STREET LEVEL VERSUS URBAN BACKGROUND CONCENTRATIONS OF SUBMICRON PARTICLES IN COPENHAGEN - MEASUREMENTS AND FIRST MODELING STUDIES

M. KETZEL[1,2,#], P. WÅHLIN[1], and R. BERKOWICZ[1]

[1]National Environmental Research Institute, Roskilde, Denmark

[2]Division of Nuclear Physics, Physics Dept., Lund University, Lund, Sweden

[#]Corresponding author. mke@dmu.dk (M.Ketzel)

Introduction

In order to characterise the size resolved particle number concentration in Danish urban environment we employ several Differential Mobility Particle Sizer (DMPS), covering a particle diameter range from 10nm to 700nm. Results of campaigns in 1999 and 2000 in Denmark were reported in Wåhlin *et al.* (2001a,b). Starting in May 2001 for the first time simultaneous particle measurements at a street location and a nearby urban background site could be performed. The objective of these measurements is to document the spatial and temporal variation of the particle size spectrum and to study the relation ship between street level and urban background concentration. For special meteorological conditions, as high wind speeds and high global radiation, regional particle formation events could be observed at both locations (Ketzel et al. 2001).

Location and Experimental

The two measurement sites are located ca. 2 km north west of central Copenhagen with a distance of ca. 500m. The street location at Jagtvej, is a busy street with about 25000 vehicles/day and is flanked on both sides with buildings about 25 m apart and about 18 m high. The urban background site is placed on the roof of the 20m high building of the H.C. Ørested Institute. Both places are permanent pollution monitoring stations operated in the frame of the Danish National Urban Monitoring Programme. A meteorological station is operated at the roof location.

The two similar DMPS systems consist of a 28cm custom built Vienna-type Differential Mobility Analyser in connection with a Condensation Particle Counter (TSI 3010), measuring particle concentrations in 15 electrical mobility channels in the submicron size range.

At the street location a traffic counting system was installed in July 2001, providing detailed information in traffic speed, traffic volume and traffic composition.

Results

This presentation gives an overview on the analysis done so far on this data set. Particle concentration measurements are presented in correlation with tracer gas measurements from the two locations, traffic profiles and meteorological parameter. Diurnal and weekly profiles elucidate the combined effect of a variable background concentration, variable meteorology and the traffic emissions.

The results show that it is essential to measure the background concentrations in order to isolate the traffic signal from the particular street from the background. The ratio background to total concentration measured in the street is in average 0.26 for NOx, 0.35 for CO and 0.41 / 0.60 for total particle number / area. The particle size spectrum in the background is variable due to changing contributions from long-range transport and show a diurnal pattern with a shift to smaller particle sizes during midday hours due to photo-chemical produced particles.

The measured particle number concentrations show in general very high correlation with NOx through a wide range of particle sizes. This indicates that dilution with the background air is the dominating process

for particles and NOx in a similar way and in a first approximation particles can be regarded as an inert tracer.
The average traffic particle emissions during day time hours (6-18) show a size distribution with maximum around 25-30nm independent on the heavy vehicle share. This indicates that petrol and diesel vehicles emit particles in a similar size range and the emissions have a similar particle/NOx ratio.
During night hours (0-5) the maximum in the emitted particle size distribution is shifted to smaller sizes of about 15-18nm. This shift to smaller particle sizes is related with an increase in the vehicle specific NOx and total particle number emission by a factor 2-3 and a reduced CO emission also by a factor 2-3. The dominating traffic during night hours are diesel taxis equipped with an oxidising catalytic converter.
The method of 'inverse modelling' (Palmgren et al., 1999) was applied to estimate average fleet emission factors typical urban conditions in Denmark. The dispersion model OSPM (Berkowicz, 2000) is used to estimate the influence of the meteorology on the distribution of the pollution inside the street canyon, while traffic counts allow to convert the total emissions to emissions per vehicle.
Emission factors per average vehicle are found in the range 1-1.5 gNOx/km, 10-16 gCO/km and $2\text{-}4 \times 10^{14}$ particles/km. This emission factors compare well with number emission factors reported in the literature.
A further analysis of the data-set including winter periods could reveal dependencies of the observed concentrations on the ambient temperature as it was observed by several groups before (Kittelson et al. 2000). In a planed future study we intend to investigate the relationship between the rural and urban background to estimate the contribution from all urban sources to the elevation of the regional background.

REFERENCES

Berkowicz, R. (2000): OSPM - A parameterised street pollution model. *Environmental Monitoring and Assessment* 65, 323-331.

Ketzel, M., Wåhlin, P., Le Bihan, O., Tymen, G. (2001): Measurements of ultrafine particles in Copenhagen. Presentation at the NOSA Aerosol Symposium in Lund 8-9 November 2001.

