SUPPORTING INFORMATION

On the spatial distribution and evolution of ultrafine particles in Barcelona

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Fig. S1. Monitoring sites in Barcelona, Spain

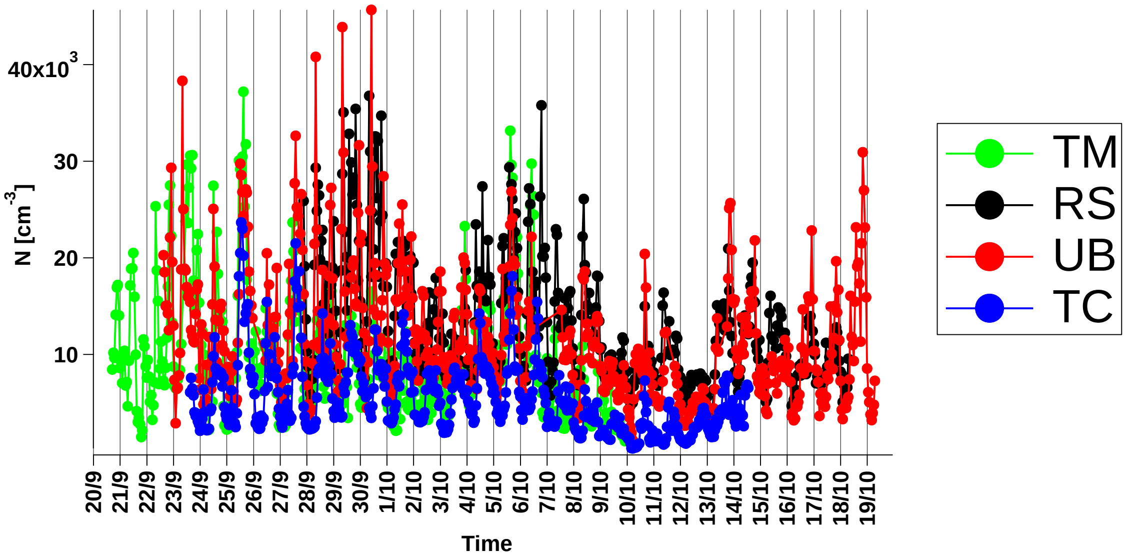


Fig. S2. Temporal trends of the particle number concentrations measured during the SAPUSS project at four different monitoring sites.

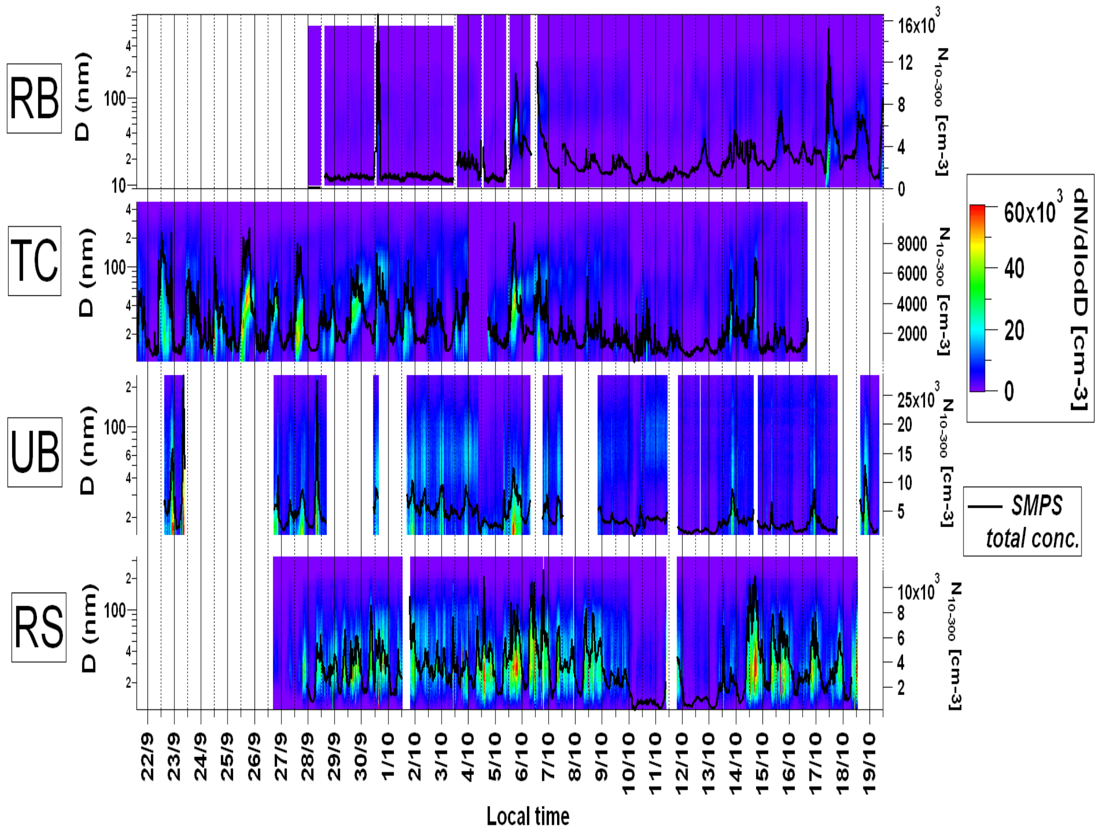
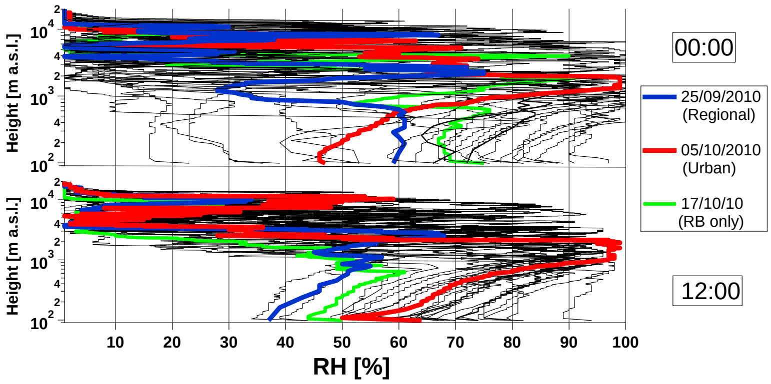
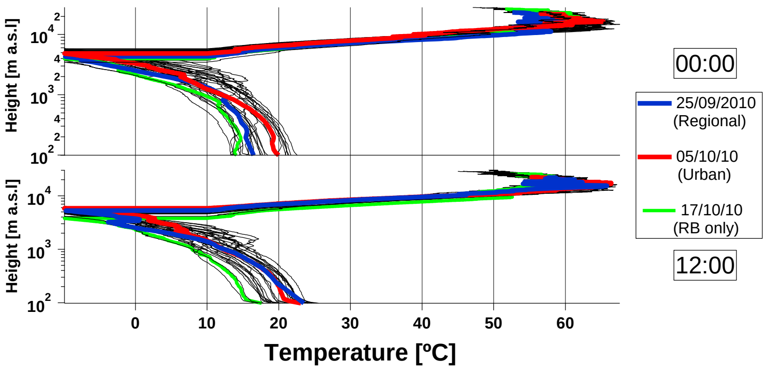


