are analyzed by three methods:

- ESEM (Environmental Scanning Electron Microscopy): Shape and size of particles.
- EDX (Energy Dispersive X-Ray Spectroscopy): Size resolved chemical composition.
- µRFA (Micro beam Roentgen Fluorescence Analysis): Size resolved chemical composition.

Figure 1 shows an ESEM micrograph of particles collected on stage 6 of the cascade impactor.

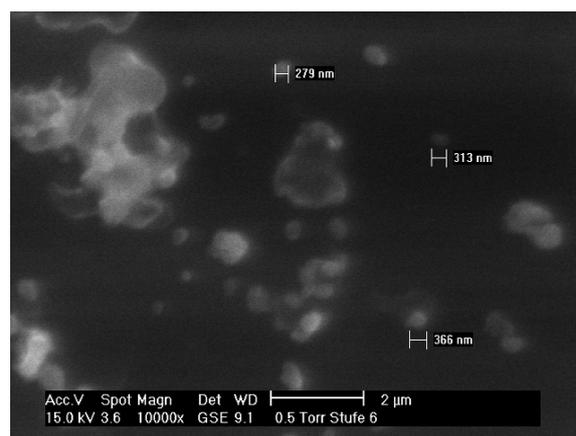


Figure 1. Particles collected on stage 6 of the cascade impactor.

The figure below reveals EDX-spectra taken from sampled particles of stage 7 to stage 2 of the cascade impactor (50% cut points of the impactor stages are given in brackets).

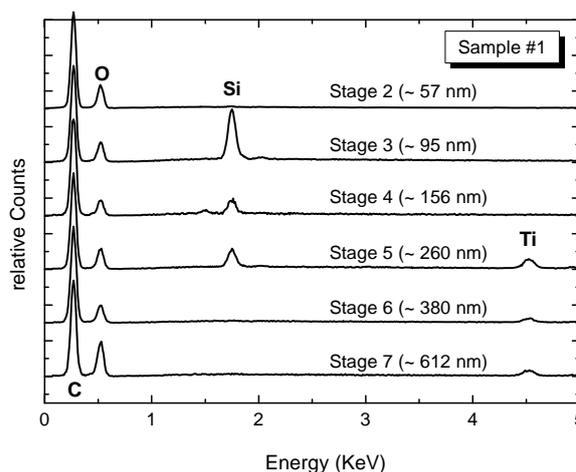


Figure 2. Comparison of chemical composition of sampled particles of different size by EDX.

Figure 3 presents data from µRFA of the same samples from the cascade impactor as in figure 2. The results of both spectroscopy methods applied are in concordance: There is a remarkable difference in the chemical composition of particles in different stages.

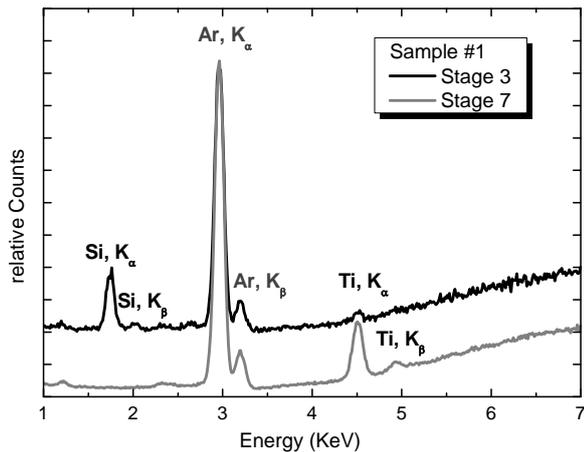


Figure 3. Comparison of chemical composition of sampled particles of different size by  $\mu$ RFA.

Only Titanium is found in stages 6 and 7, while Si is missing. In stage 2, 3 and 4, however, Si is the dominant contribution and Ti is clearly not visible. Oxygen and Carbon are visible in both spectra because these elements are ubiquitous. The Argon peak in figure 3 is a contribution from the ambient air.

The particle number concentration maximum for the printer in this example is around 110 nm diameter. Thus, the number concentration maximum consists of small particles containing Si, while the mass maximum consists of bigger particles containing Ti.

This work was supported by the German Federal Environment Agency (UBA, 204 95 373).