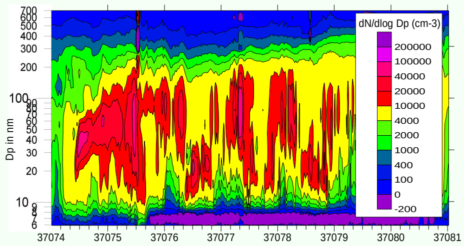
Kittelson, D. et al. (2000): Diesel Aerosol Sampling in the Atmosphere. *SAE Technical Paper Series* 2000-01-2122.

Palmgren, F., Berkowicz, R., Ziv, A., and Hertel, O. (1999): Actual car fleet emissions estimated from urban air quality measurements and street pollution models. *The Science of The Total Environment* 235, 101-109.

Wåhlin, P., Palmgren, F., Dingenen, R. Van and Raes, F. (2001a): Pronounced decrease of ambient particle number emissions from diesel traffic in Denmark after reduction of the sulphur content in diesel fuel. - *Atmospheric Environment* **35**: 3549-3552.

Wåhlin, P., Palmgren, F. and Dingenen, R. Van (2001b): Experimental Studies of Ultrafine Particles in Streets and the Relationship to Traffic. - *Atmospheric Environment* **35**(Supplement 1): S63-S69.

Street level versus urban background concentrations of submicron particles in Copenhagen



6. ETH Conference on Nanoparticle Measuremetns

M. Ketzel[1,2], P. Wåhlin[1] and R. Berkowicz[1]

1 - National Environmental Research Institute, Roskilde, Denmark

2 - Division of Nuclear Physics, Physics Dept., Lund University, Lund, Sweden

DMU Danmarks Miljøundersøgelser

National Environmental Research Institute of Denmark

LUND INSTITUTE OF TECHNOLOGY

Lund University

Outline of the presentation

- Motivation
- Data set at Copenhagen
- Results from measurements
  - time series
  - average diurnal / weekly cycles
- Estimation of fleet emission factors using "inverse modelling"

Motivation

- concern about health effects of ultrafine particles
- measure of the particle size distribution under “real world” conditions
  - average over the vehicle fleet mix,
  - driving conditions,
  - meteorology
- effect of the urban background
- estimation of number emission factors
- create a validation data set for aerosol models, to be included in our air quality forecast system

Dataset

- using permanent monitoring stations, street - background
- long term DMPS measurements, started May 2001
  - no TD; dry sheath air; 3 min scan time
- analysis of 12 weeks of simultaneous measurements 5-11/2001
  - mostly (Danish) summer conditions: 16±5 °C

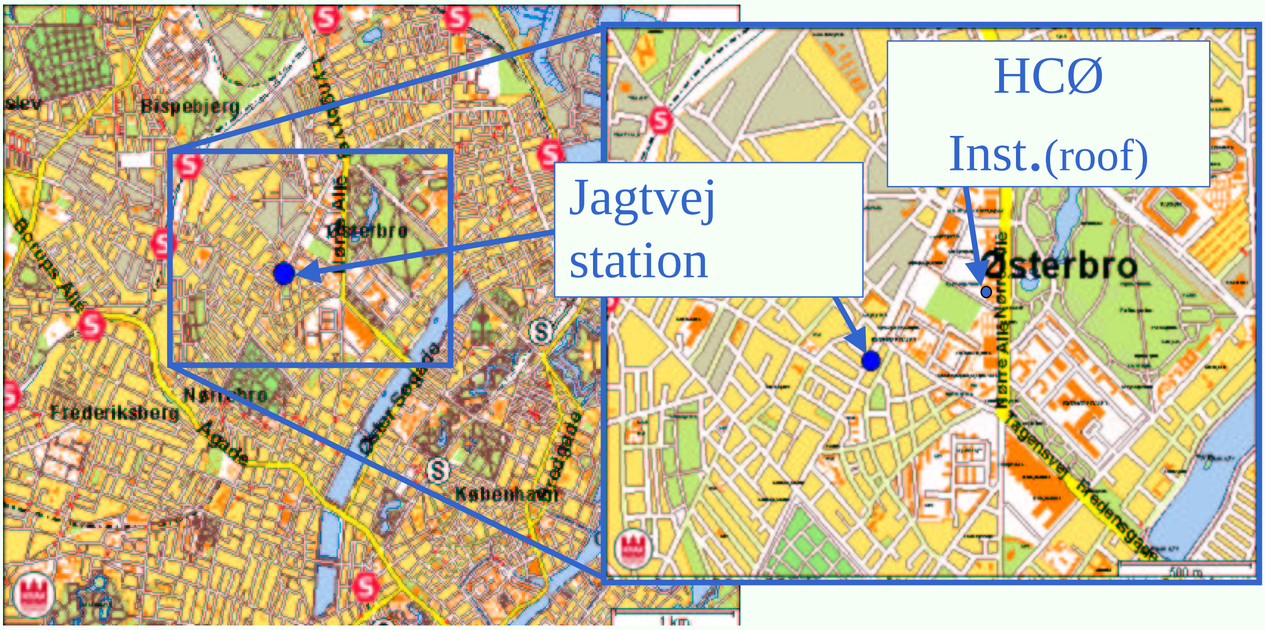
HCØ (background)

particles	***trace gas***	***meteorology***
DMPS 10-700nm	CO	wind direction
(UCPC > 3nm)	NO	wind speed
PM10 - Teom	NOx	temperature
	O3	RH
		global radiation