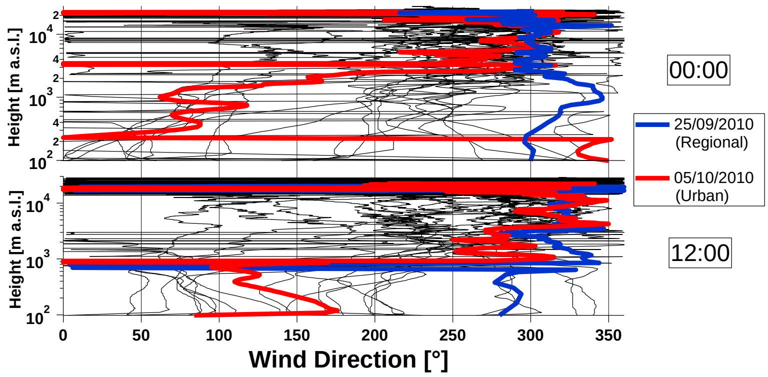
Fig. S3. Temporal trends of the aerosol size distributions during the SAPUSS project at four different monitoring sites.


00:00
12:00
25/09/2010 (Regional)
05/10/2010 (Urban)
17/10/10 (RB only)
Height [m a.s.l.]
RH [%]

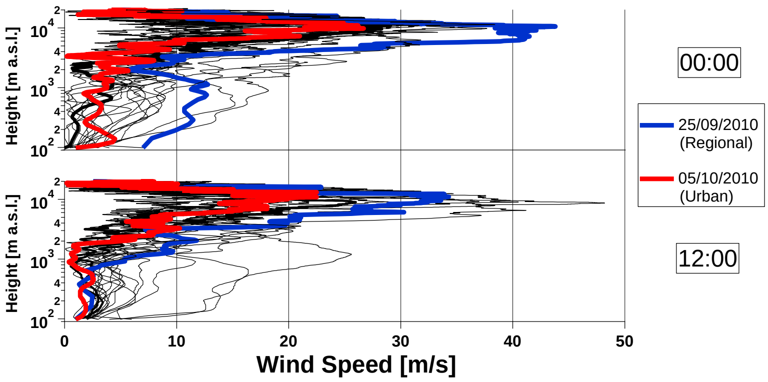
1


00:00
12:00
25/09/2010 (Regional)
05/10/10 (Urban)
17/10/10 (RB only)
Height [m a.s.l]
Temperature [ºC]

2


00:00
12:00
25/09/2010 (Regional)
05/10/2010 (Urban)
Height [m a.s.l.]
Wind Direction [°]

3


00:00
12:00
25/09/2010 (Regional)
05/10/2010 (Urban)
Height [m a.s.l.]
Wind Speed [m/s]

