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Daily patterns of traffic-generated particles and gaseous pollutants at a kerbside site in Milan, Italy.

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Introduction

Particle number size distribution measurements have been recently receiving growing attention since adverse health effects appear to better correlate with submicron particle number concentration rather than particle mass concentration (Sioutas et al., 2005; Su et al., 2006; Weichenthal et al., 2007). Though taking only a few percentage of particle mass, submicron particles account for 80%-90% of the particle number concentration (Wichmann et al., 2000) and have more reparability in deep lung region, inducing cellular damage (Hoek et al., 2010). Several studies have shown that motor vehicle emissions are the major source of fine and ultrafine particles in urban environments, namely responsible for the largest fraction of the particle number concentration in urban areas (Morawska et al., 1999; Hitchins et al., 2000; Morawska et al., 2008). Therefore, considerable interest has been directed towards the understanding of the evolution of number concentration, size, and composition of aerosol particles after their first release in the atmosphere, tracing plumes from tailpipe to road, from road to kerbside, from kerbside to urban or near-regional scale where aged particles become important for regional exposure levels of the population and climate effects (Zhang et al., 2004; Roldin et al., 2011).

The city of Milan has been classified as one of the most polluted areas in Western Europe (Putaud et al. 2004), afflicted with high particle levels exceeding PM10 and PM2.5 standards. However, as most of the investigations have been rather extensively focusing on PM mass-based features, the ultrafine particle number concentration has only been scarcely explored and limited to urban background sites (Lonati et al., 2011), neglecting traffic exposed sites.

In order to bring a first piece of information on this topic for Milan, this work deals with the particle number concentration data measured at a monitoring site directly exposed to urban traffic emissions, providing some insight in the relationships between particle concentration and both traffic and traffic-related gaseous pollutants.

Material and methods

Monitoring site

The monitors were placed in a kerbside position at a site located on the city centre ring road, with a daily traffic of about 55000 vehicles per day and traffic peaks of about 3500 vehicles per hour on rush hours. Concurrently with particle number measurements data for the traffic-related criteria pollutant (CO, NO and NO_2) and for the main meteorological parameters (temperature, humidity, wind speed and direction) have been recorded at 1-hour time resolution; furthermore, the traffic volume on the ring road passing in front of the monitoring site has been tracked daily through traffic data counts at 5-minute time resolution.

Instruments

Measurement campaigns for particle number have been performed during two separate sessions in October 2008. During a first 14-day campaign total particle number concentrations were measured using a butanol based condensation particle counter (CPC TSI 3775-TSI Inc., USA) which detects particles larger than 4 nm working at nominal flow rate of 0.3 litre per minute. For the measurement particles are first grown to a detectable size by condensation of a supersaturated butanol vapor and then detected by means of laser scattering.

During a further 7-day campaign total particle number and size segregated concentration data in 12 size bins in the 7 nm-10 µm size range have been concurrently collected by means of an Electrical Low Pressure Impactor (ELPI™ - Dekati Ltd., Finland), working at a 10 litre per minute nominal flow rate. The ELPI™ operating principle is based on particle charging in a unipolar corona charger, size classification in a cascade impactor (12 impactor stages and a final filter stage) and electrical detection through sensitive electrometers located in every stage of the impactor. The current signal measured at each impactor stage is directly proportional to the particle number concentration and size. Since ELPI™ measures ambient air particle concentration in real-time without any sample conditioning, a particle dryer based on a co-polymer Nafion® tube was used for removing humidity from the sample. In fact, since most ambient air particles are hygroscopic the particle-bound water increases the particle size affecting the particle size distribution with larger concentration data readings for the larger particles.
During both the campaigns particle number concentrations (PNCs) have been recorded at 1-min time resolution.

Results and discussion

On the average the PNC levels measured in Milan are in the same orders than those reported for trafficked urban sites (~$3{\cdot}10^4$ cm^{-3}); however, the typical daily pattern of particle concentrations displays a strong diurnal variation leading to 1-h average peak concentrations up to $6.5{\cdot}10^4$ cm^{-3}, typically on weekdays' early morning rush hours (Fig. 1). Particle concentration spots up to $1.2{\cdot}10^5$ cm^{-3} have been observed on both morning and evening rush hours. Saturdays' and Sundays' patterns reflect the corresponding patterns of the traffic, with relevant concentration levels on late evening hours too, associated to weekends' night-life traffic.
Notwithstanding the similar daily and weekly time patterns, the ELPI™ concentration data are about 50% lower than those measured by means of the CPC: this is partially due to the different size range, with ELPI™ data not accounting for particles in the 4-7 nm range, but more reasonably for the more perturbed weather conditions that occurred during the second campaigns, with some heavy showers washing the lower atmosphere.

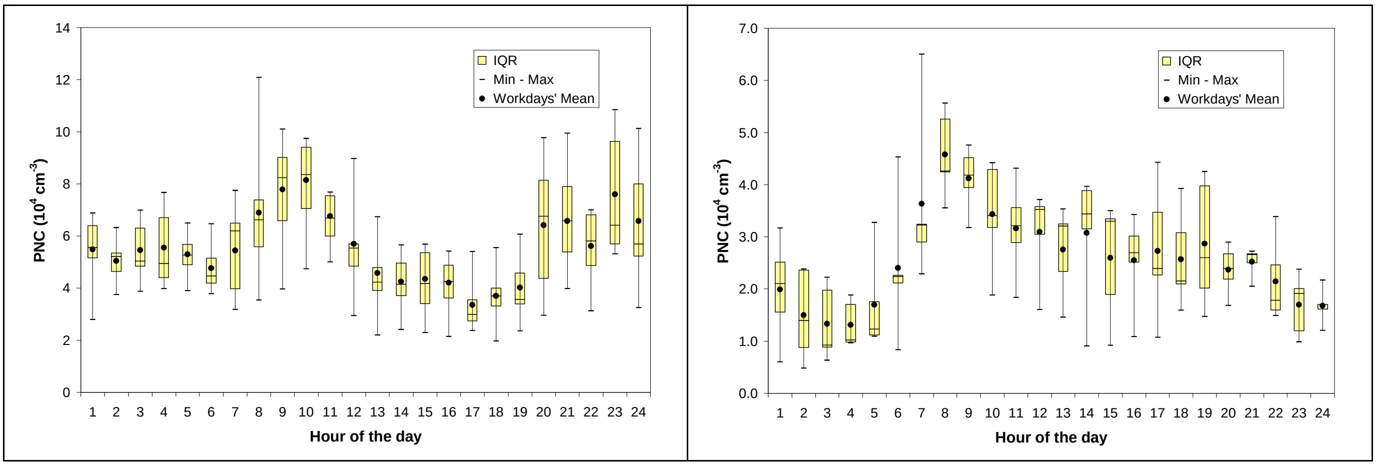

Fig. 1. *Daily patterns of particle number concentration on weekdays (Left: CPC data; right: ELPI data)*

Particle concentration is much more correlated with traffic-related pollutants than with the traffic flow: in fact, even though traffic flow and concentration levels of both particles and gaseous pollutants concurrently increase from the early morning hours until the peak rush hours (usually between 8 and 9 AM), their daily evolutions take different patterns, with traffic intensity rather uniform during the late morning and the afternoon hours whereas CO, NO, and PNC display a strong decrease in the early afternoon hours as a consequence of the diurnal evolution of the mixing layer height (Fig. 2). In general, the correlation between PNC and traffic-related gaseous pollutants is slightly higher for the diurnal data subset: PNC data are more strongly correlated with primary NO (R = 0.76), followed by CO (R = 0.48) and by secondary NO_2 (R = 0.34); due to the higher levels of NO compared to NO_2 correlation with NO_X is still relevant (R = 0.69).

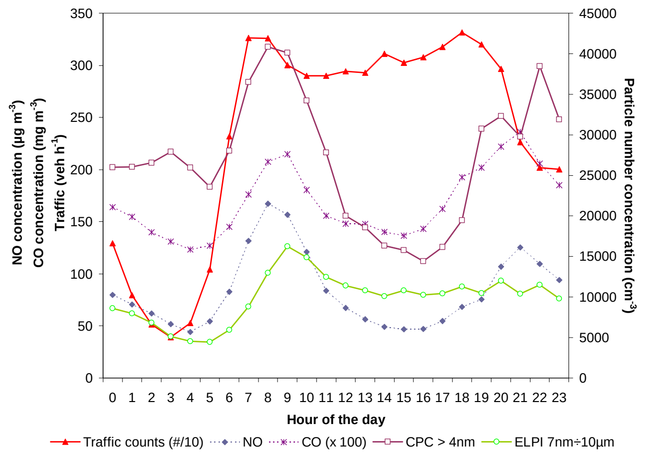

Fig. 2. *Weekdays' average daily patterns of CO, NO, PNC, and traffic*

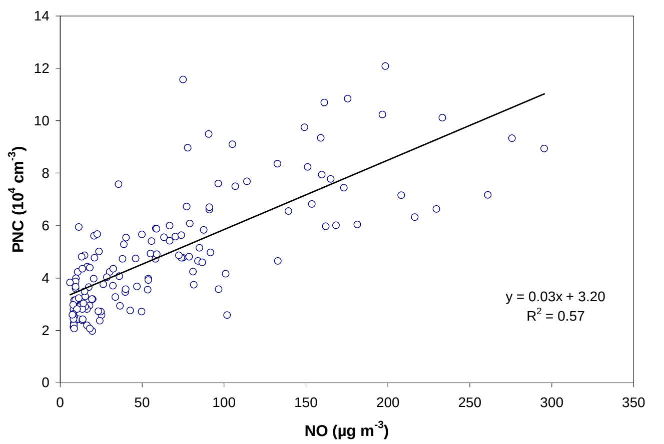

Fig. 3. *Scatter of 1-h NO and PNC diurnal data*

The size-resolved concentration data provided by ELPI™ data allow to analyze the daily evolution of the particle size distribution, pointing out its bimodal structure with a main mode located around 70 nm, coherently with the features of an urban traffic site, and a second mode located around 300 nm. On the average the size distribution maintains the same shape during the day (Fig. 4), reflecting the prevailing role of the local traffic emissions during the whole day, but with concentration levels progressively decreasing from the morning to the night hours as a combined consequence of the traffic flow pattern and of the mixing layer height. Furthermore, the cluster analysis performed on the 12 bins of size-segregated concentration data points out three main clusters of strongly correlated (R > 0.85) size bins (Fig. 5). These clusters groups size bins that can be associated to different particle generation processes, namely: freshly emitted engine exhaust nanoparticles (7-55 nm range), aged traffic-emitted particles (55-157 nm range) clustered with fine supermicron and coarse particles originated by surface wear and dust resuspension, and fine submicron particles (264-955 nm range) from local background.

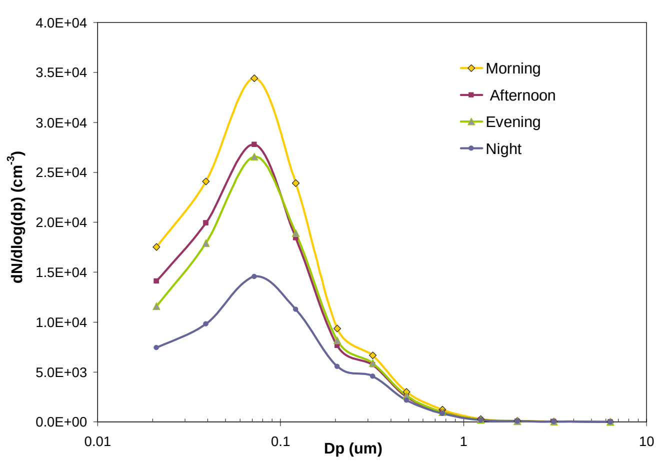

Fig. 4. *Average particle size distributions*

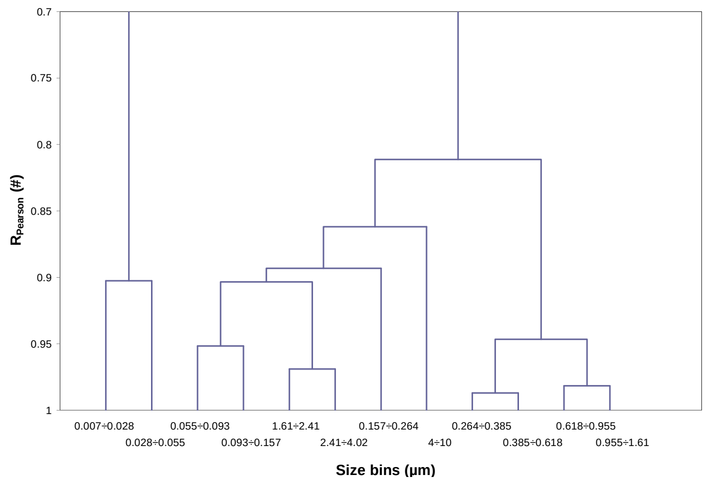

Fig. 5. *Cluster analysis dendrogram for the size-resolved particle number concentration data*

Conclusions

Particle number concentration data point out the particulate matter concerns in Milan are more a question of mass rather than number concentration, with concentration levels at the traffic-exposed site in substantial agreement with literature data for similar sites. PNC is well correlated with traffic-related gaseous pollutants (in particular with primary NO), whereas is not correlated with traffic due to the strong influence of the mixing layer daily evolution on PNC levels on afternoon hours.