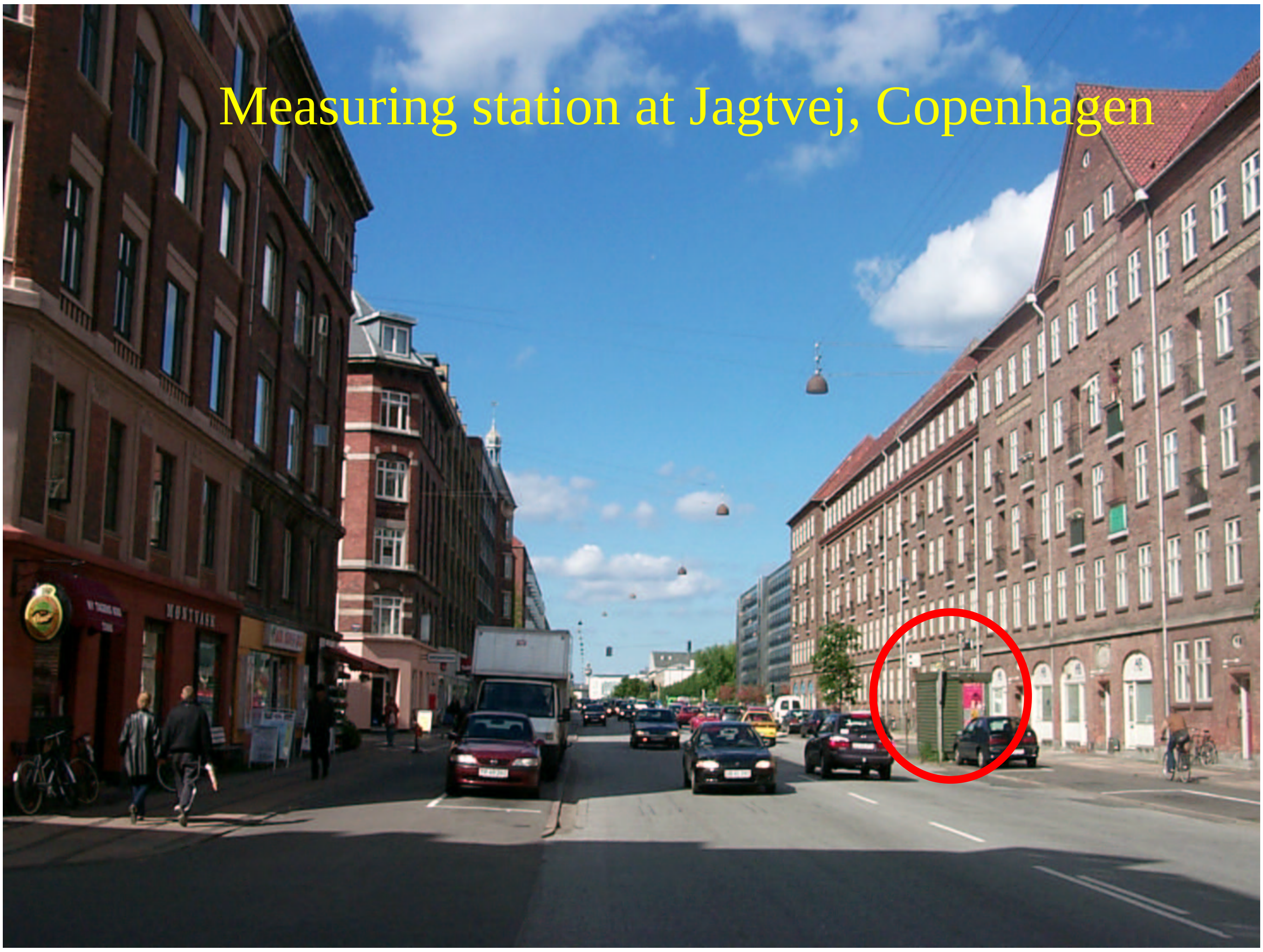
Jagtvej (street canyon)

particles	***trace gas***	***div***
DMPS 10-700nm	CO	traffic counts
PM10 - Teom	NO	rain
PM10 - filter packs	NOx	