4

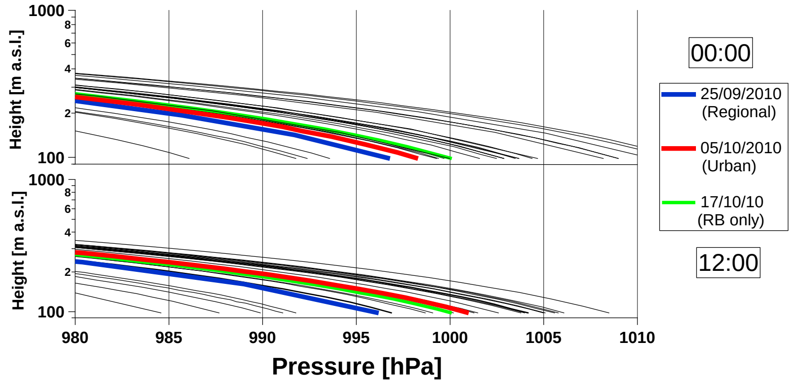
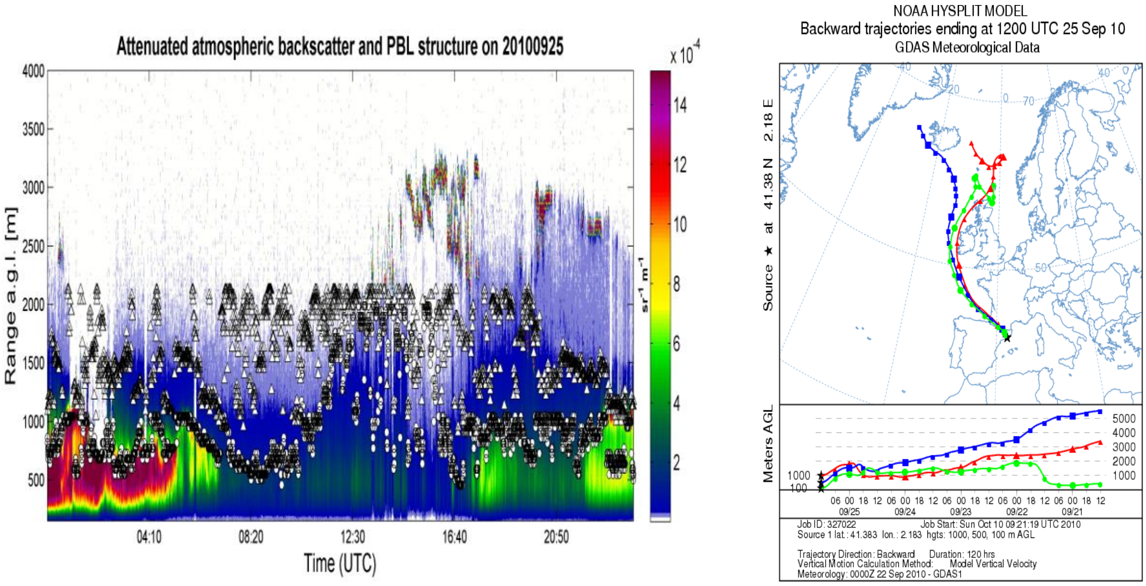
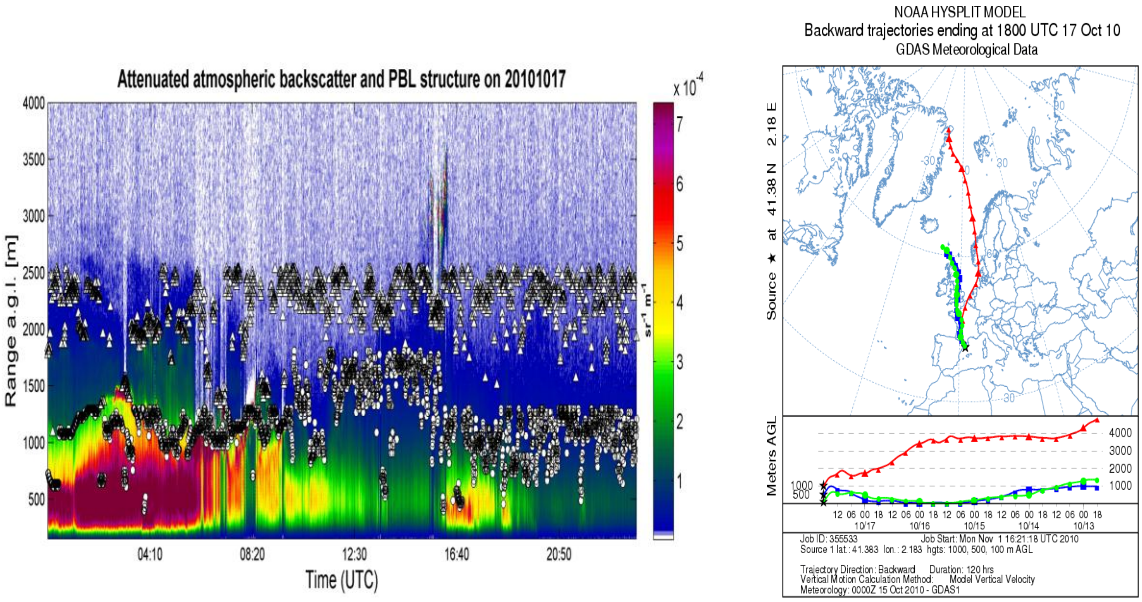


