

**Ultrafine aerosols in
urban air: spatial
distribution and
evolution**

M. Dall'Osto et al.

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On the spatial distribution and evolution of ultrafine aerosols in urban air

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Abstract

Sources and evolution of ultrafine particles ($<0.1\mu\text{m}$ diameter) were investigated both horizontally and vertically in the large urban agglomerate of Barcelona, Spain. Within the SAPUSS project (Solving Aerosol Problems by Using Synergistic Strategies), a large number of instruments was deployed simultaneously at different monitoring sites (road, two urban background, regional background, urban tower 150 m a.s.l., urban background tower site 80 m a.s.l.) during a 4 week period in September-October 2010. Particle number concentrations ($N_{>5\text{nm}}$) are highly correlated with black carbon (BC) at all sites only under strong vehicular traffic influences. By contrast, under clean atmospheric conditions (low condensation sinks, CS) such correlation diverges towards much higher N/BC ratios at all sites, indicating additional sources of particles including secondary production of freshly nucleated particles. This is also evident in the urban background annual mean diurnal trend of N/BC, showing a midday peak in all seasons. Size-resolved aerosol distributions (N_{10-500}) as well as particle number concentrations ($N_{>5\text{nm}}$) allow us to identify two types of nucleation and growth events: a regional type event originating in the whole study region and impacting almost simultaneously the urban city of Barcelona and the surrounding background area; and an urban type which originates only within the city centre but whose growth continues while transported away from the city to the regional background. Furthermore, during these clean air days, higher N are found at tower level than at ground level only in the city centre whereas such a difference is not so pronounced at the remote urban background tower. In other words, this study suggests that the column of air above the city ground level possesses the best compromise between low CS and high vapour source, hence enhancing the concentrations of freshly nucleated particles. By contrast, within stagnant polluted atmospheric conditions, higher N and BC concentrations are always measured at ground level relative to tower level at all sites. Our study suggests that the city centre is a source of both non-volatile traffic primary (29–39 %) and secondary freshly nucleated particles (up to 61–71 %) at all sites. We suggest that organic

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compounds evaporating from freshly emitted traffic particles are a possible candidate for new particle formation within the city and urban plume.

1 Introduction

The quality of air we breathe is defined by a number of air pollutants that have adverse effects on human health and the environment. Ultrafine particles (UF) are defined as those with diameters smaller than 0.1 μm (100 nm), and make a dominant contribution to urban total particle number concentrations, but only a small contribution to particle volume or mass (Harrison et al., 2000a; Pope et al., 2002). Toxicological studies have suggested that UF, the main component of ambient particles by number, are more toxic than coarser particles, per unit mass (Davidson et al., 2005; Seaton et al., 1995). Apportioning the sources of ultrafine particles has become of large interest in urban areas where population density and human activities are highly concentrated. Moreover, Reddington et al. (2011) recently stressed the importance of understanding the relative contribution of primary and secondary particles in regional and global aerosol so that models can attribute aerosol radiative forcing to different sources.

There are at least two main sources contributing to particle number concentrations in the urban atmosphere: primary and secondary. Traffic is often found to be the most important primary source for ultrafine particles in urban areas (Aalto et al., 2005; Morawska et al., 2008). In general, particles from vehicle exhaust can be divided into two broad categories, depending on the location of their formation. They can be formed in the engine (mostly 30–500 nm agglomerates of solid phase carbonaceous material) or in the atmosphere after emission from the tailpipe (mostly smaller than 30 nm, semi-volatile and consisting mainly of hydrocarbons and hydrated sulfuric acid). Particle formation in dilute engine exhaust depends strongly on meteorological factors (Zhang et al., 2004). For example, strong winds result in both higher particle numbers and smaller modal diameters (Charron and Harrison, 2003).

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Secondary particles formed by nucleation are generated by gas-to-particle conversion. Such particles have been observed at many surface locations around the world and also within the free and upper troposphere (Kulmala et al., 2004). New particle formation events can occur on a large regional scale, and can impact urban areas (Stanier et al., 2004; Wehner et al., 2007; Hussein et al., 2009; Jeong et al., 2011; Boulon et al., 2011).