The time pattern of the size-resolved particle concentration data points out three size clusters with similar behaviour; these clusters are reasonably associated with different sources, but still traffic-related particle generation processes, namely: freshly emitted engine exhaust nanoparticles, aged traffic-emitted particles clustered with fine super micron and coarse particles originated by surface wear and dust resuspension, and fine submicron particles from local background.

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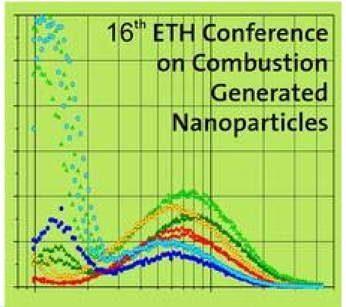

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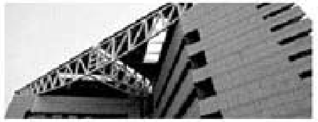

Daily patterns of traffic-generated particles and gaseous pollutants at a kerbside site in Milan, Italy

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Background & motivation

- Growing interest for particle number concentration data and related size distribution for health impact studies:
  - session 5a Health effects – 16th ETH Conference:

    "There is (some) evidence on the short term health effects of UFP but not yet clear and consistent" *K. Katsuoyanni, 2012*
  - hazard more correlated with number than mass concentration

    Peters et al., 1997; Wichmann et al., 2000; Donaldson et al., 2002
  - UFP (> 90% on particle number) toxic due to adsorption of toxic pollutants

    Sioutas et al., 2005
  - UFP deposition in the alveoli region of the lung and accession to blood circulatory system *Jaques and Kim, 2000*

Background & motivation

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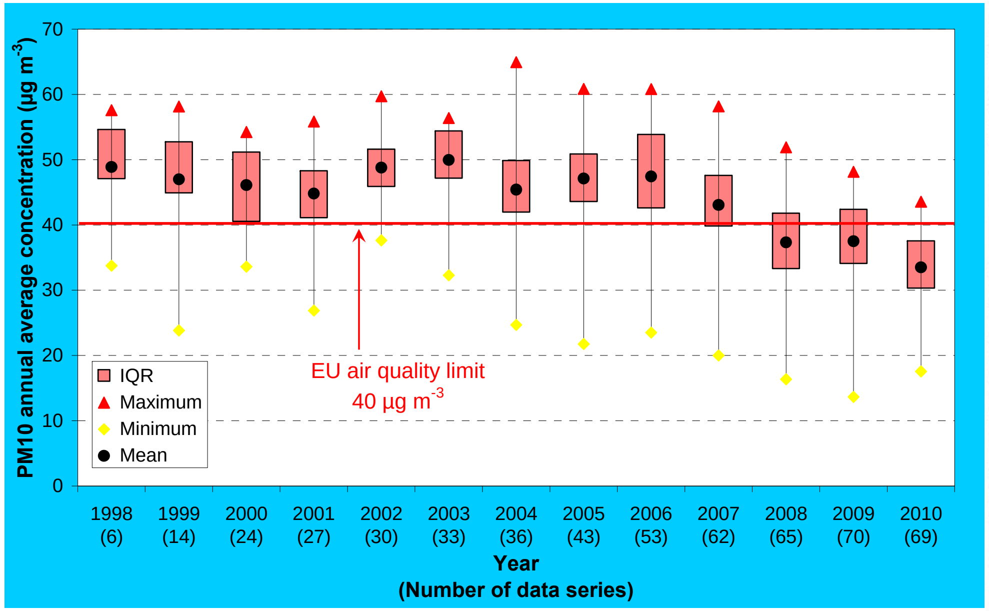
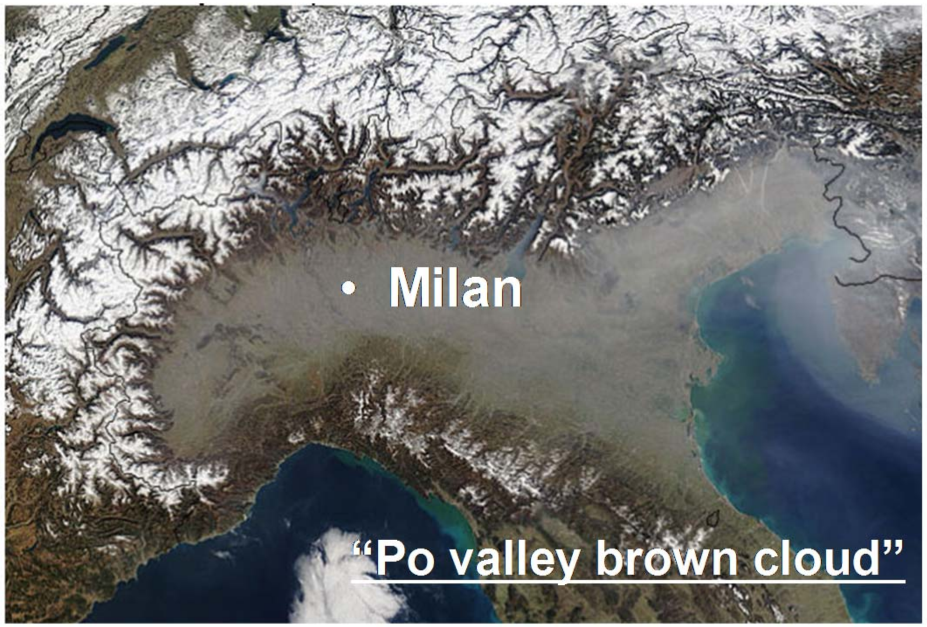
➢ Particulate matter concerns in Milan area

- extensive monitoring network for PM10 and PM2.5
- few studies for Milan area focused on particle number concentration
  Baltensperger et al., 2002; Rodriguez et al., 2007; Lonati et al, 2011
- no investigation of the relationship between traffic emission and particle number levels





Methods: Monitoring site

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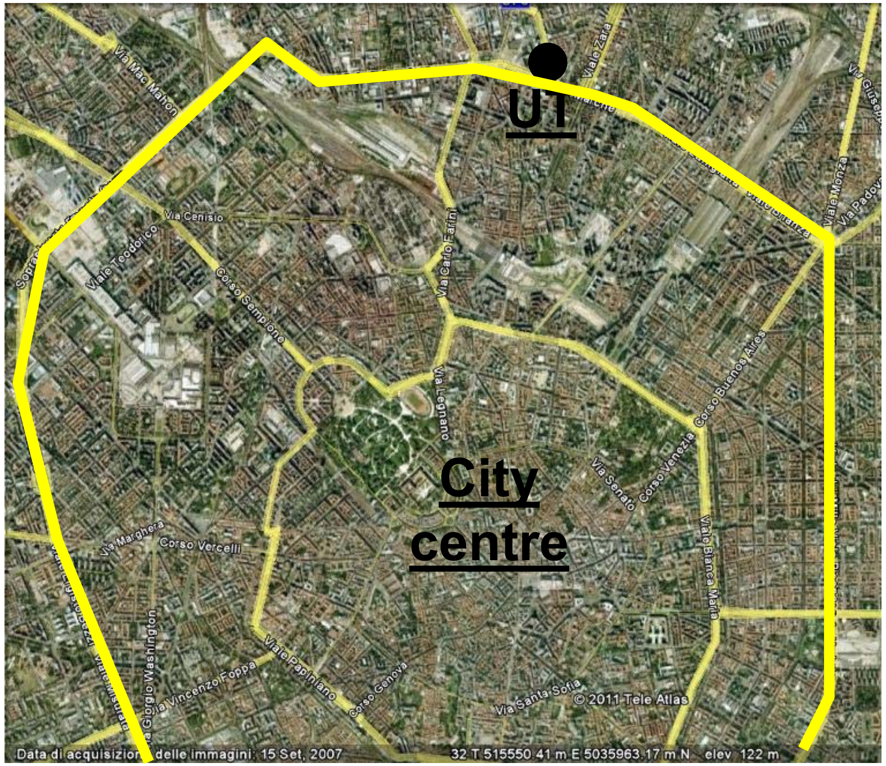
- Location: kerbside site on the city centre outer ring road
- Road features: double carriageway - 4 lanes



- Queue of idling or creeping vehicles due to traffic lights crossing
- Unpaved parking area on the road median

Methods: Monitoring site

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- Traffic data: vehicles counts at 5-min time resolution
- Air quality data: CO, NO_X (NO & NO_2) data at 1-h time resolution
- Meteorological data: wind speed and direction, RH, temperature

Methods: Monitoring site traffic

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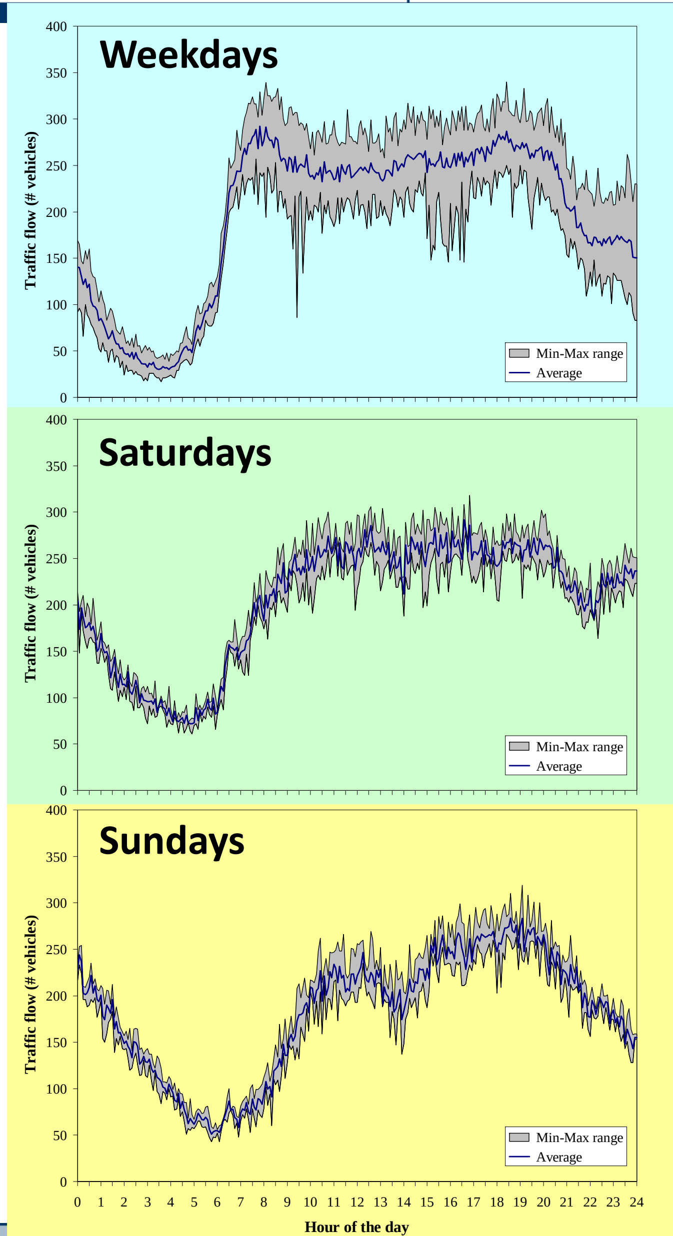
- Workdays' and Saturdays' daily traffic:
  - average 2800 vehicles hour^{-1}
  - range 600 - 3600 vehicles hour^{-1}
  - rather constant flow during daytime
  - Saturdays' night traffic
- Sundays' daily traffic:
  - average 1900 vehicles hour^{-1}
  - range 600 - 3000 vehicles hour^{-1}
  - no morning peak
- Traffic composition on rush hours:

morning		evening
70%	passenger cars (G & D)	78%
14%	motorbikes & mopeds	15%
16%	duty vehicles (Diesel)	7%