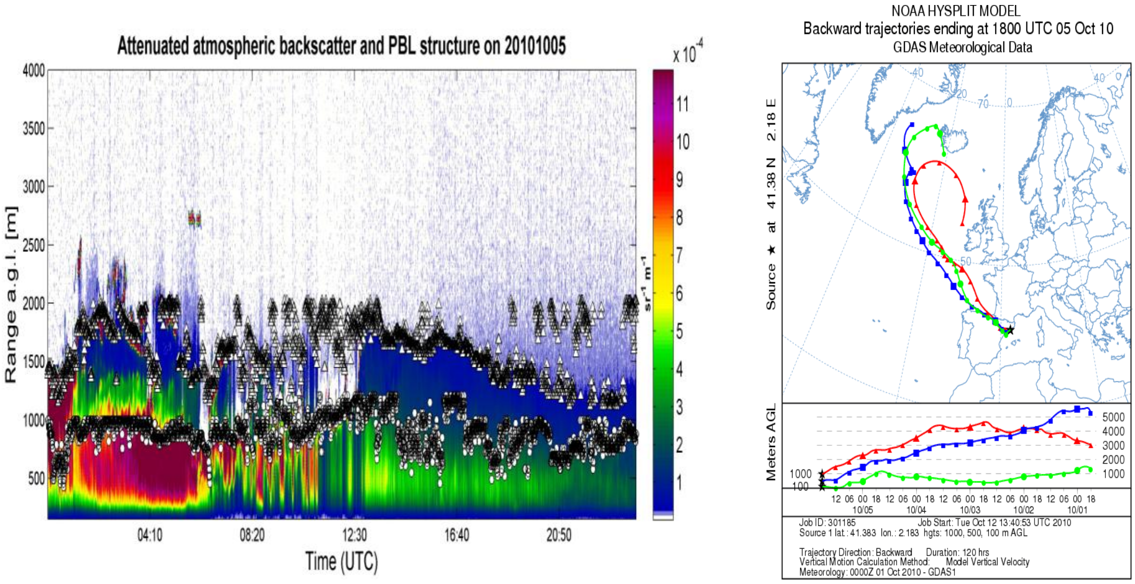
Fig. S4. Vertical profiles of RH, temperature, wind direction, wind speed and air pressure obtained with radio sounding balloons performed twice per day (at 12:00 UTC and 00:00 UTC) at the UB site throughout the SAPUSS campaign (64 balloons in total). Enhanced in colour are the profiles obtained during the regional (blue), urban (red) and regional background only (green) nucleation events. Wind data for day 17/10/10 were not available.


Attenuated atmospheric backscatter and PBL structure on 20100925
Range a.g.l. [m]
Time (UTC)
x 10^-4
sr^-1 m^-1
NOAA HYSPLIT MODEL
Backward trajectories ending at 1200 UTC 25 Sep 10
GDAS Meteorological Data
Source ★ at 41.38 N 2.18 E
Meters AGL
Job ID: 327022 Job Start: Sun Oct 10 09:21:19 UTC 2010
Source 1 lat.: 41.383 lon.: 2.183 hgts: 1000, 500, 100 m AGL
Trajectory Direction: Backward Duration: 120 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 22 Sep 2010 - GDAS1

(a)


Attenuated atmospheric backscatter and PBL structure on 20101017
Range a.g.l. [m]
Time (UTC)
x 10^-4
sr^-1 m^-1
NOAA HYSPLIT MODEL
Backward trajectories ending at 1800 UTC 17 Oct 10
GDAS Meteorological Data
Source ★ at 41.38 N 2.18 E
Meters AGL
Job ID: 355533 Job Start: Mon Nov 1 16:21:18 UTC 2010
Source 1 lat.: 41.383 lon.: 2.183 hgts: 1000, 500, 100 m AGL
Trajectory Direction: Backward Duration: 120 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 15 Oct 2010 - GDAS1

(b)



(c)

Fig. S5. Diurnal profile of the boundary layer and air mass back trajectories during the (a) regional nucleation event on 25th September 2010, (b) regional background nucleation on 17th October 2010 and (c) the urban nucleation event on 5th October 2010.