In general, new particle formation (NPF) events in the urban atmosphere are expected to be far less favoured than in the rural atmosphere due to the high existing surface area for condensation of nonvolatile materials needed for homogeneous nucleation. Despite this, observational studies have suggested that new particle formation occurs in urban atmospheres (Harrison et al., 2000b; McMurry et al., 2000; Pey et al., 2008). Depending on the location, nucleation events in urban atmospheres can occur on approximately 5 % of the days observed (Alam et al., 2003) up to 25–30 % (Stanier et al., 2004; Petaja et al., 2007; Salma et al., 2011).

Little is known about the vertical distribution of primary and secondary UF particles in the urban atmosphere. Urban UF flux cycles showed clear diurnal trends which were linked to traffic activity and local sources (Martin et al., 2009). However, UF concentrations at urban street level are often found significantly higher than those at rooftop levels (Vakeva et al., 1999; Hitchins et al., 2002; Longley et al., 2003). Li et al. (2007) also reported that the particle number concentrations in both the nucleation mode and in the Aitken mode significantly dropped with height. By contrast, short-time non-simultaneous measurements suggested larger concentrations of 5–30 nm aerosols at rooftop level than at street level (Kumar et al., 2009). Dall'Osto et al. (2011) recently provided compelling evidence for the volatile loss of semi-volatile hydrocarbon components of the aerosol during advection of traffic-generated particles into a park and vertical transfer to a tower 160 m a.g.l. At the tower site, within one hour the traffic nucleation mode particles are totally lost by evaporation from the measured size distribution having shrunk to below 5 nm diameter (Dall'Osto et al., 2011).

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Even less is known about the vertical distribution of secondary new UF particles, especially in the urban atmosphere. The origin of NFP is still uncertain, with evidence for both new aerosol particle formation at the surface layer mixing upwards (O'Dowd et al., 2009) and nucleation occurring aloft and subsequently blended downwards (Siebert et al., 2004). Up to which altitude these NPF events take place, and where they are initiated is still under debate. Crumeyrolle et al. (2010) observed that NPF events were limited to the boundary layer while Hamburger et al. (2010) have observed high concentrations of ultrafine particles in the upper free troposphere. Wehner et al. (2010) observed peaks in the number concentrations of ultrafine aerosol particles at ground level connected with new aerosol particle formation in the residual layer by boundary layer development and vertical mixing, and Pryor et al. (2011) concluded that the variation of the ultrafine particle concentrations was due to in situ production rather than horizontal advection. The occurrence of new particle formation episodes depends on various factors including the emission strength of precursors, composition and number concentration of pre-existing aerosol population, meteorological parameters (in particular solar radiation, temperature and relative humidity) and photo-chemical processes (Kulmala et al., 2001, 2004).

In summary, the evolution of primary anthropogenic particles and the formation of new secondary particles still remains one of the greatest challenges in urban atmospheric aerosol research. Primary and secondary UF particles may have different chemical composition and hence different toxicity. Furthermore, UF particles generated in urban areas are important sources of cloud condensation nuclei (CCN), and hence important for the climate (Spracklen et al., 2008; 2010).

This study aimed to provide extensive measurements on particle number concentrations in urban air, including in the vertical above the surface where highly dynamic conditions may play a role. The study area is a large urban European Mediterranean agglomerate which, under high solar irradiance, is a very favourable location for nucleation processes. Pey et al. (2008) first studied events and cycles of urban aerosols in the Western Mediterranean, and a follow-up study (Pey et al., 2009) reported source

apportionment of urban fine and ultrafine particle number concentration, with traffic being by far the most important component (69 %). Recently, Reche et al. (2011) elaborated new considerations for black carbon (BC) and particle number concentrations (N) for air quality monitoring across different European cities, showing that in Barcelona nucleation events can occur in the middle of the day all year round. In this work, we extend previous measurements by using a three-dimensional approach to study the sources and the evolution of UF particles.