Methods: Instruments & monitoring periods

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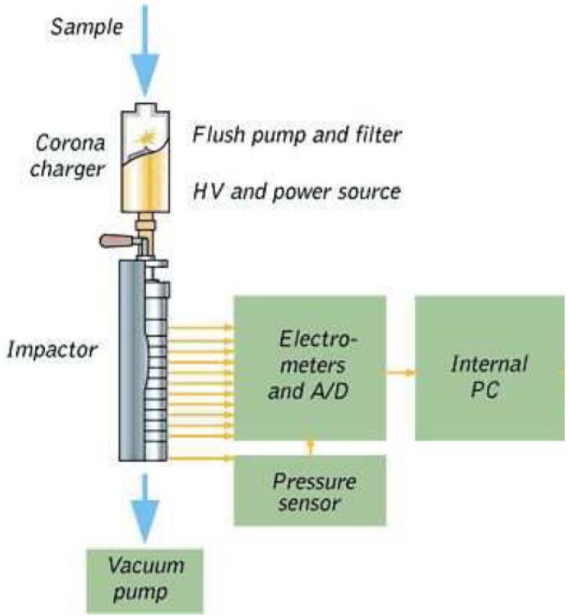
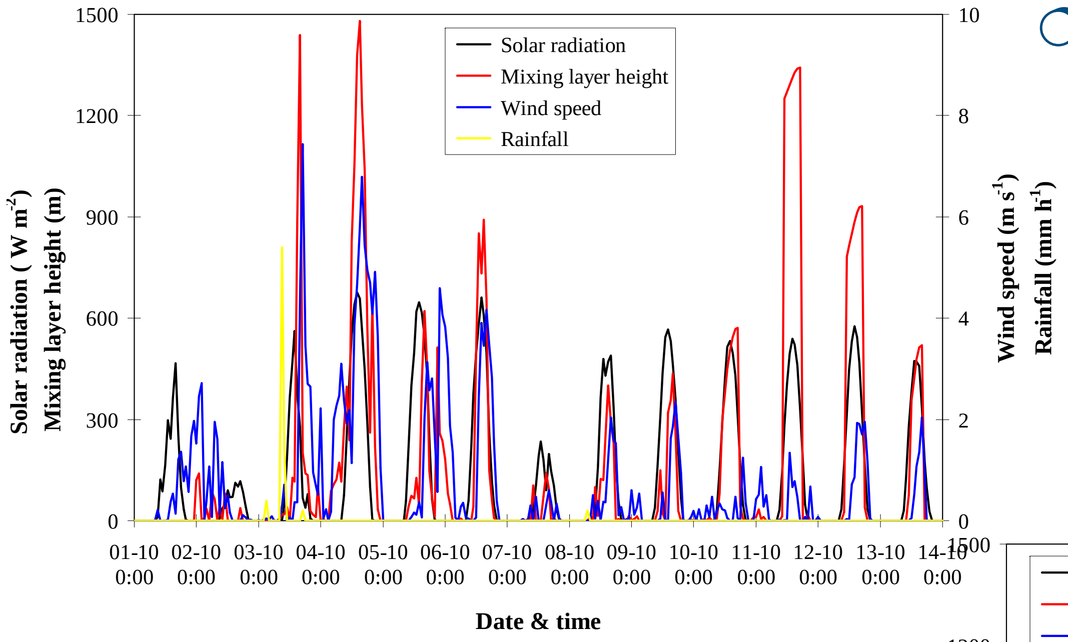
- Condensation particle counter (TSI CPC 3775)
  - Total particle number concentration (dp > 4 nm) at 1-min time resolution
  - Period: 1-14 October 2008

- Electrical Low-Pressure Impactor (DEKATI ELPI™)
  - Size-resolved particle number concentrations in the 7 nm-10 μm size range at 1-min time resolution for 12 size bins
  - Period: 23-30 October 2008

D_{min}	D_{max}	D_{50}
4.02	9.98	6.334
2.41	4.02	3.113
1.61	2.41	1.97
0.955	1.61	1.24
0.618	0.955	0.768
0.385	0.618	0.488
0.264	0.385	0.319
0.157	0.264	0.204
0.093	0.157	0.121
0.055	0.093	0.072
0.028	0.055	0.039
0.007	0.028	0.021

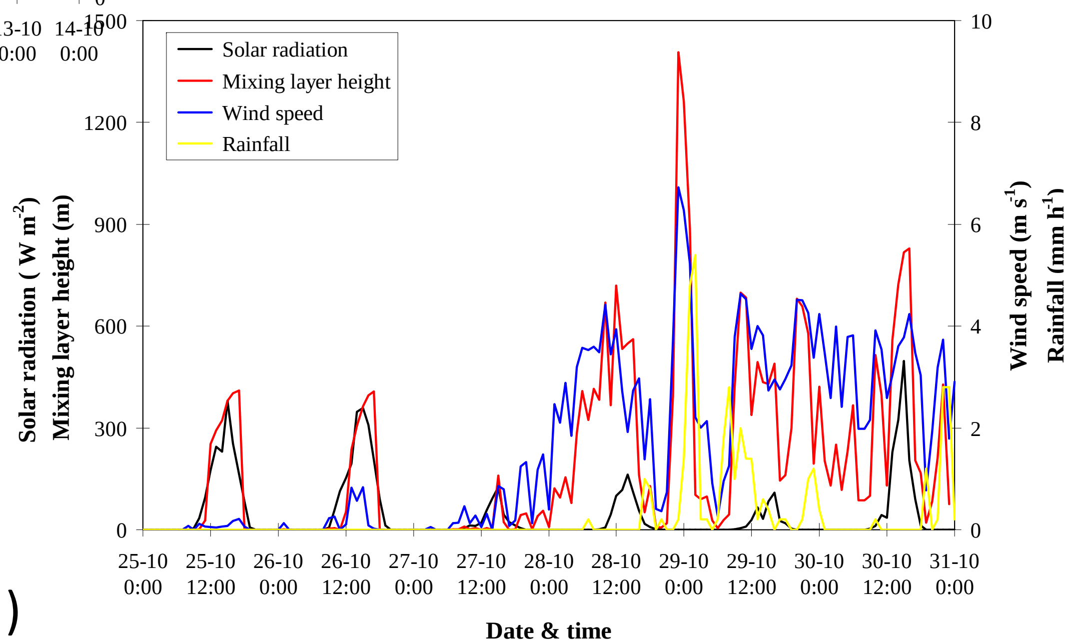


Methods: Meteo and PM data



- CPC campaign
  - Rather homogeneous conditions
  - Well defined daily evolution of the mixing layer
  - Just one short rain event
  - PM2.5: avg. 39 µg m^{-3} (15-63 µg m^{-3})

- ELPI campaign
  - More perturbed conditions
  - Higher wind speed and rain events on the final days
  - Mixing layer mainly driven by mechanical turbulence
  - PM2.5: avg. 39 µg m^{-3} (11-73 µg m^{-3})



CPC results: Weekly and daily patterns

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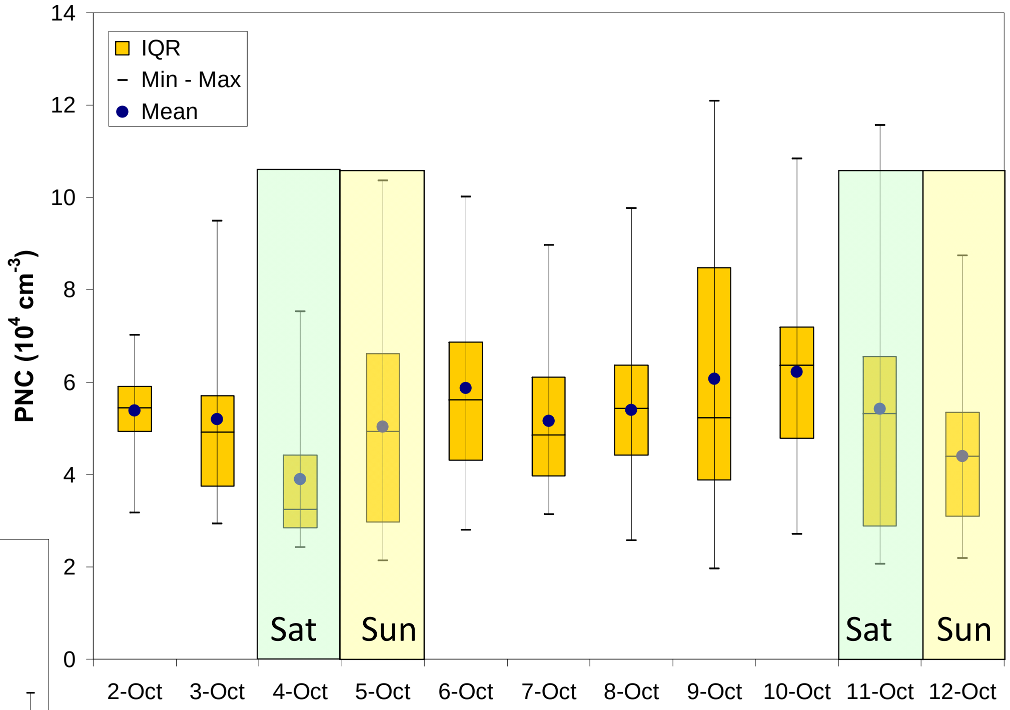
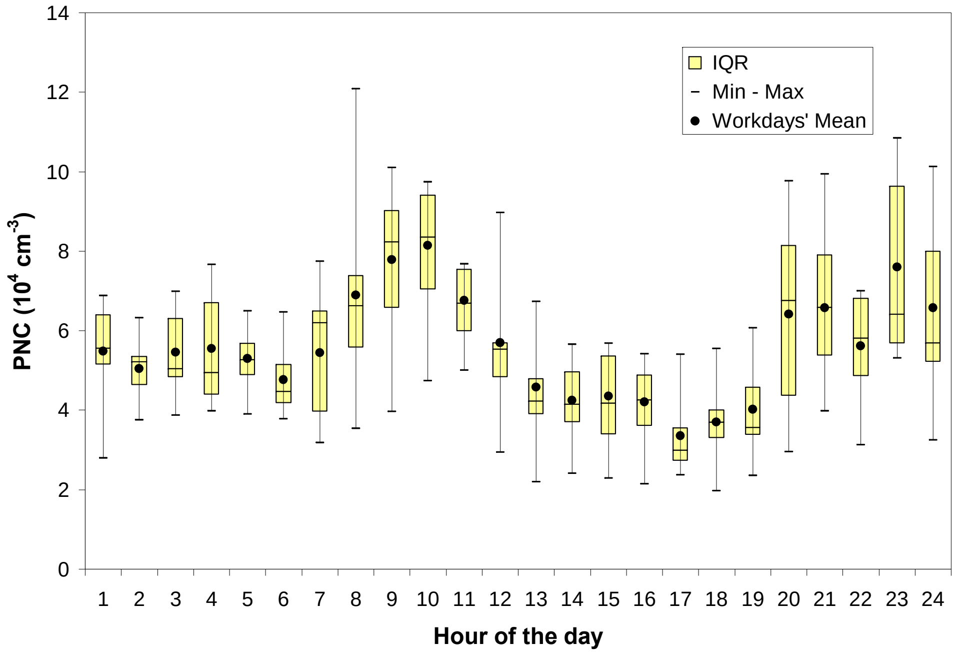
❍ Weekly pattern

- No significant changes either in PNC levels or in data dispersion
- Daily average within 4-6·10^4 cm^{-3}
- Peaks in excess of 10^5 cm^{-3} on both weekdays and weekends