Measuring stations near Jagtvej



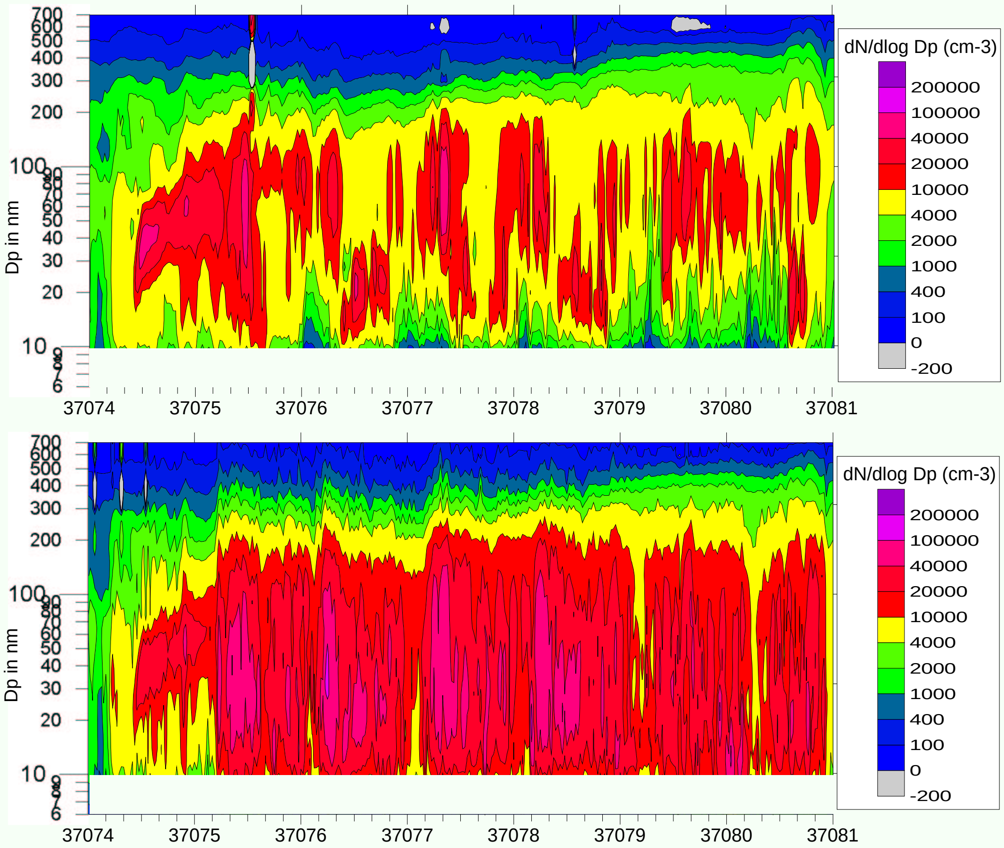
- 26 000 veh./day; 6-8% HDV
- Danish diesel: 50 ppmS; gasoline 60-70 ppm S



Particle size spectrum; 30 min averages (ca. 10 scans)
2.-8.7. 2001

HCØ (roof)

Jagtvej (street)

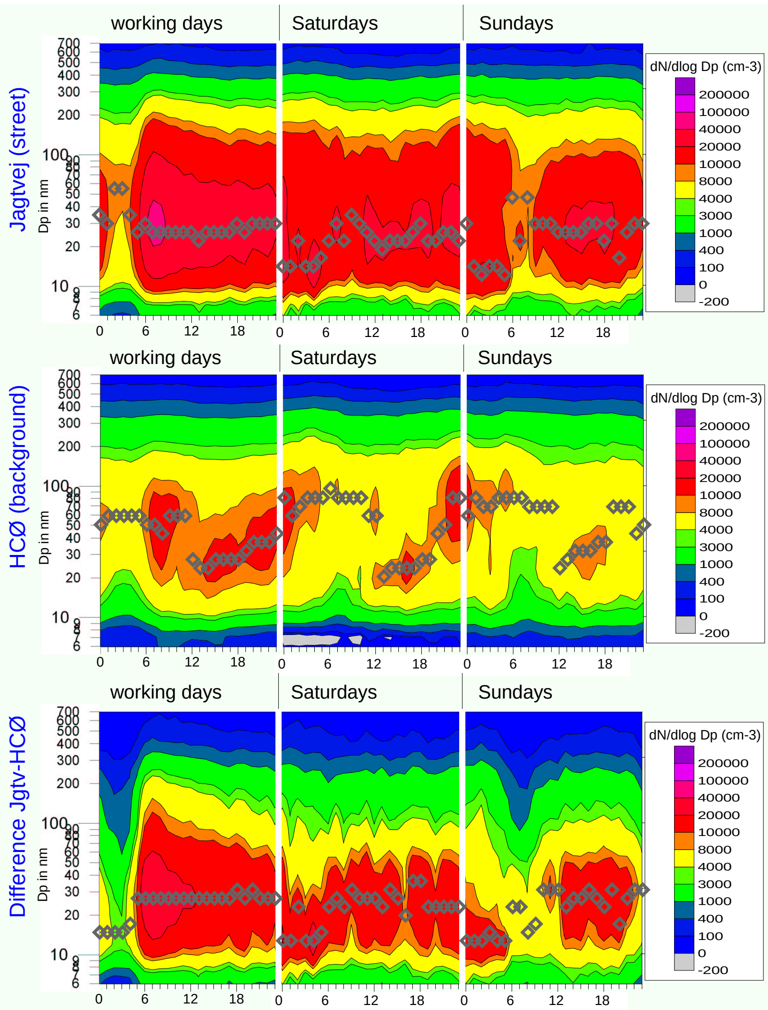


time (7 days)