BAM (2003), *Test method for the determination of emissions from hardcopy devices with respect to awarding the environmental label for office devices RAL-UZ 62, RAL-UZ 85 and RAL-UZ 114*, BAM 06/2003, [www.blauer-engel.de](http://www.blauer-engel.de)

# Quantitative Characterization of Nanoparticle Emissions from Office Machines with Printing Function (Poster 1)

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Keywords: Aerosols, Nanoparticles, Emissions, Laserprinters

Office machines with printing function, i.e. laser printers and photocopiers, may be substantial sources of gaseous emissions as well as emitters of fine and ultrafine particles (Bake & Moriske, 2006; Jann & Wilke, 2006). In the last years symptoms such as asthma and pseudo allergic inflammations of the respiratory tracts, irritations of the skin and eyes, headache and sick building syndrome have been linked to these emissions (Kagi, 2007; Jaakkola, 2007; Smola, Georg & Hohensee, 2002).

Although it is obvious that laser printers and copiers are sources of particles, neither the qualitative nor the quantitative details of the particle emissions are understood in full detail. A reliable and comprehensive quantitative and qualitative database on those emissions is still lacking. Such a database could enable environmental toxicologists to estimate the related health risks in an appropriate way.

However, few studies on emissions have already been published (He & Morawska, 2007), revealing methodical shortcomings like the following:

- different environments for emission tests (outdoor, test chambers, offices),
- different instrumentations and measuring ranges,
- different measuring conditions (e.g. air exchange, aerosol background level),
- different methods of data analysis.

Due to these uncertainties it is at the moment nearly impossible to compare results from different working groups and to get a consistent overview. Consequently, the conclusions regarding health effects, drawn from different studies, are not consistent. What is necessary in order to eliminate the discrepancies and to provide reliable and comparable data? A standardized and determined protocol is needed in order to identify gaseous and particulate emissions from photocopiers and laserprinters.

BAM has worked out such a protocol, which comprises a description of the experimental setup as well as a step-by-step test procedure (Seeger et al., 2006; BAM, 2003). BAM now utilizes this time- and size-resolved quantitative and qualitative method for the characterization of laserprinters and office machines with printing function under controlled conditions.

This poster presents some results of recent measurements. The particle spectrometers (TSI 3936,

Grimm 1.108) operate at 1 liter/minute and cover a size range between 10 nm and 20  $\mu\text{m}$  particle diameter.

Figure 1 reveals a comparison of the detailed particle emission behaviour of two laser printers from different manufacturers during printing and after. The graphs show particle size distributions versus time. Perpendicular lines indicate the printing periods. The intensity of the grey scale code marks the particle concentration levels (the respective concentration maxima were set to 100 % for better comparison). The measurements were taken in a 1 m<sup>3</sup> environmental test chamber at an air exchange rate of 4.4 h<sup>-1</sup>. The data are not corrected for air exchange.

Printer B (right) clearly shows a shift of the emission maximum from initially 35 nm to 55 nm at the end of the printing while the particle size maximum is not changing with time for printer A (left). It is also remarkable, that the size distribution maximum of printer A is much higher and reaches 125 nm in diameter. An other interesting detail is the fact that the emission maximum of printer A occurs at the end of the printing period while it is in the middle for printer B. For both printers - as also for many other printer models investigated and not shown in detail here - there is no particle emission observable above 500 nm diameter. As toner particles usually have diameters between 3 and 6  $\mu\text{m}$  our findings indicate that there is no direct emission of toner particles from laser printers and alternative explanations for particle emissions have to be found.

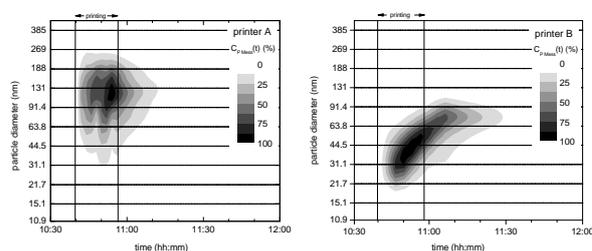


Figure 1. Comparison of particle emission characteristics of two different laser printers during printing and afterwards

Figure 2 shows a comparison of particle emission rates (unit particles per page printed) from 10 measurements on monochrome and colour laser printers, all operating in monochrome mode. Particle emission rates in the size range from 10 to 500 nm have been calculated according to the procedure de-

scribed in (Seeger et. al., 2006). Capital letters of x-axis labels refer to different manufacturers while figures denote diverse printer types. Thus the diagram compares 6 manufacturers and up to four printers of each brand. Except for types C1, C5, D1 and E2 measurements have been repeated. The results show a remarkably good repeatability of emission rates for each printer type and therefore the experimental efforts for constant conditions have been quite successful. The emission rates range from approximately  $2 \cdot 10^7$  particles/page (C3) up to approximately  $3 \cdot 10^9$  particles/page (D2). This is a huge span over more than two orders of magnitude.