❍ Daily pattern

- Typical 2-peak pattern
- Larger fluctuations on peak hours



CPC results: Weekly and daily patterns

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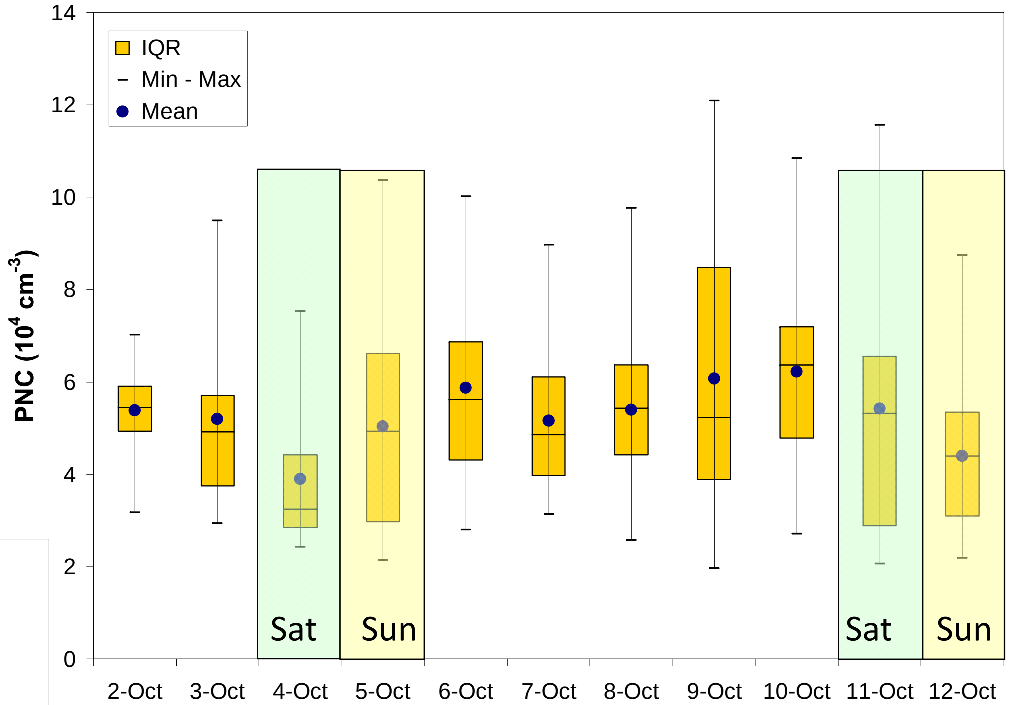
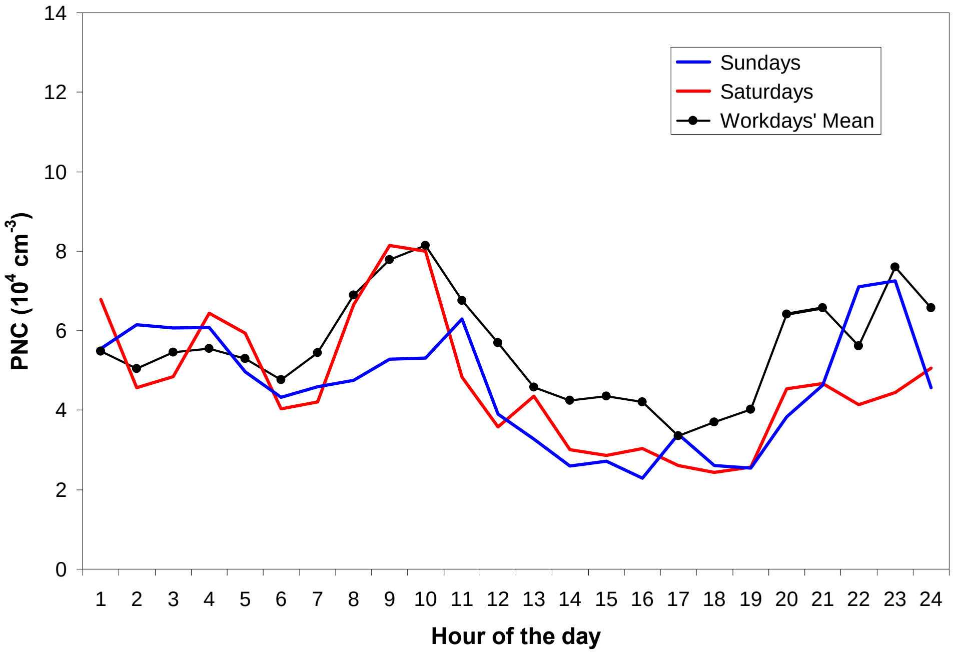
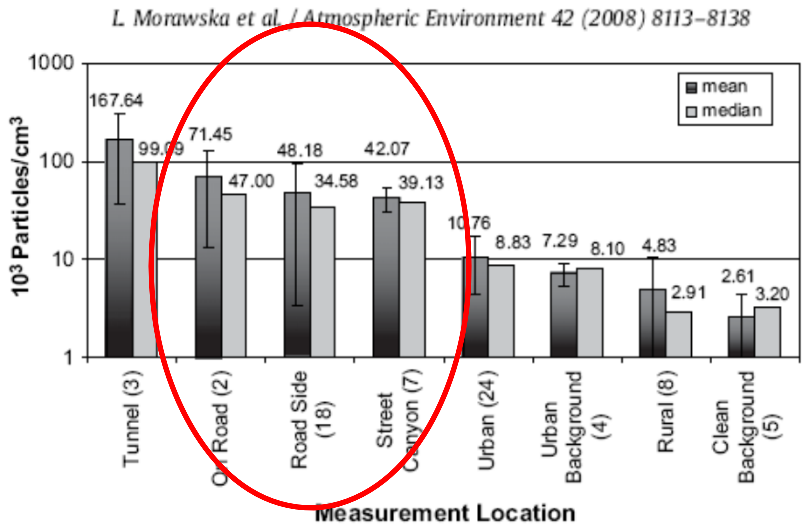




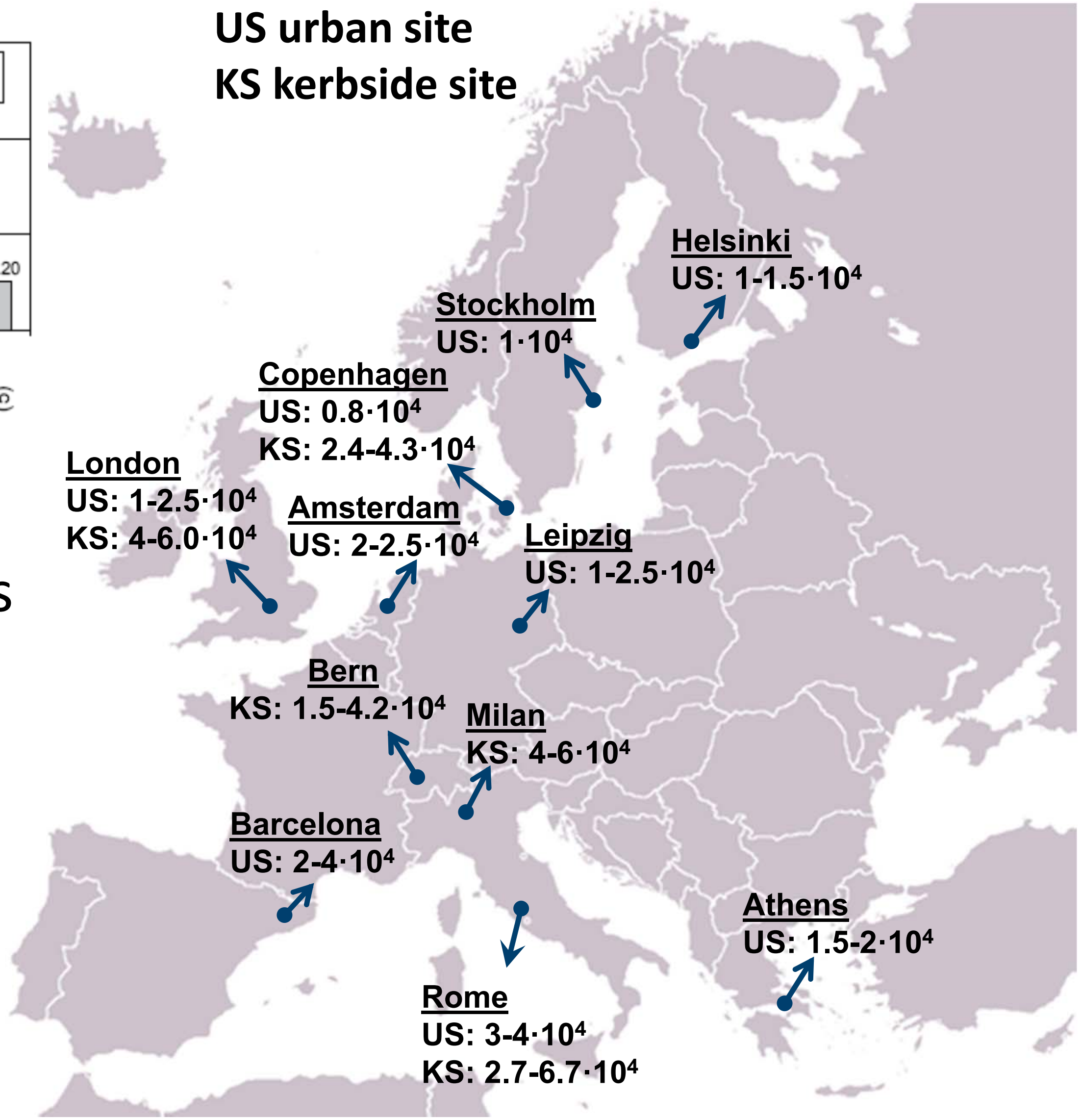
❍ Daily pattern

- Typical 2-peak pattern
- Larger fluctuations on peak hours
- Different pattern on weekends
  - ➢ low concentrations on Sat. afternoon
  - ➢ no morning peak on Sundays

CPC results: Comparison with literature data



Milan data in the same orders reported for kerbside sites in European cities

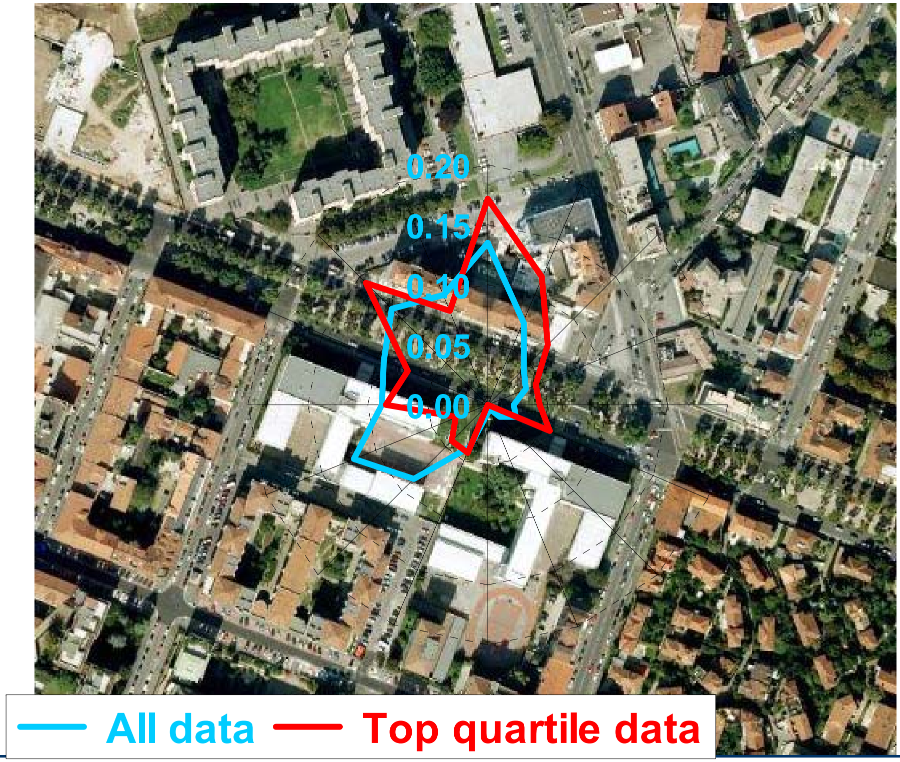


Data from:
Ketzel et al., 2004; Van Dingenen et al. 2004
Aalto et al., 2005; Marconi et al., 2007
Rodriguez et al., 2007 Puustinen, 2007
Reche et al., 2011