Week 27

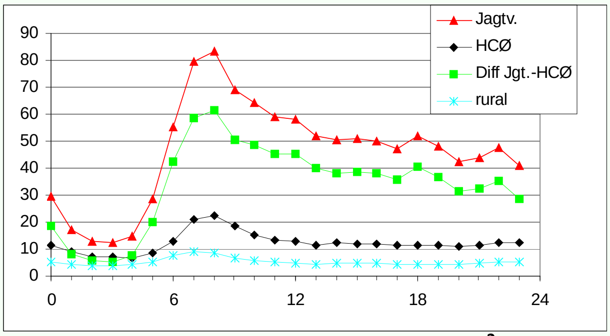
Average diurnal profile
periode 15-05 to 23-11-2001
ca. 12 weeks of data
(1h averages)

- variability in background
  - long range transport
  - mixing layer height
  - photo chemistry
- difference str.-backg.: constant mode diameter during day
  - diesel ≈ gasoline
- night time shift to smaller sizes
  - diesel taxi + oxicat.
  - nucleation ?!

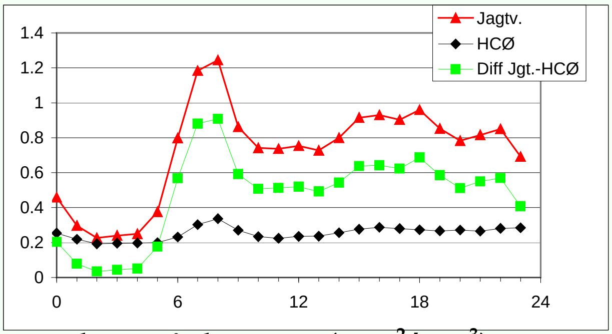


Diurnal variation of NOx, CO and particles during working days

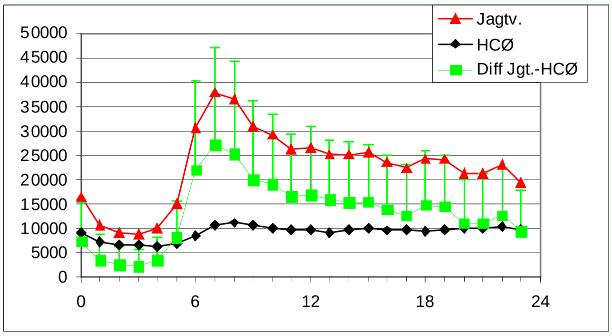
NOx (ppb)



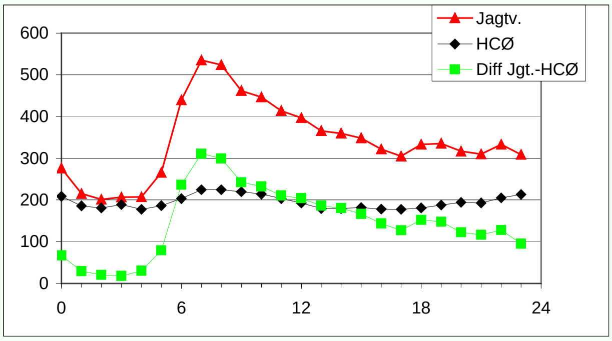
CO (ppm)



Total particle number (#/cm^3)



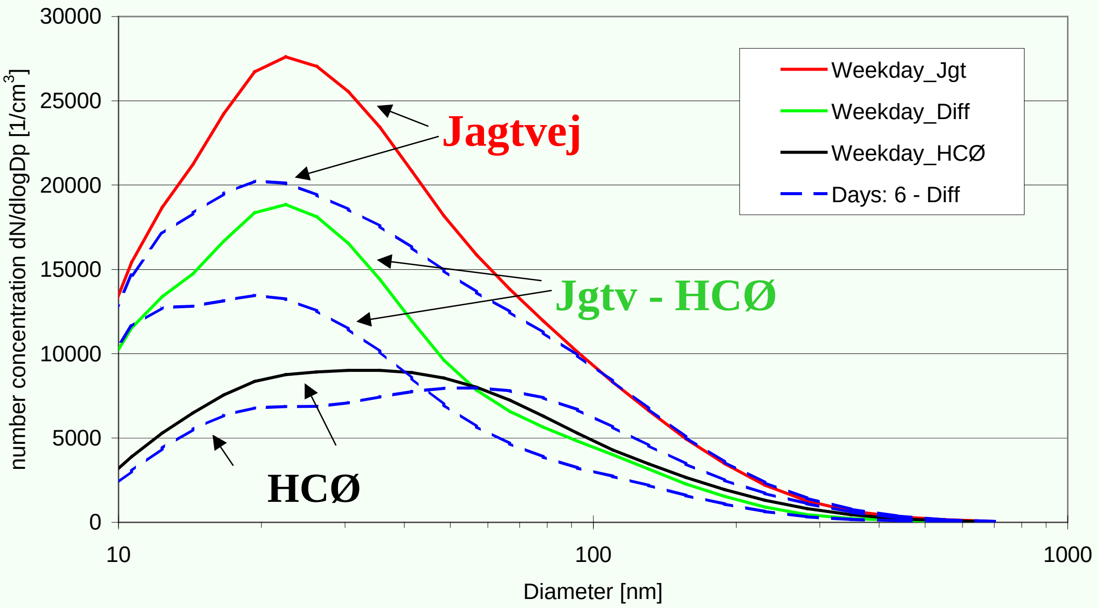
Total particle area (µm^2/cm^3)