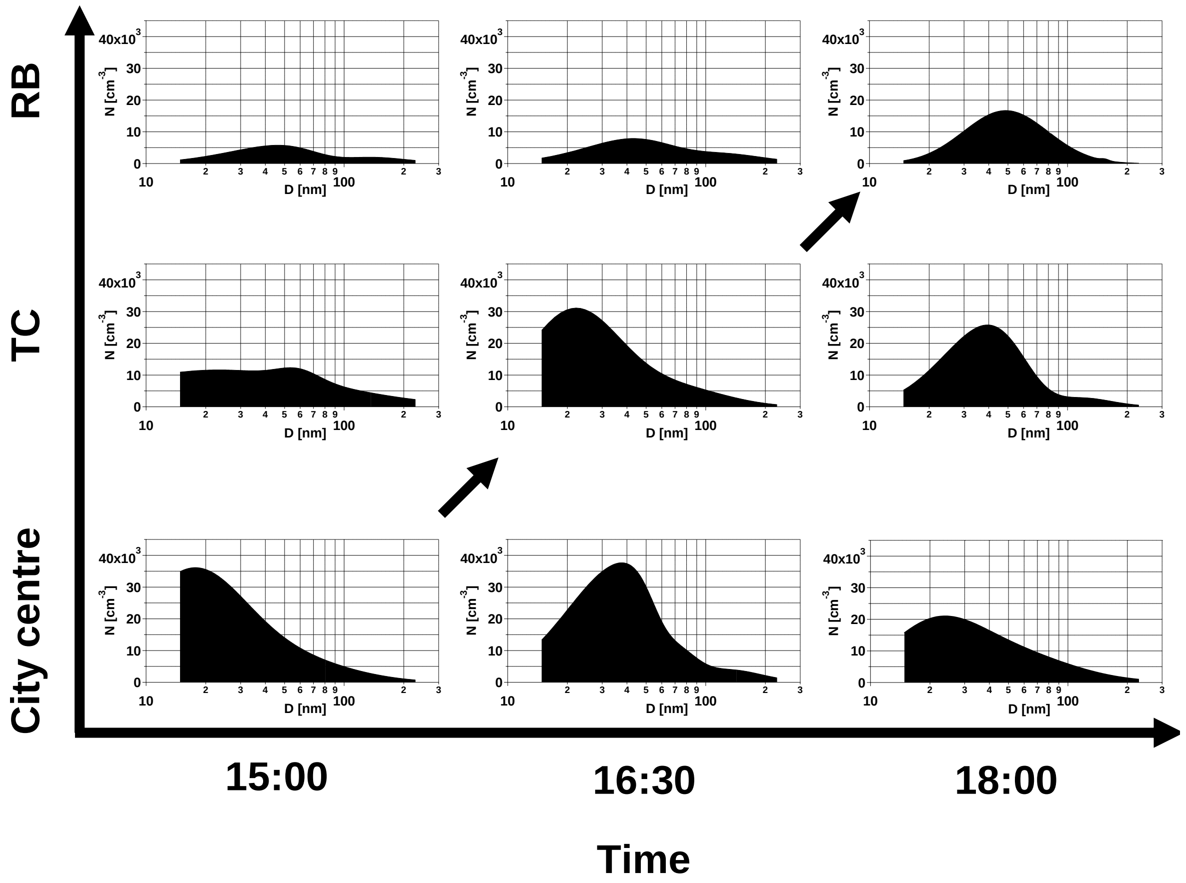
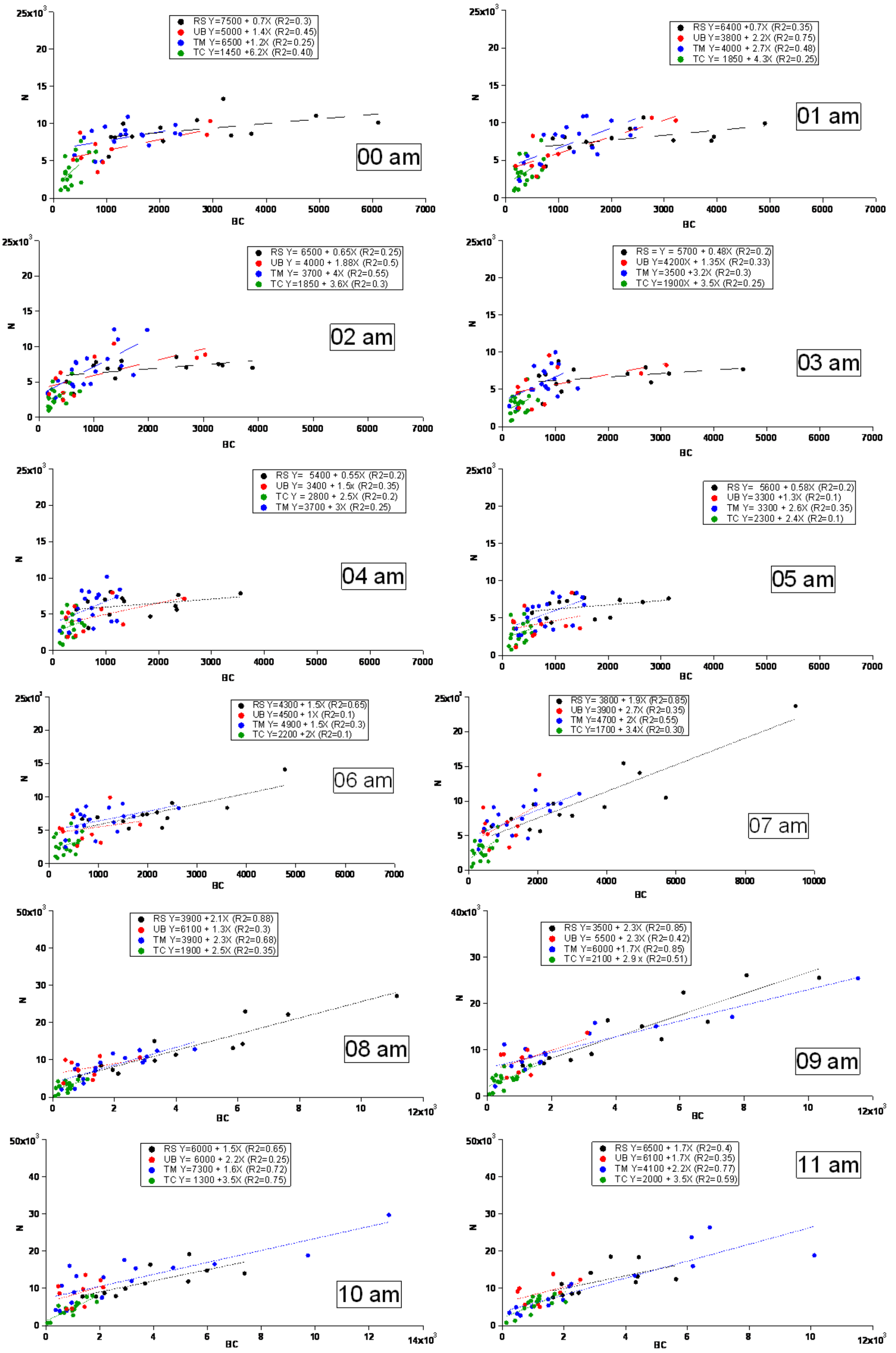


Fig. S6. Aerosol size distributions shown during the urban nucleation event (05/10/2010), starting at about 15:00 in the city (shown for simplicity as an average between RS and UB), moving to the urban background TC and reaching the regional background site (RB) at about 18:00 as a plume of ultrafine particles grown to about 50 nm (SMPS size distributions shown also in Fig. 6).