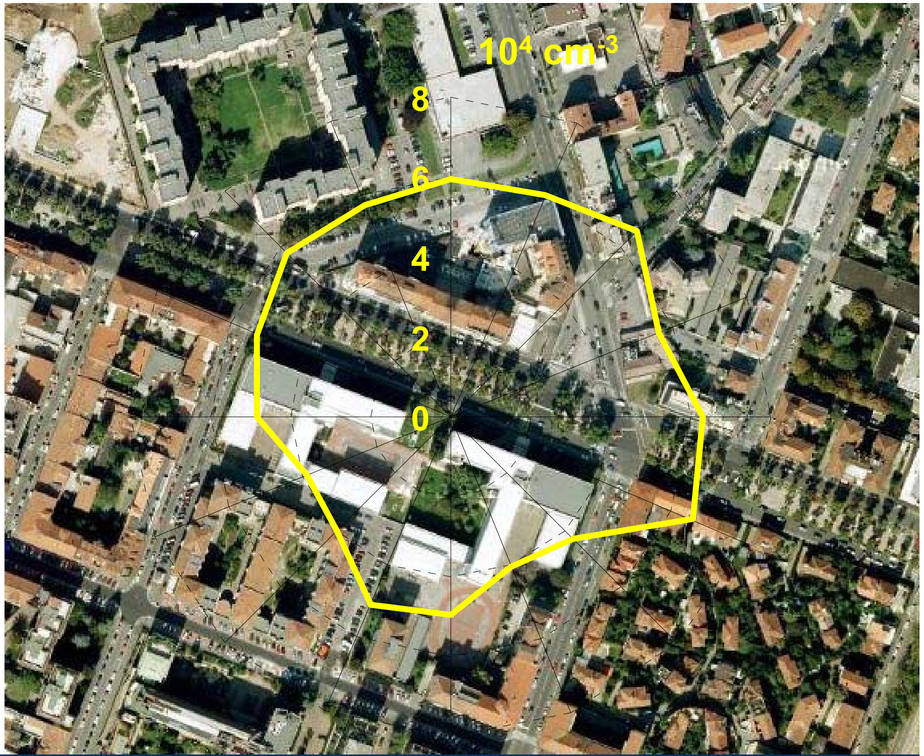
CPC results: Local traffic influence

- Wind-resolved analysis shows the superposition of local emissions on urban background level
- Highest PNC when monitor on the downwind side of the road

Conditional windrose plot



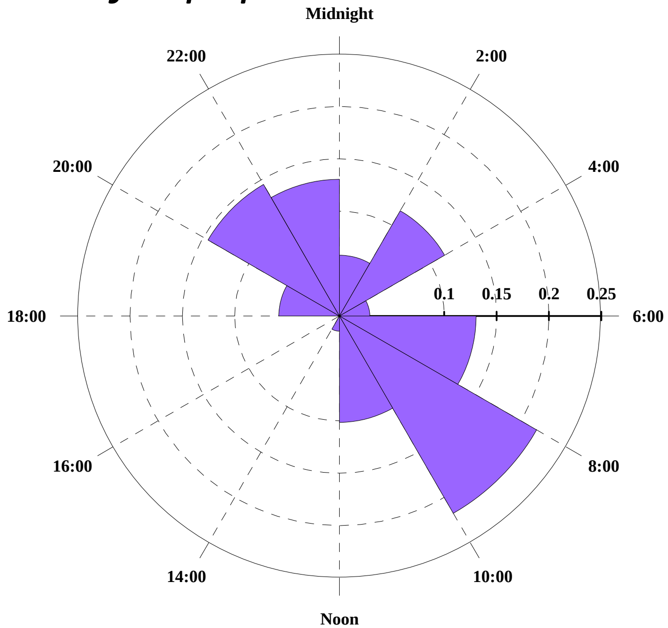
PNC rose plot

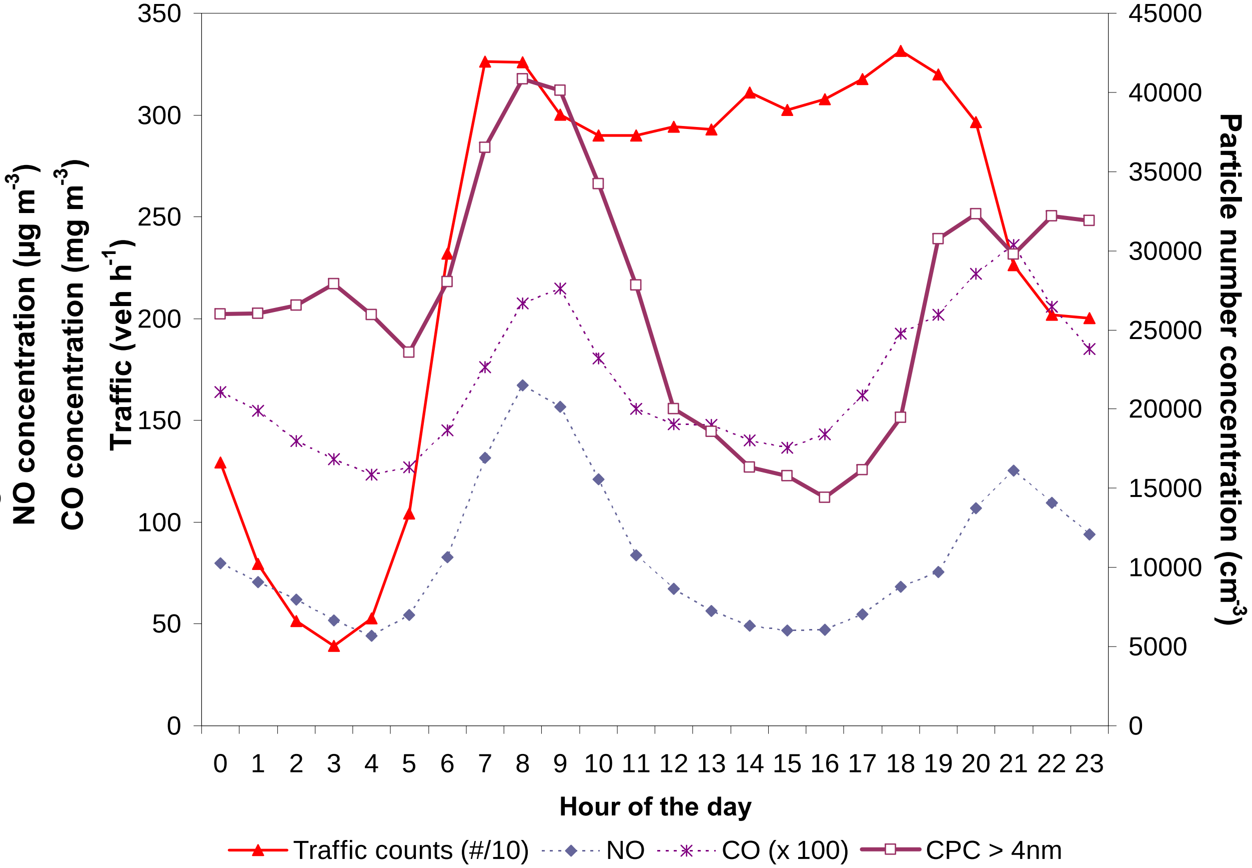


CPC results: relations with air quality and traffic

- Top PNC levels on traffic rush hours
- Concurrent increase of traffic and PNC from the early morning hours until the peak rush hour
- Different pattern in the afternoon ➢ **no overall correlation**

Hour-of-the-day distribution of top quartile PNC data





CPC results: relations with air quality and traffic

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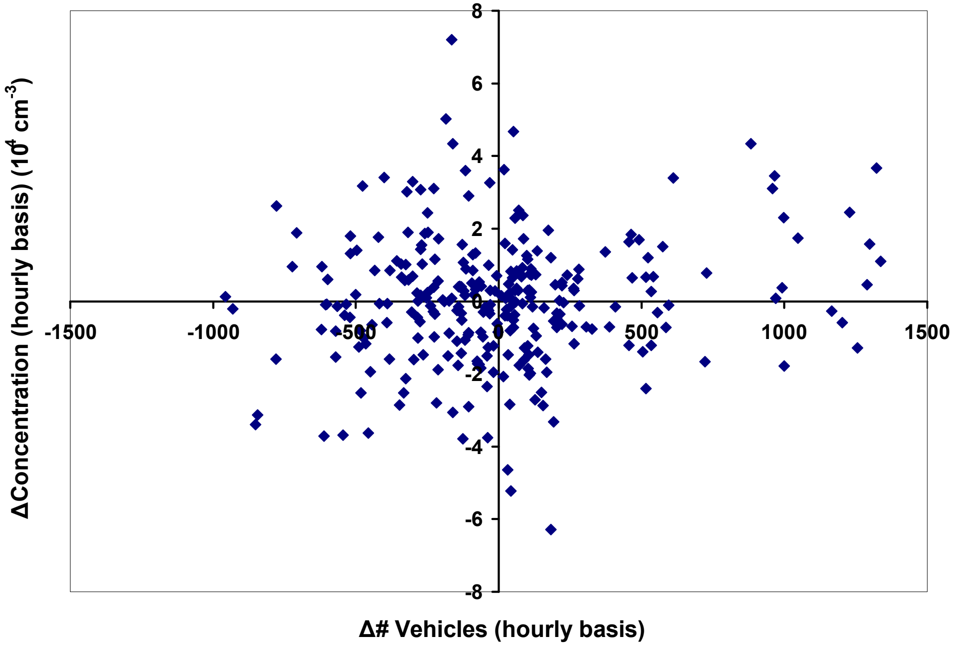
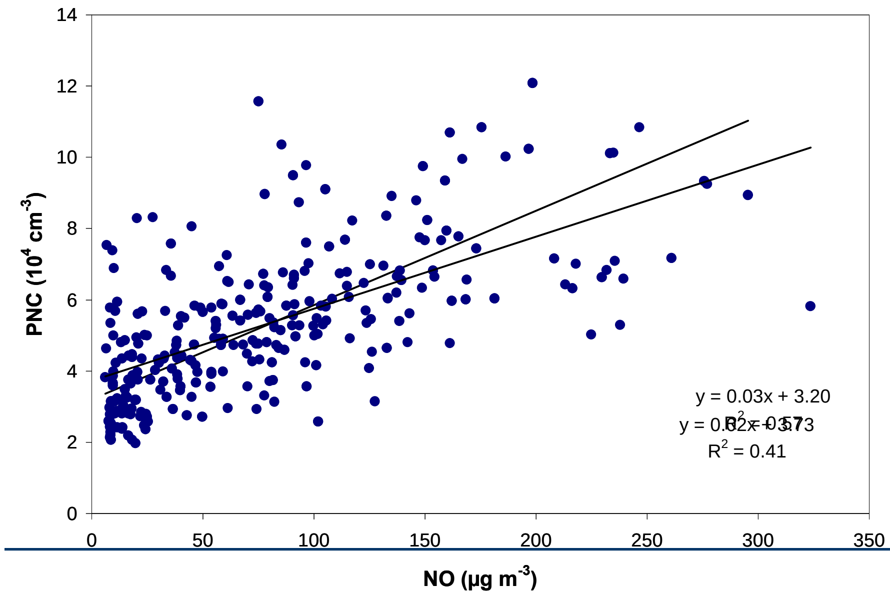
Hour-by-hour PNC change not univocally related to traffic change