- ratio roof/street conc. 0.26, 0.35, 0.41, 0.60 resp. for NOx,CO,ToN,ToA

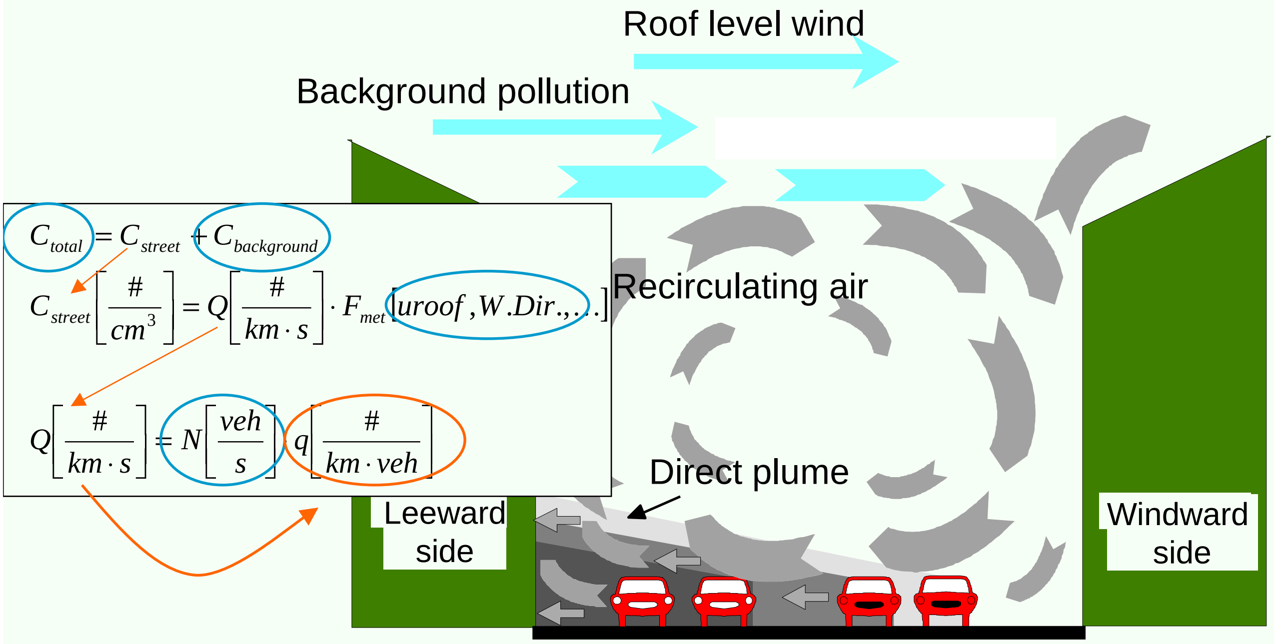
Average size distribution

Average periode 15-05 to 23-11-01



- background: 20 - 60 nm mode
- street: 20-30 nm mode

Estimation of emission factors using “inverse modelling”