2 Methodology

2.1 SAPUSS project and measurement sites

The SAPUSS campaign (Dall'Osto et al., 2012a) was carried out in the region of Barcelona, a city located in the Western Mediterranean basin (WMB) in the north-east (NE) part of Spain. With a population of about 1.7 million inhabitants in the city and around 4 million in the surrounding area, Barcelona is the fifth most populated metropolitan area in Europe. The sampling campaign took place between 20 September and 20 October 2010 (local time, UCT + 2). The unique approach involved a large variety of instrumentation deployed simultaneously at 5 monitoring sites in the metropolitan area, shown in Fig. S1 and a regional background site:

- Road site (RS, 41° 23' 18" N; 02° 09' 0" E) was situated in a car park next to C/ Urgell, a major highway composed of a one-way four lane road. Vehicle intensity for the measurement period was about 17 000 vehicles per day.
- Urban Background site (UB, 41° 23' 15" N, 02° 07' 05" E) was situated at the northwestern periphery of the city centre in a small park (elevation 80 m a.s.l.) situated about 6 km from the sea. This monitoring site was used in previous studies, providing important information on the variability of UF particles in Barcelona (Pey et al., 2008, 2009; Reche et al., 2011).

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- Tower Mapfre site (TM, 41° 23' 16" N, 2° 11' 51" E) is a skyscraper (154 m a.s.l.) in the Port Olímpic, about 200 m from the sea. A large recreational area, as well as ring road motorways can be found at street level.
- Tower Collserola site (TC, 41° 25' 02" N, 02° 06' 51" E) is a telecommunications tower located at 445 m a.s.l. Measurements were taken on the fourth floor, at 80 m a.g.l. (525 m a.s.l.).
- Due to both limited access and space at the TC monitoring site, the Fabra Observatory (41° 25' 56" N, 2° 07' 27" E) was also used as a monitoring site during the SAPUSS project. The Fabra Observatory is an astronomical observatory at 415 m altitude above sea-level of the sea, and located about 450 m (900 m road distance) from the TC site. Measurements were taken up in the terrace at about 10 m from the ground. Although not ideal, we treat the Collserola Tower (TC) and the Fabra Observatory (TC ground, TCg) as one monitoring site, but we will clearly state when measurements were taken at ground level (TCg, 10 m above ground, 425 m a.s.l.) or at higher level (TC, 80 m above ground, 525 m a.s.l.). It is important to note that whilst TM is well within the Barcelona urban city centre, TC is located in the hills of the urban background of Barcelona (Fig. S1).
- Montseny Regional Background site (RB, 41° 46' 45" N 02° 21' 29" E) is part of the EUSAAR network (European Supersites for Atmospheric Aerosol Research, <http://www.eusaar.net>, abbreviation "MSY") and is located within a regional natural park about 50 km to the north-northeast side of the city of Barcelona and about 30 km from the Mediterranean coast. This RB site is only used in this study to describe an urban nucleation event presented in Sect. 3.3.

In summary (see Fig. S1), the RS site and the TM site are located in the core of the busy city centre of Barcelona, at ground level and at about 154 m a.s.l., respectively. In contrast, the UB site is more representative of the urban background conditions at ground level. Additionally, the TCg and the TC sites are indicative of the outskirts of

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the Barcelona agglomerate, and are confined to the hills (TCg, 10 m above ground, 425 m a.s.l. and TC, 80 m above ground, 525 m a.s.l., respectively) about 4 km from the city centre. Finally, the RB site reflects more the regional background conditions. It is worthwhile to stress that the two monitoring towers are the tallest buildings within several kilometres of the sites (Fig. S1), with good exposure to winds from all directions. The TCg, TC and RB sites are all aligned with the Barcelona city centre in a position to receive urban plumes from the city, especially in the afternoon when the sea breeze arrives from the south-west encountering in order: TM, RS, UB, TCg-TC, RB.

2.2 Instrumentation

2.2.1 Meteorological and remote sensing parameters

Meteorological variables (atmospheric pressure, wind components, solar radiation, temperature and relative humidity) were recorded at all the SAPUSS monitoring sites during the whole field study. The atmospheric boundary layer (ABL) above Barcelona was monitored by simultaneous measurements of two Jenoptik CHM15K LIDAR ceilometers (Heese et al., 2010; Martucci et al., 2010) with 1064 nm wavelength and 15 km vertical range. The ABL height and vertical structure can be retrieved with standard 30 s and 15 min temporal and vertical resolution, respectively, by applying the Temporal-Height-Tracking (THT, Martucci et al., 2010; Haeffelin et al., 2011) algorithm to the LIDAR returns from the CHM15K). The vertical profiles of atmospheric temperature, pressure and relative humidity as well as wind components were also obtained by radiosoundings performed twice per day (at 12:00 UTC and 00:00 UTC) at the UB site.