- Traffic gradients bad proxy



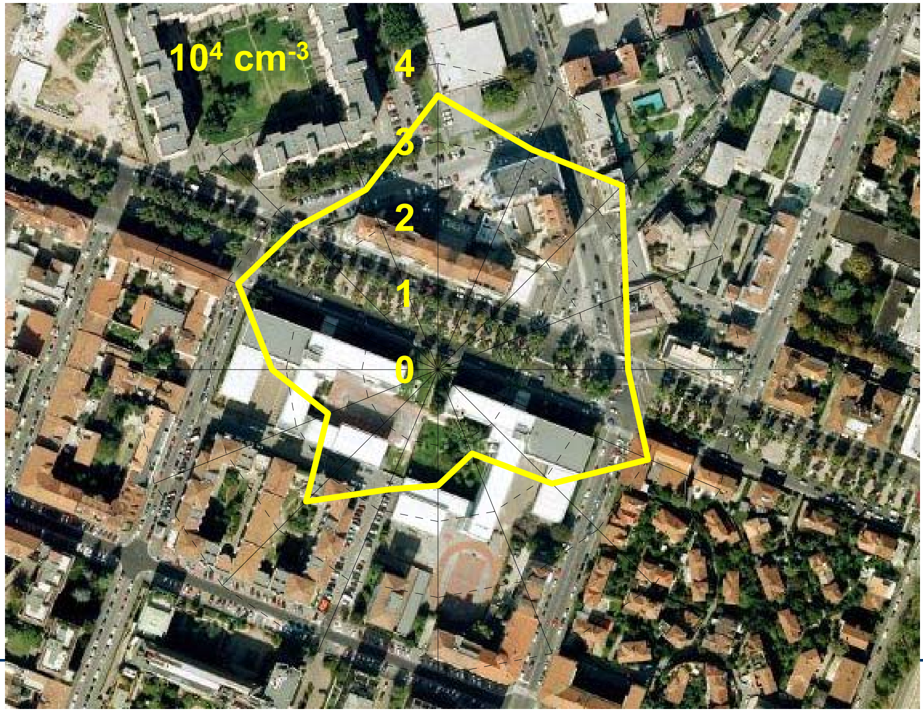


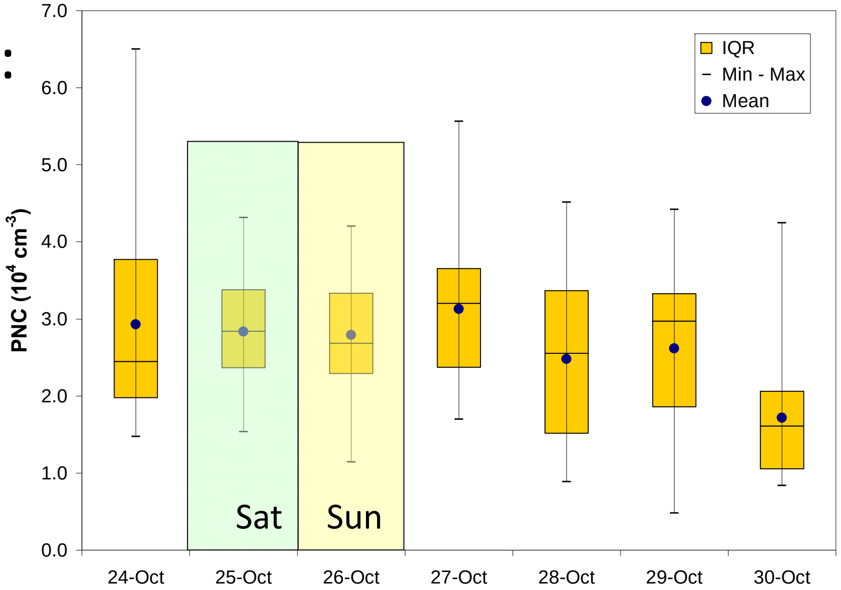
Interesting correlation with primary traffic exhaust gas

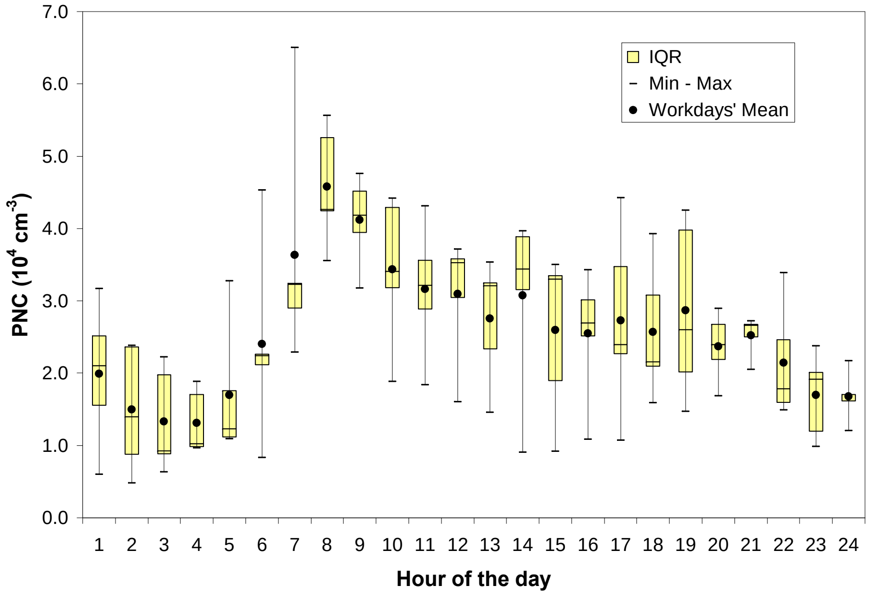
	NO_X	NO	NO_2	CO
All data	0.59	0.64	0.40	0.47
Diurnal data	0.69	0.76	0.34	0.48

ELPI results: patterns & traffic influence

- Same results than CPC campaign, but:
  - lower PNC levels (Daily avg. ≈ $2\text{-}3 \cdot 10^4\ cm^{-3}$)
    - measurement range
    - measurement principle
    - effect of weather conditions
  - no clear evening peak but rather stable levels during afternoon







ELPI results: particle size distribution

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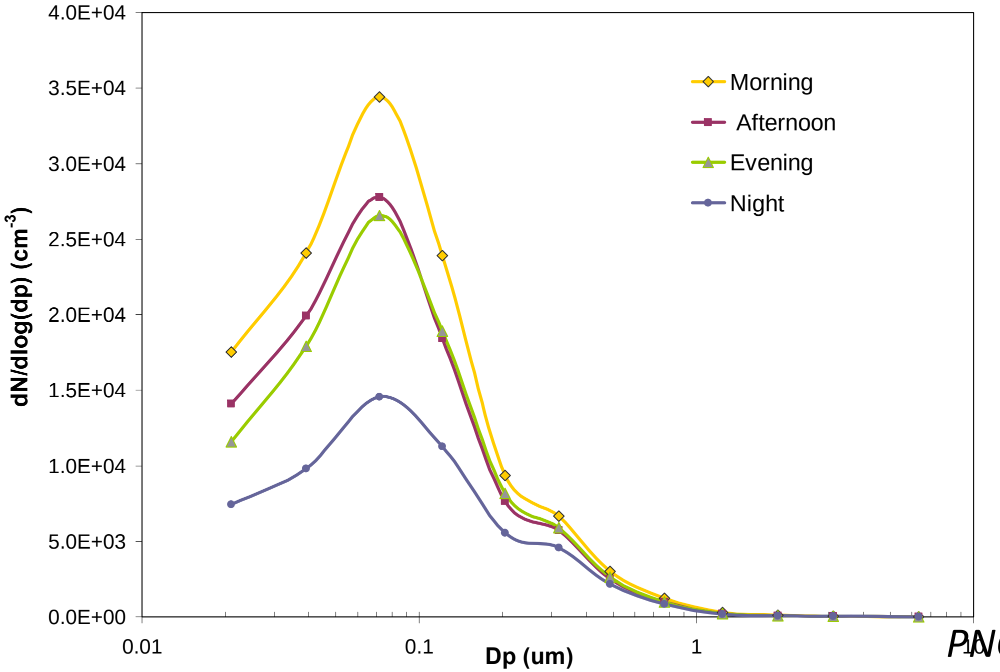
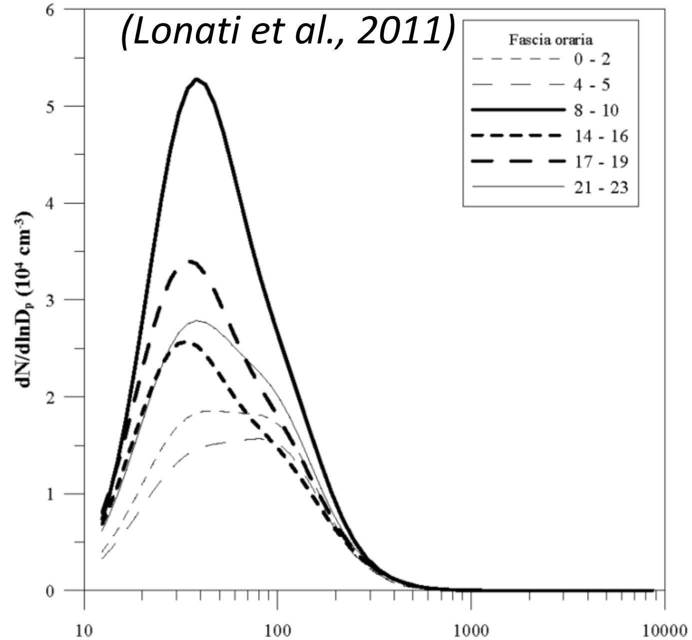
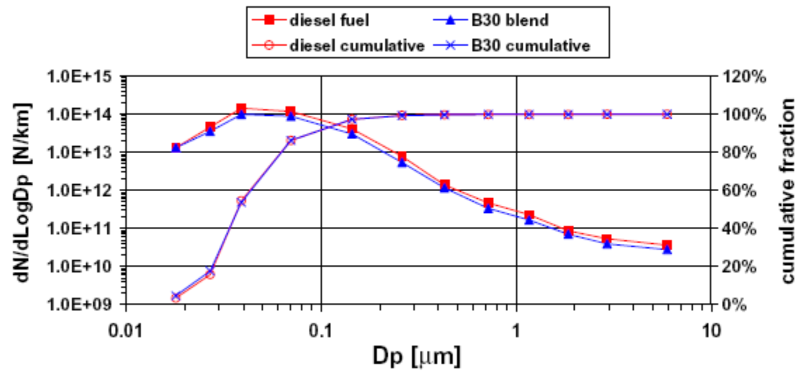
- Size distribution features:
  - bimodal structure: main mode ≈ 70 nm, second mode ≈ 300 nm
  - same shape throughout the day
  - combined effect of the decreasing traffic flow pattern and of the increasing mixing layer height



DMPS PNC size distribution UB site in Milan (Lonati et al., 2011)





PNC size distribution of diesel LDV (Macor et al., 2008)