RS Y=7500 + 0.7X (R2=0.3)
UB Y=5000 + 1.4X (R2=0.45)
TM Y=6500 +1.2X (R2=0.25)
TC Y=1450 +6.2X (R2=0.40)
00 am
RS Y=6400 +0.7X (R2=0.35)
UB Y=3800 + 2.2X (R2=0.75)
TM Y=4000 + 2.7X (R2=0.48)
TC Y= 1850 + 4.3X (R2=0.25)
01 am
RS Y= 6500 + 0.65X (R2=0.25)
UB Y = 4000 + 1.88X (R2=0.5)
TM Y= 3700 + 4X (R2=0.55)
TC Y=1850 + 3.6X (R2=0.3)
02 am
RS = Y = 5700 + 0.48X (R2=0.2)
UB Y=4200X + 1.35X (R2=0.33)
TM Y=3500 +3.2X (R2=0.3)
TC Y=1900X + 3.5X (R2=0.25)
03 am
RS Y= 5400 + 0.55X (R2=0.2)
UB Y= 3400 + 1.5x (R2=0.35)
TC Y = 2800 + 2.5X (R2=0.2)
TM Y=3700 + 3X (R2=0.25)
04 am
RS Y= 5600 + 0.58X (R2=0.2)
UB Y=3300 +1.3X (R2=0.1)
TM Y= 3300 + 2.6X (R2=0.35)
TC Y=2300 + 2.4X (R2=0.1)
05 am
RS Y=4300 + 1.5X (R2=0.65)
UB Y=4500 + 1X (R2=0.1)
TM Y= 4900 + 1.5X (R2=0.3)
TC Y=2200 +2X (R2=0.1)
06 am
RS Y= 3800 + 1.9X (R2=0.85)
UB Y=3900 + 2.7X (R2=0.35)
TM Y=4700 + 2X (R2=0.55)
TC Y=1700 + 3.4X (R2=0.30)
07 am
RS Y=3900 +2.1X (R2=0.88)
UB Y=6100 + 1.3X (R2=0.3)
TM Y=3900 + 2.3X (R2=0.68)
TC Y=1900 + 2.5X (R2=0.35)
08 am
RS Y=3500 + 2.3X (R2=0.85)
UB Y= 5500 + 2.3X (R2=0.42)
TM Y=6000 +1.7X (R2=0.85)
TC Y=2100 + 2.9 x (R2=0.51)
09 am
RS Y=6000 + 1.5X (R2=0.65)
UB Y= 6000 + 2.2X (R2=0.25)
TM Y=7300 + 1.6X (R2=0.72)
TC Y= 1300 +3.5X (R2=0.75)
10 am
RS Y=6500 + 1.7X (R2=0.4)
UB Y=6100 +1.7X (R2=0.35)
TM Y=4100 +2.2X (R2=0.77)
TC Y=2000 + 3.5X (R2=0.59)
11 am
N
BC