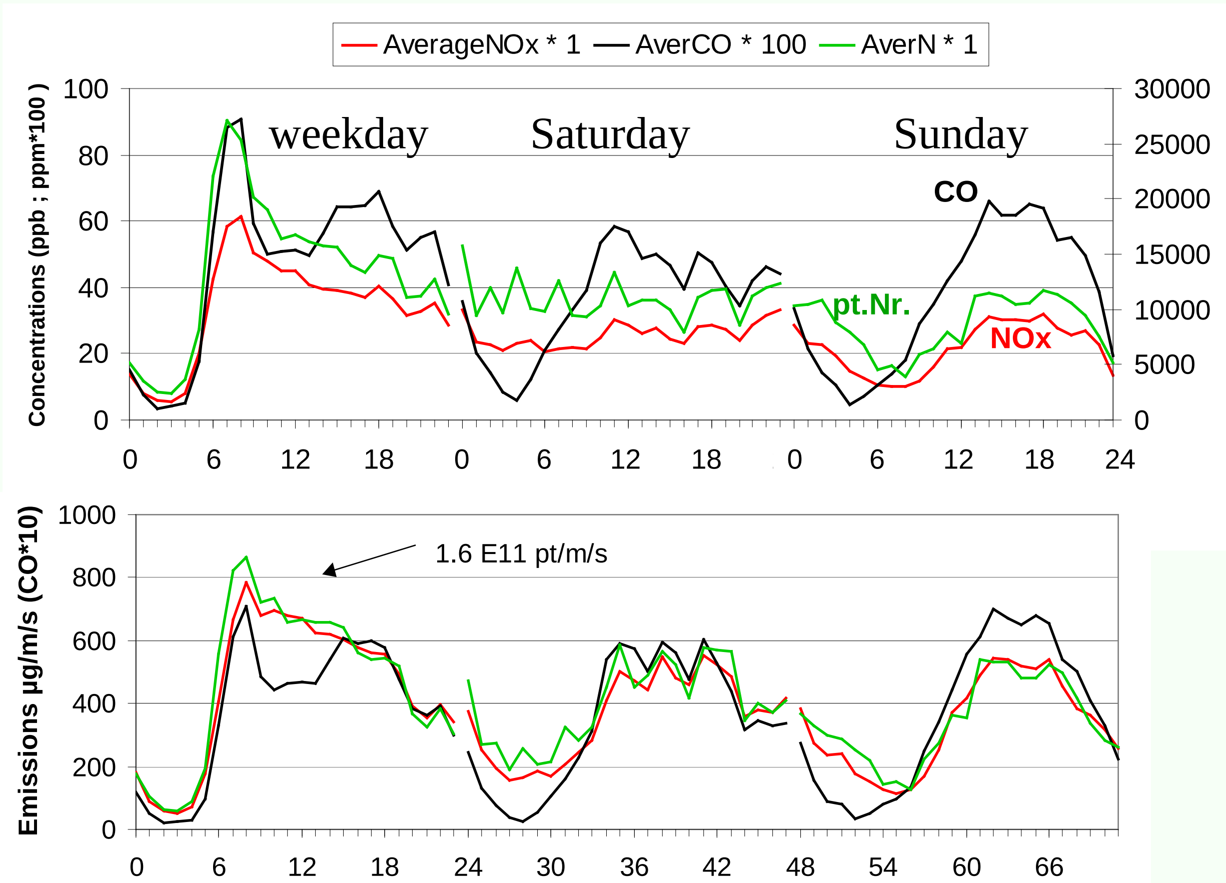
$$C_{total} = C_{street} + C_{background}$$

$$C_{street}\left[\frac{\#}{cm^3}\right] = Q\left[\frac{\#}{km\cdot s}\right]\cdot F_{met}\left[uroof, W.Dir., \ldots\right]$$

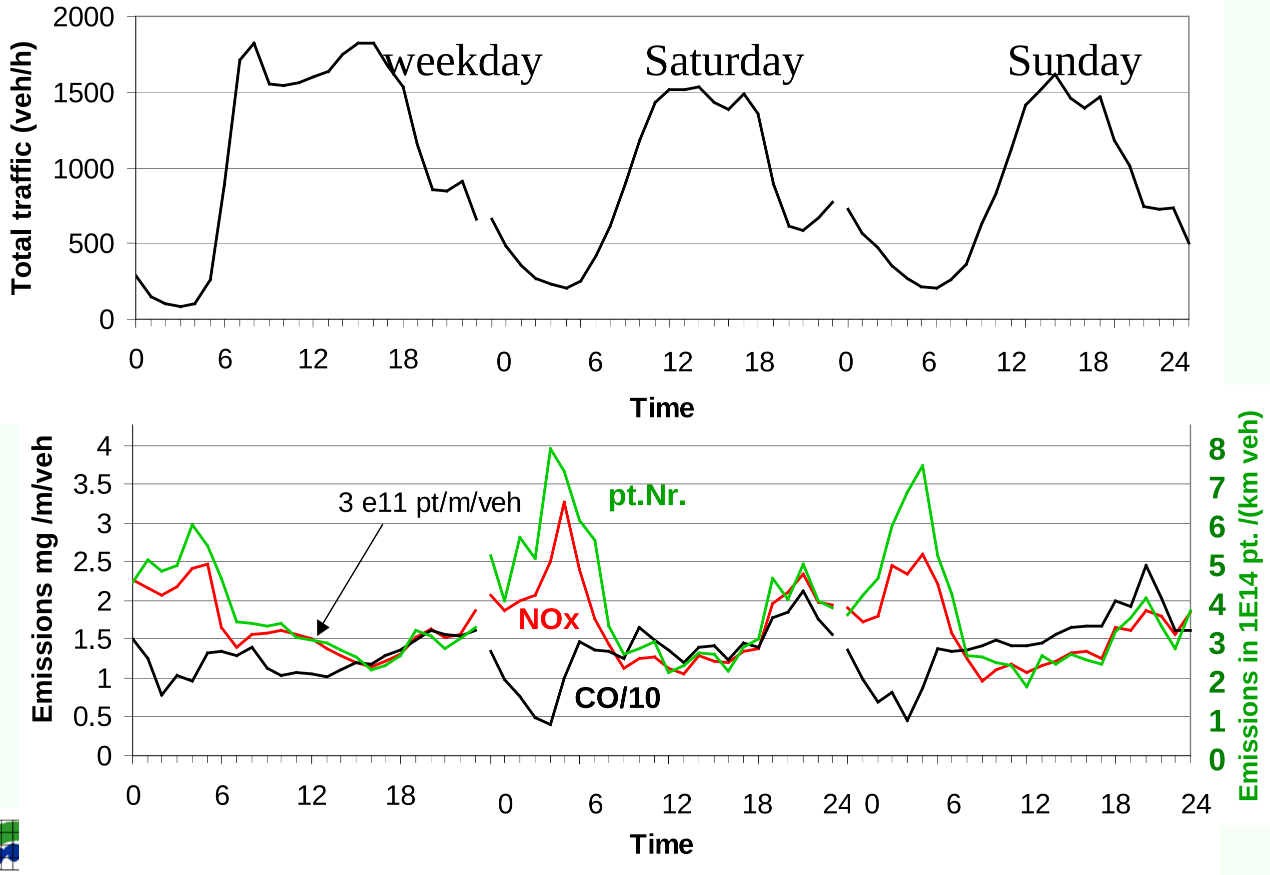
$$Q\left[\frac{\#}{km\cdot s}\right] = N\left[\frac{veh}{s}\right] q\left[\frac{\#}{km\cdot veh}\right]$$