ELPI results: particle size distribution

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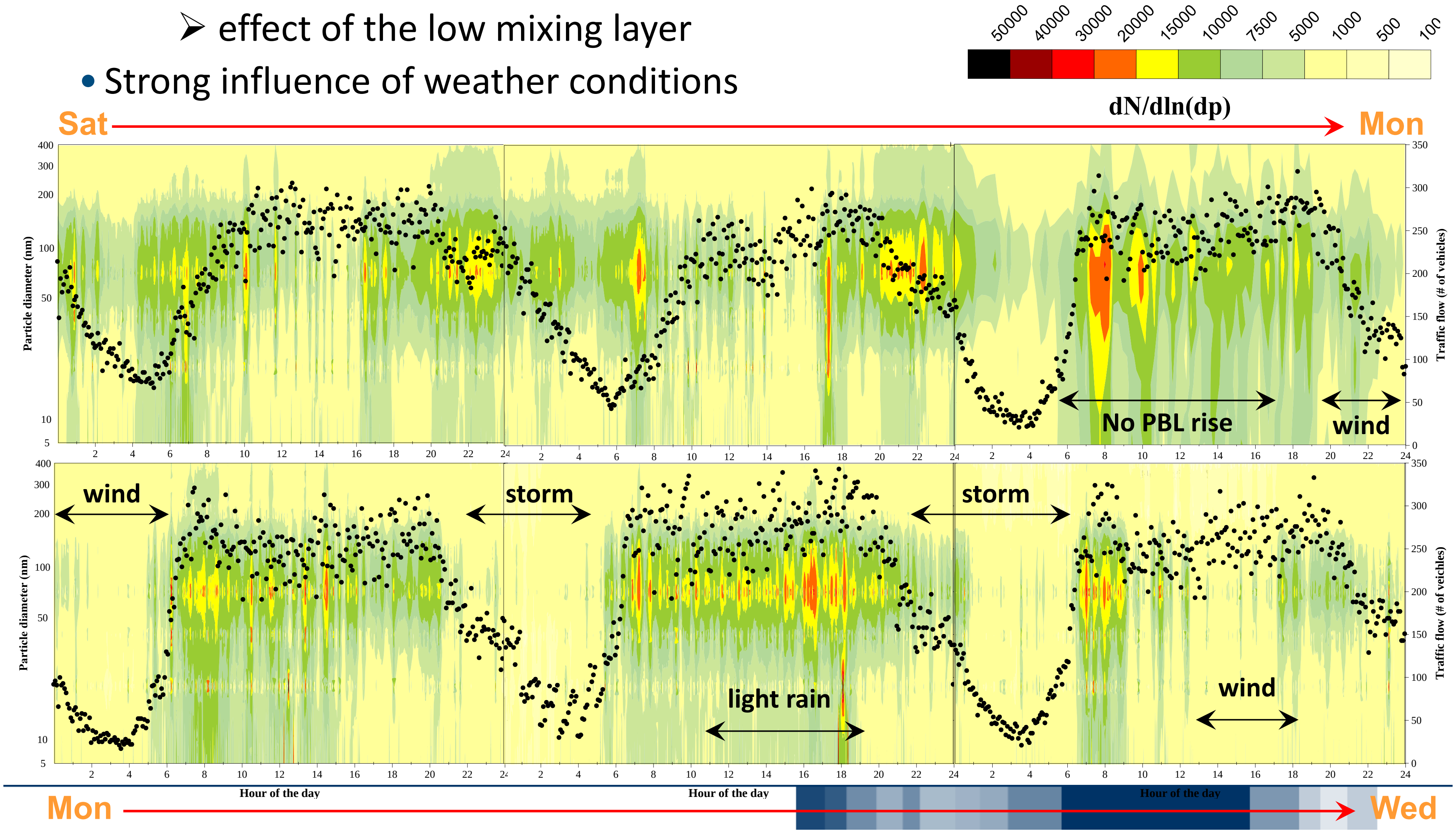
- Size distribution daily pattern at 1-min time resolution:
  - High concentration spots prior/after morning/evening traffic peak
    - ➢ effect of the low mixing layer
  - Strong influence of weather conditions



ELPI results: particle size distribution

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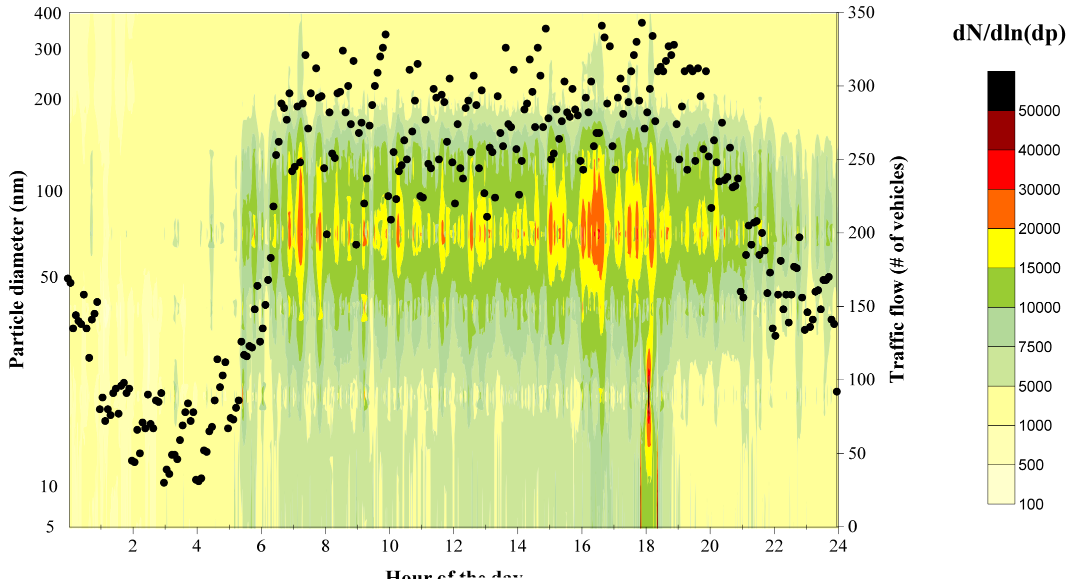
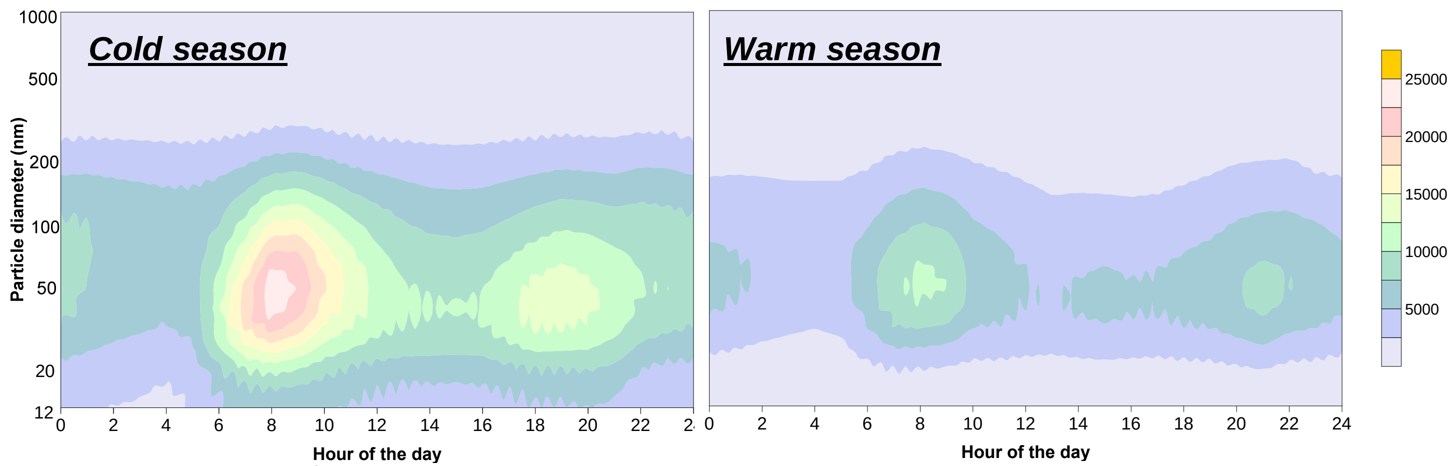
- Size distribution daily pattern at 1-min time resolution:

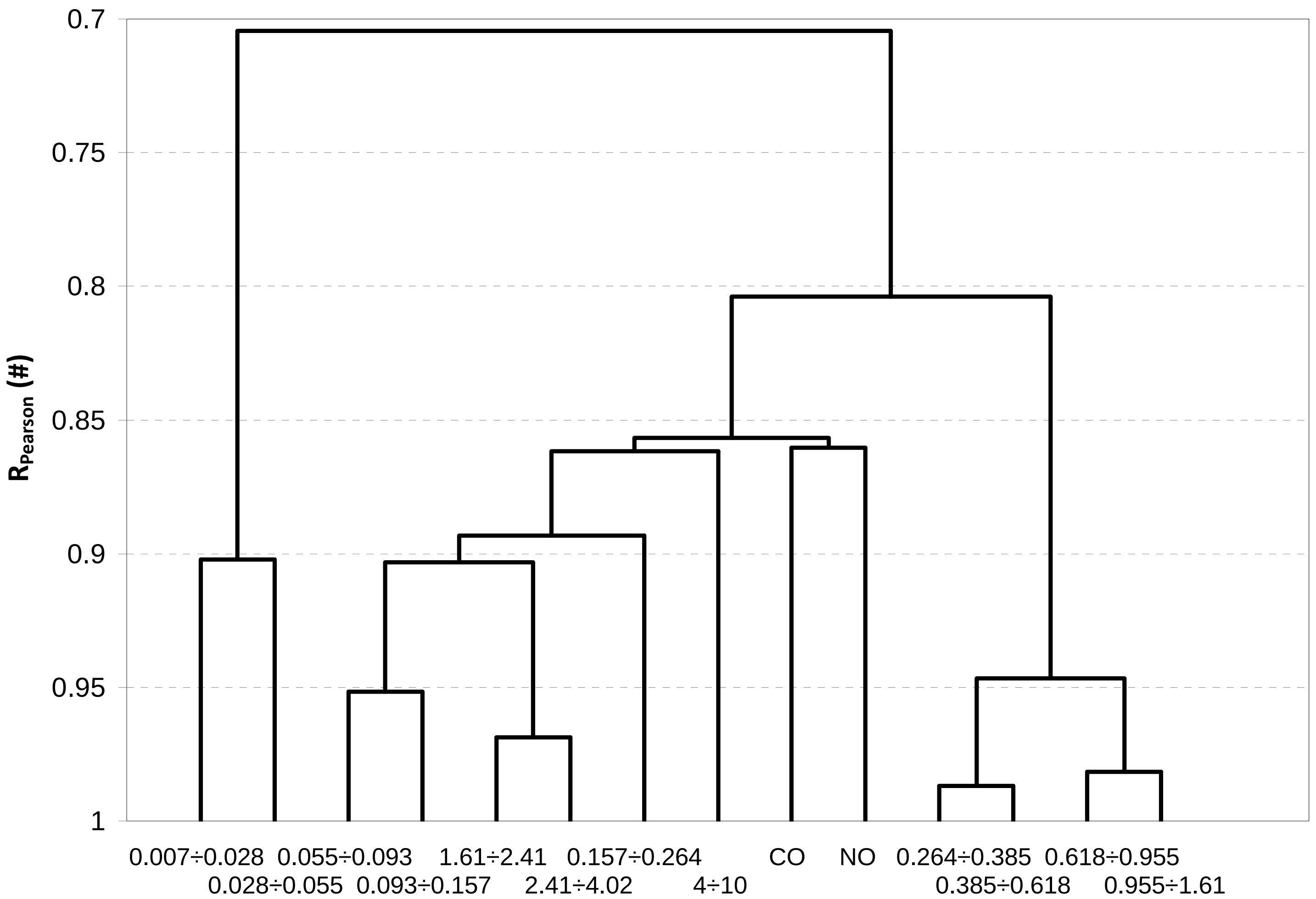




DMPS data from an urban background site in Milan, Crippa 2011

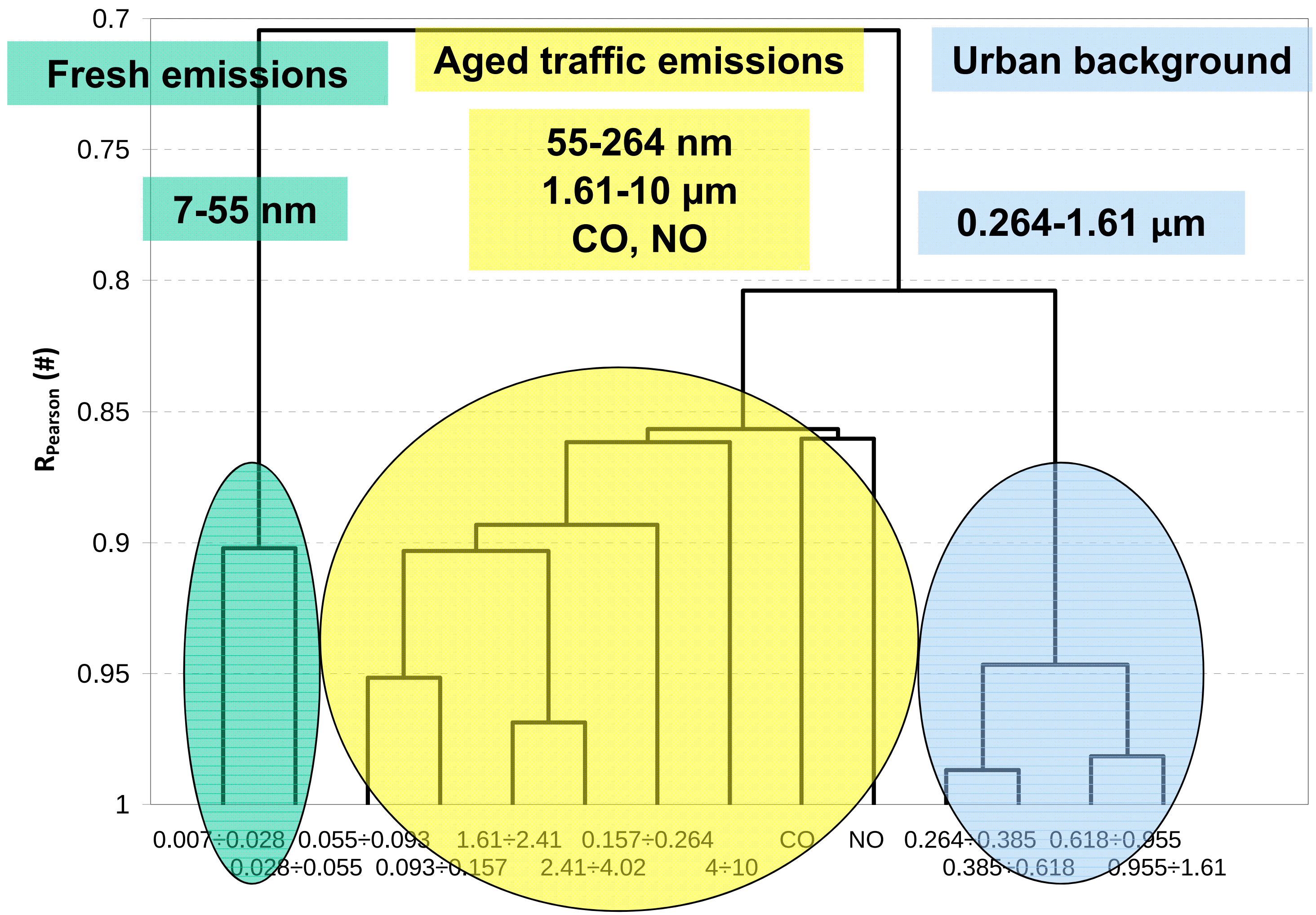
ELPI results: particle size distribution