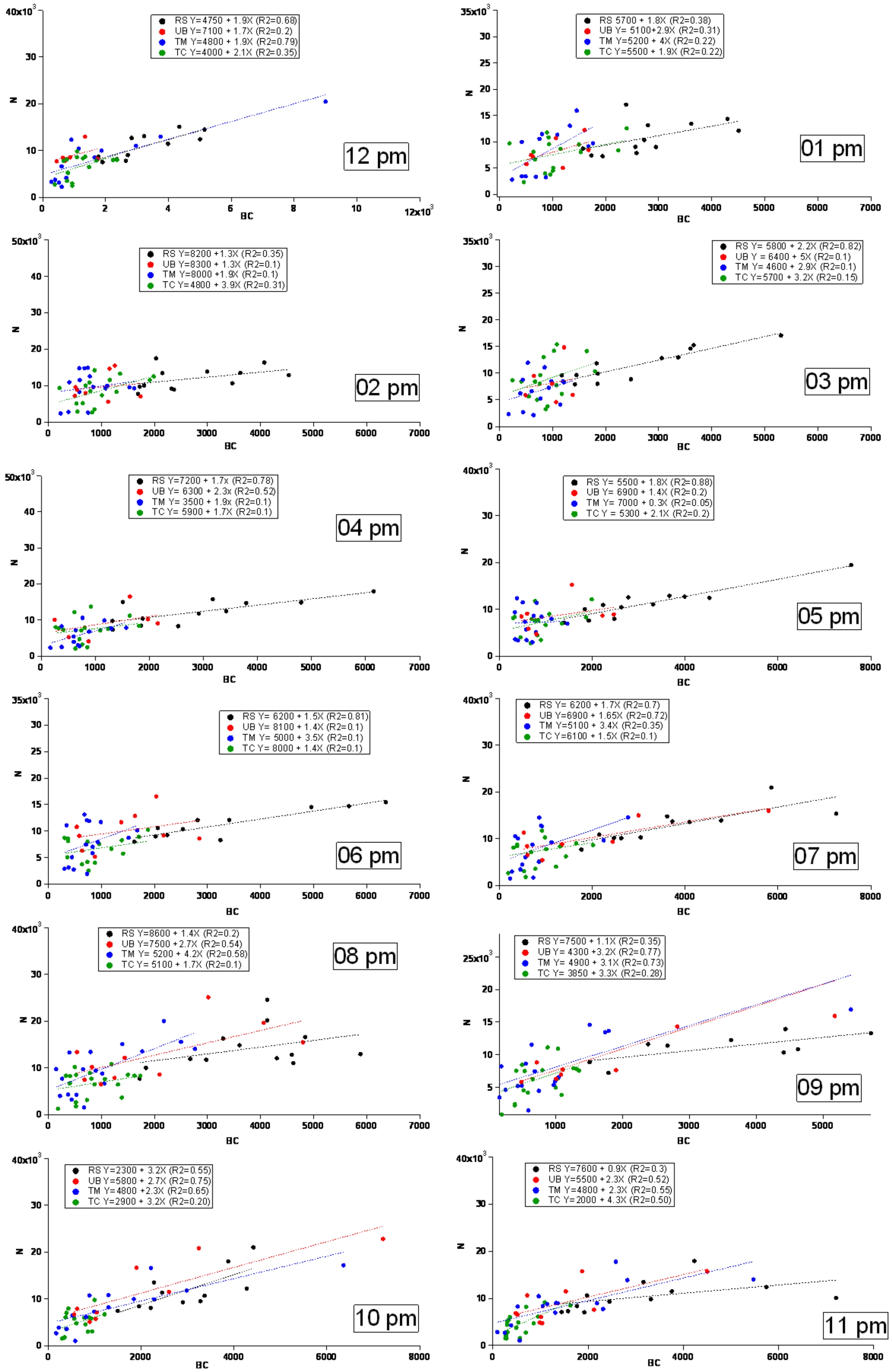

RS Y=4750 + 1.9X (R2=0.68)
UB Y=7100 + 1.7X (R2=0.2)
TM Y=4800 + 1.9X (R2=0.79)
TC Y=4000 + 2.1X (R2=0.35)
12 pm
N
BC
RS 5700 + 1.8X (R2=0.38)
UB Y= 5100+2.9X (R2=0.31)
TM Y=5200 + 4X (R2=0.22)
TC Y=5500 + 1.9X (R2=0.22)
01 pm
RS Y=8200 +1.3X (R2=0.35)
UB Y=8300 + 1.3X (R2=0.1)
TM Y=8000 +1.9X (R2=0.1)
TC Y=4800 + 3.9X (R2=0.31)
02 pm
RS Y= 5800 + 2.2X (R2=0.82)
UB Y = 6400 + 5X (R2=0.1)
TM Y = 4600 + 2.9X (R2=0.1)
TC Y=5700 + 3.2X (R2=0.15)
03 pm
RS Y=7200 + 1.7x (R2=0.78)
UB Y= 6300 + 2.3x (R2=0.52)
TM Y= 3500 + 1.9x (R2=0.1)
TC Y= 5900 + 1.7X (R2=0.1)
04 pm
RS Y= 5500 + 1.8X (R2=0.88)
UB Y= 6900 + 1.4X (R2=0.2)
TM Y= 7000 + 0.3X (R2=0.05)
TC Y = 5300 + 2.1X (R2=0.2)
05 pm
RS Y= 6200 + 1.5X (R2=0.81)
UB Y= 8100 + 1.4X (R2=0.1)
TM Y= 5000 + 3.5X (R2=0.1)
TC Y=8000 + 1.4X (R2=0.1)
06 pm
RS Y= 6200 + 1.7X (R2=0.7)
UB Y=6900 + 1.65X (R2=0.72)
TM Y=5100 + 3.4X (R2=0.35)
TC Y=6100 + 1.5X (R2=0.1)
07 pm
RS Y=8600 + 1.4X (R2=0.2)
UB Y=7500 +2.7X (R2=0.54)
TM Y= 5200 + 4.2X (R2=0.58)
TC Y= 5100 + 1.7X (R2=0.1)
08 pm
RS Y=7500 + 1.1X (R2=0.35)
UB Y= 4300 +3.2X (R2=0.77)
TM Y= 4900 + 3.1X (R2=0.73)
TC Y= 3850 + 3.3X (R2=0.28)
09 pm
RS Y=2300 + 3.2X (R2=0.55)
UB Y=5800 + 2.7X (R2=0.75)
TM Y=4800 +2.3X (R2=0.65)
TC Y=2900 + 3.2X (R2=0.20)
10 pm
RS Y=7600 + 0.9X (R2=0.3)
UB Y=5500 +2.3X (R2=0.52)
TM Y=4800 + 2.3X (R2=0.55)
TC Y=2000 + 4.3X (R2=0.50)
11 pm

Fig. S7. Linear correlation between BC (ng m^{-3}) and N (cm^{-3}) at four different monitoring sites for each hour of the day.

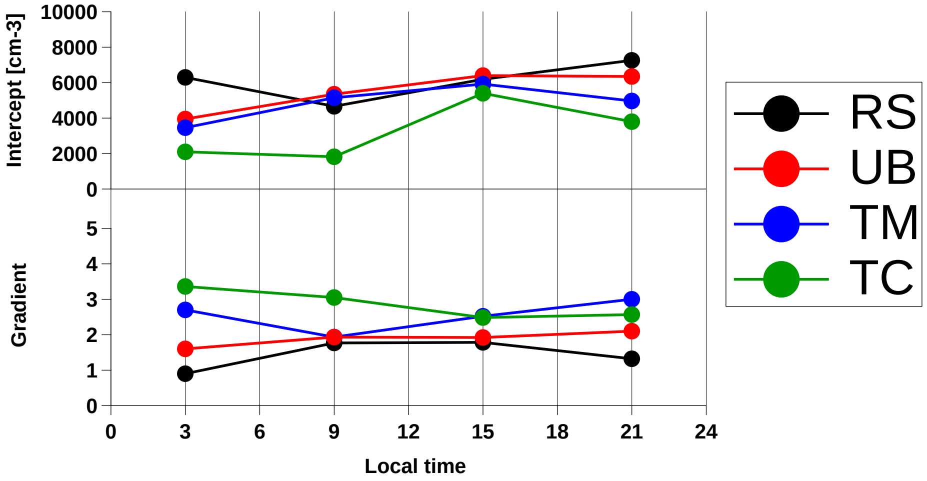


Fig. S8. Gradient and intercept binned at six hour intervals obtained from the linear correlations of Fig. S6.