- Operational Street Pollution Model (OSPM); Berkowicz et al. 1989...2002
- requirements for inverse modelling: longer time series + background measurements

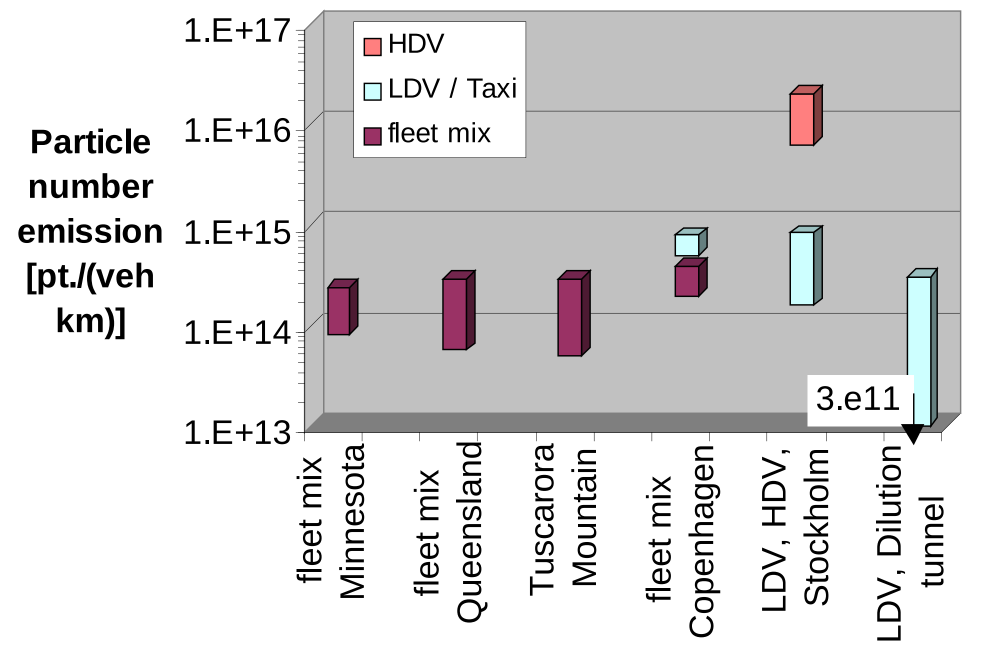
Estimation of emission factors



Estimation of emission factors



Comparison with literature data:



Kittelson, 5. ETH Conf. Zurich 2001
Jarmiska et al. 2001
Abu-Allaban et al. 2002
This study, Jagtvej 2001
Kristensson, Johansson et al .2002
Graskow et al. 1998/99

Summary

- urban background:
  – pt. size distr. shows a strong diurnal variation in all size ranges
  – important contribution to street level pt. conc. (more than NOx, CO)
- street level:
  – in general very high correlation particle number - NOx (R=0.93)
  – dilution seems to be the dominant process
- Nanoparticle events 10-20 nm
  – in background (+ street level) air; photo chemistry
  – night shift to smaller diameters; diesel taxi
- average cycles
  – constant particle mode 20-30 nm, diesel $\approx$ gasoline
  – fleet emission factors: 1-1.5 gNOx/km, 10-16 gCO/km, 2-4 $\times 10^{14}$ pt./km

- Next steps:
  – analyse winter measurements, temperature dependence?
  – look at rural -> urban relationship

Acknowledgements

- Danish EPA - Funding for the aerosol measurements
- Danish Research Council - Funding for PhD project
- Copenhagen University, Ole John Nielsen et al. - operating the background (roof) station
- NERI monitor lab. - running the measurements at the stations: Kåre Kemp et al.
- Finn Palmgren - manager of all the particle activities at NERI