- Particle size bins and gaseous pollutants clustering



ELPI results: particle size distribution

- Particle size bins and gaseous pollutants clustering



ELPI results: particle size distribution

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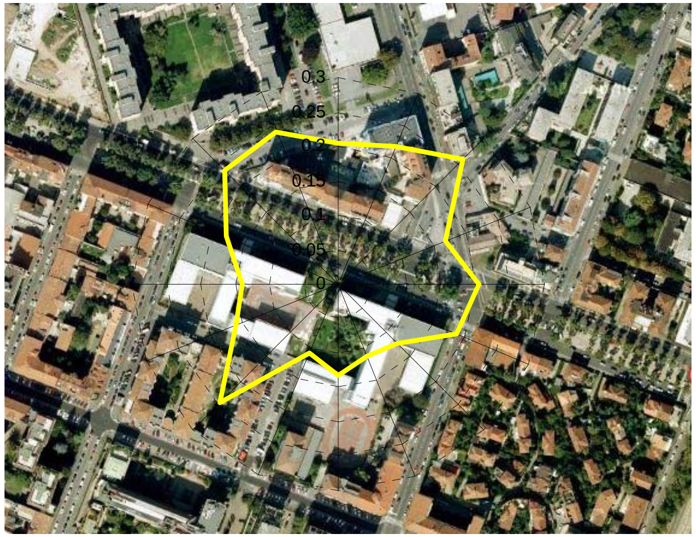
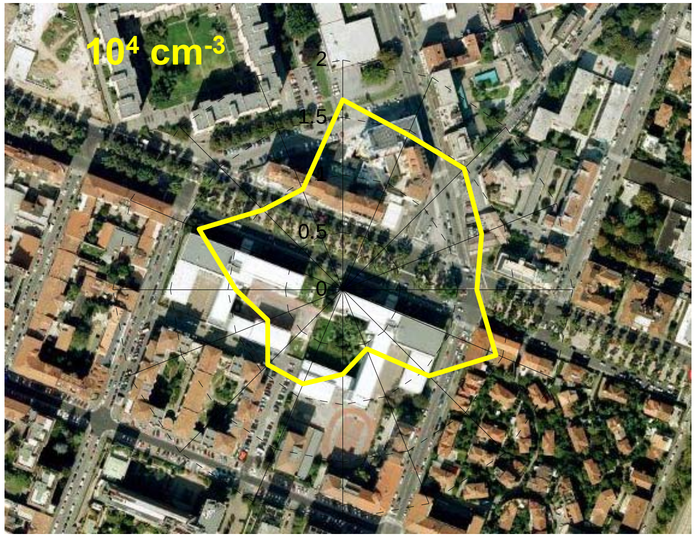
Wind-resolved analysis shows for PN clusters

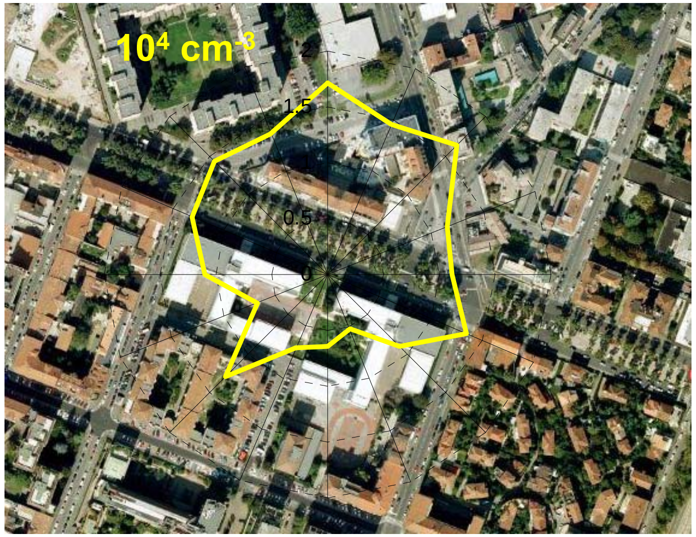


Cluster 3 – Urban background
- ➢ 10-fold lower levels

Cluster 1 – Fresh emissions



Cluster 2 – Aged traffic emitted



Concluding remarks and perspectives

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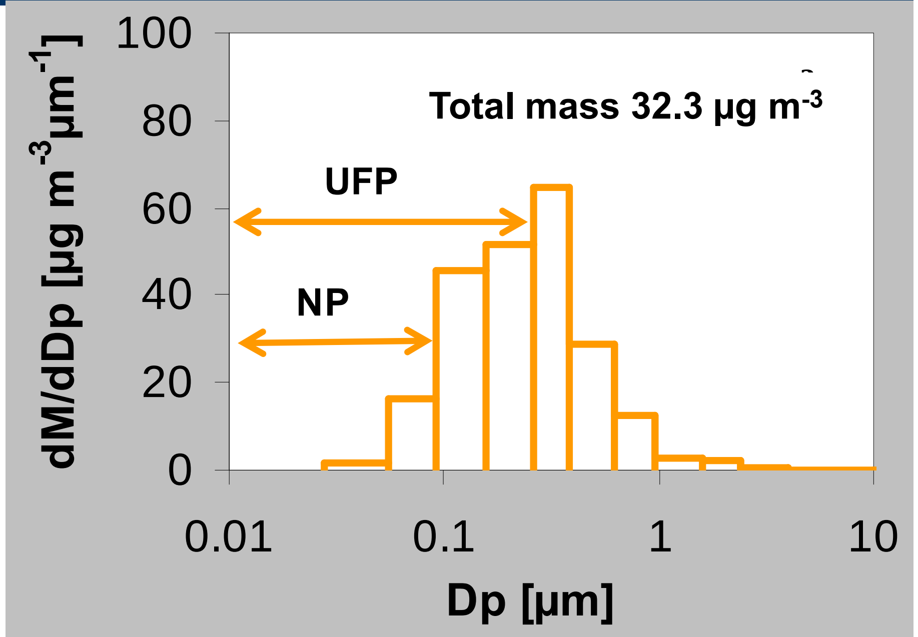
- PM concern in Milan is a question of mass rather than number!
  - Milan PM hot-spot not PN hot-spot!
- Weather conditions important even for kerbside measurements
- PNC daily evolution similar to those of traffic related pollutants
- Cluster analysis to group size bins with similar behaviour and origin

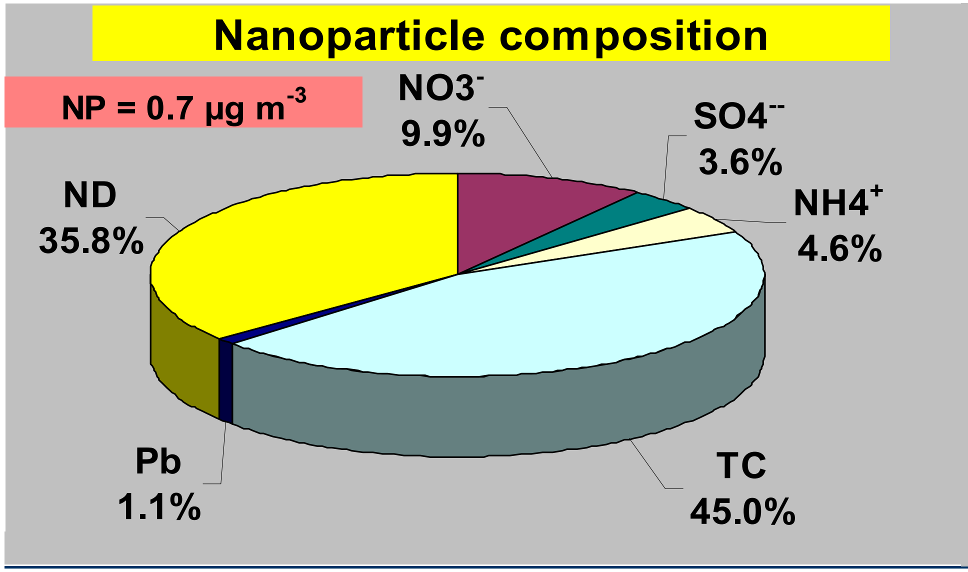
- Combined measurements with both instruments (+ other ??)
- Concentration data modelling:
  - Multiple regression model
  - Emission-based model (traffic data)
- ELPI+ vs. ELPI (additional size bin split at 17 nm)
- *"substances have to come into the game for toxicity assessment"*

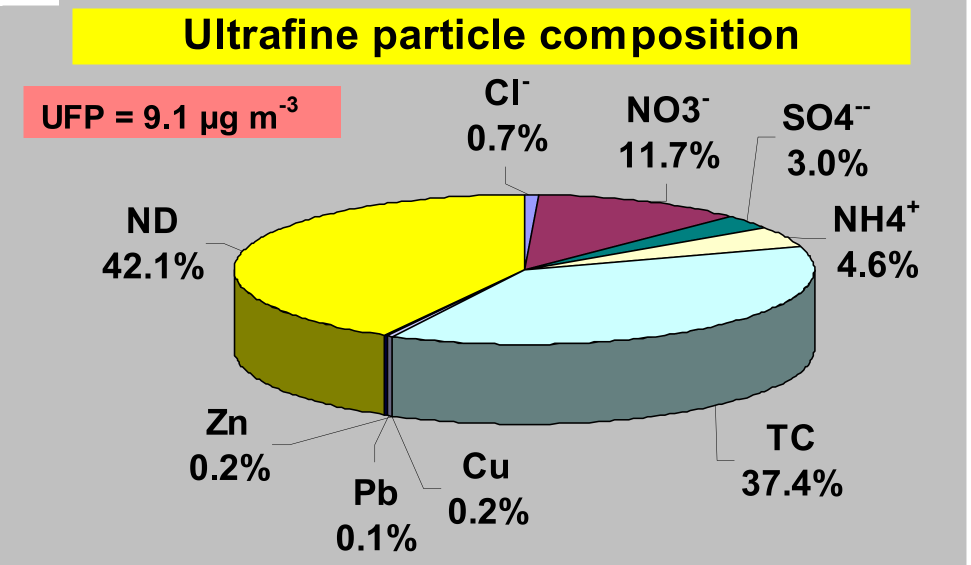
A. Mayer, two days ago

NP & UFP chemical speciation

- UFP about 28% of the mass
- NP only 2% of the mass
- Relevant Not Determined share
- TC first contributor to the mass
- Secondary ionic species about 20% of NP and UFP mass







Thank you for your attention

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