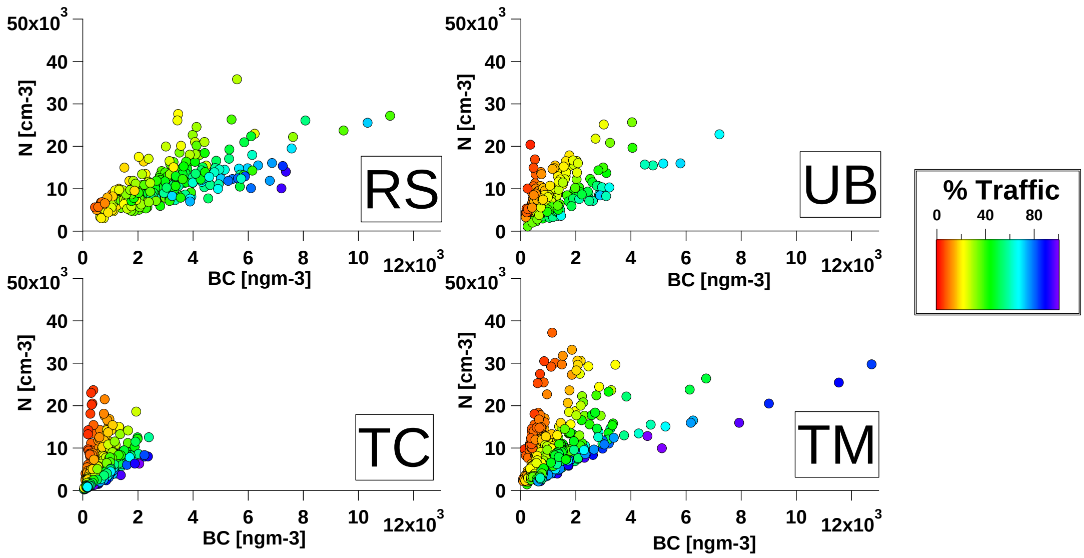


Fig S9. Percentage of primary non volatile traffic particles (% Traffic) at the four different monitoring sites.

Calculation of the Condensation Sink

The condensation sink (CS) describes how rapidly condensable vapour molecules will condense on the existing aerosol. Specifically this quantity describes the loss rate of molecules with diameter d_p, diffusion coefficient D, and mean free path λ_v onto a distribution $n(d_p)$ (or N_i in the discrete case) of existing particles and as such, can be obtained from integrating over the particle size spectrum (Dal Maso *et al.* 2002).

$$\begin{aligned} CS &= 2\pi D\int_0^\infty d_p \beta_M(d_p) n(d_p) dd_p \\ &= 2\pi D\sum_i^o \beta_{M_i} d_{p,i} N_i \end{aligned} \qquad (1)$$

The transitional correction factor β_M can be expressed as (Fuchs *et al.* 1971)

$$\beta_M = \frac{Kn+1}{0.377Kn+1+\frac{4}{3}\alpha^{-1}Kn^2+\frac{4}{3}\alpha^{-1}Kn} \qquad (2)$$

Where Kn is the Knutson number $Kn = 2\lambda_v/d_p$ and α is the mass accommodation coefficient.

In Eq. (2), α is assumed to be 1 while Kn can be expressed in terms of particle diameter and the mean free path of vapour molecules (Pirjola *et al.* 1999) as

$$Kn = \frac{2\lambda_v}{d_p} \qquad (3)$$

The mean free path λ_v is pressure and temperature dependent and can be determined from the following formula from (Willeke 1976)

$$\lambda_v = \lambda_r \left(\frac{101}{P}\right)\left(\frac{T}{293}\right)\left(\frac{1+\frac{110}{293}}{1+\frac{110}{T}}\right) \quad (4)$$

Where P is in kPa and T in K.

Dal Maso, M., M. Kulmala, et al. (2002). "Condensation and coagulation sinks and formation of nucleation mode particles in coastal and boreal forest boundary layers." J. Geophys. Res. **107**(D19): 8097.

Fuchs, N. A. and A. G. Sutugin (1971). Highly Dispersed Aerosols. New York, Pergamon.

Hanson, D. R. and F. Eisele (2000). "Diffusion of H2SO4 in Humidified Nitrogen:â€‰ Hydrated H2SO4." The Journal of Physical Chemistry A **104**(8): 1715-1719.

Pirjola, L., M. Kulmala, et al. (1999). "Formation of sulphuric acid aerosols and cloud condensation nuclei : An expression for significant nucleation and model comparison." Journal of Aerosol Science **30**(8): 1079-1094.

Willeke, K. (1976). "Temperature Dependence of Particle Slip in a Gaseous Medium." Journal of Aerosol Science **7**: 381-387.