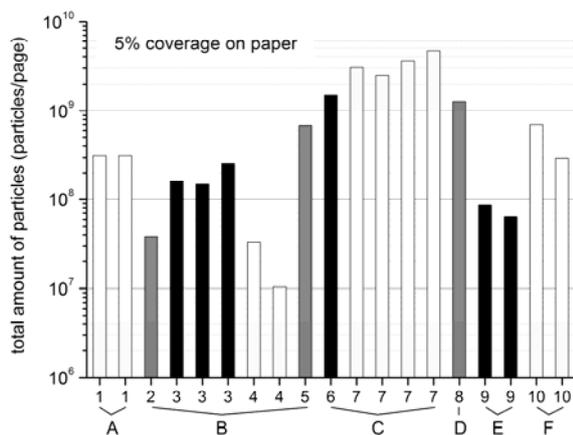


Figure 2. Comparison of particle emission rates for ten different laser printers in monochrome printing mode (capital letters indicate manufacturers, numbers indicate printer types)

Therefore our results indicate that even under comparable and controlled conditions different laser printers - even from the same manufacturer - reveal construction-conditioned extremely different particle emission behaviour both qualitatively and quantitatively. Both facts have to be considered in order to increase the comparability and reproducibility of results, in the estimation of health risks and in future measures to substantially reduce the exposure to particles from office machines.

Poster 2 presents first results on chemical analysis of particles, emitted from laser printers.

This work was supported by the German Federal Environment Agency (UBA, 204 95 373).

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# PM Measurements with ELPI (Electrical Low Pressure Impactor) and TEOM 1400a in the City of Vienna at the turn of the year 2004/2005

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Keywords: particle number size distribution, PM10, comparison of results, urban air, measuring technique

The Results from a PM-measuring campaign performed around New Years Eve 2004 are reported. Two different methods were used during the measurements, TEOM (Tapered Element Oscillating Microbalance) PM10 and ELPI.

With the TEOM true mass measurements can be performed in real time whereas the ELPI delivers valuable information about the particle number concentration based on its aerodynamic diameter. The particle size could be measured in twelve channels between <math>0.007</math> and

The TEOM Data show a peak of up to more than

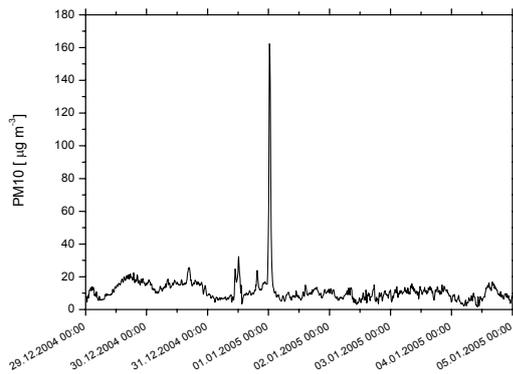


Figure 1. PM10 mass concentration at the turn of the year, obtained by the TEOM

The number concentration did not show significant peaks during midnight. The number of particles in urban air is dominated by the ultrafine particles being smaller than

A detailed evaluation of the ELPI-data show the mean particle diameter increases at the new year event as shown in Figure 2. This leads to an increased PM10 mass concentration.

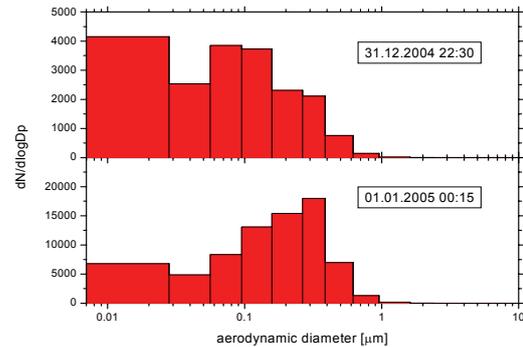


Figure 2. Particle number size distribution before and after turn of the year

As a conclusion of this measurements it seems to be obvious that both parameters, particle mass concentration and particle number size distribution are not necessarily in a fixed relationship with each other. Special events can show different effects on each parameter. This means that rather both parameters need to be separately measured to get a better idea of the composition of urban air.