2.2.2 Aerosol properties

Total particle number concentrations were measured at all the SAPUSS monitoring sites. The same type of Condensation Particle Counter (CPC) (TSI CPC Model 3022A; lower 50 % cut-point 7 nm) was simultaneously deployed at 3 of the SAPUSS sites (RS,

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TM, TC), whilst the instruments at the RB and UB sites were TSI water based (WB) CPCs 3785 (with a lower 50 % cut-point at 5 nm) which would lead to slightly higher readings relative to the Model 3022A instrument. Instruments were inter-compared before and after the campaign, giving excellent agreement, with uncertainty of about 5 % both before and after the campaign. CPC data capture ranged between 65 % and 83 %, with simultaneous measurements at four sites taken for 55 % of the time (Fig. S2). Nano CPCs (TSI 3025 and TSI WB 3786, 50 % cut-point at 3 nm) were also located at the TM tower site (20–27 September 2010) and at the TC tower site (24–26 September 2010), respectively. A number of different Scanning Mobility Particle Sizer (SMPS) systems were deployed simultaneously at 4 different sites (Fig. S3), providing size resolved aerosol particle number concentrations. In summary, during the SAPUSS experiment there was a total of 4 Differential Mobility Analysers (DMAs) and 11 CPCs deployed. Black Carbon mass was derived by light absorption using both a McGee scientific aethalometer AE 16 and a Multi-Angle Absorption Photometer (MAAP). A total of 4 MAAP instruments were deployed most of the time during the SAPUSS whereas an aethalometer was deployed at the TM site. MAAP instruments gave excellent agreement between them (within 5 %). The MAAPs and the aethalometer were inter-compared before and after the field study, also giving excellent agreement.

3 Results

3.1 N and BC concentrations across different monitoring sites

The highest concentration of averaged particle number concentrations (N) was found at the RS ($12\,890 \pm 6\,300 \text{ cm}^{-3}$), followed by UB ($10\,490 \pm 5\,900 \text{ cm}^{-3}$), TM ($9\,588 \pm 6\,400 \text{ cm}^{-3}$) and TC ($5\,500 \pm 3\,800 \text{ cm}^{-3}$) sites. Previous studies at the UB (Reche et al., 2011) reported higher values for the 2009 annual mean (about $17\,000 \pm 5\,000 \text{ cm}^{-3}$), although similar values were seen when considering the 2010 annual mean (about $12\,000 \pm 4\,000 \text{ cm}^{-3}$) (C. Reche, personal communication, 2012).

A similar trend was seen for BC: the highest values for RS ($2987 \pm 1700 \text{ ng m}^{-3}$), followed in the same order by UB ($1350 \pm 1100 \text{ ng m}^{-3}$), TM ($1405 \pm 1500 \text{ ng m}^{-3}$) and TC ($650 \pm 450 \text{ ng m}^{-3}$). This is consistent with road traffic being a major source of both BC and N. The slightly higher value of BC at the TM site relative to UB is likely due to the busy highways and roads found below the TM site. The diurnal variation of BC and N for the four different sites is presented in Fig. 1. As expected, the BC trends show a marked profile peaking during the morning and evening traffic rush hours at the urban sites RS, UB and TM. In contrast, the TC site shows a peak in the late morning due to the fact that the urban pollution arrives at this background location at a later time, when the boundary layer is fully developed and the sea breeze commences, bringing plumes from the city to the TC site. The diurnal trends of N are more complex and do not always follow those for BC. Reche et al. (2011) have already extensively described the variability of N and BC levels across different European areas. Although in Central and Northern Europe, N and BC levels tend to vary simultaneously, not only during the traffic rush hours but also during the whole day, in urban background stations in Southern Europe maximum N levels coinciding with minimum BC levels and attributed to nucleation episodes are recorded at midday in all seasons. Within the SAPUSS campaign (Fig. 1), a strong peak in N is observed in the morning for the ground sites UB and RS, whereas the tower sites TM and TC do not follow the BC diurnal profiles. N concentrations at TM show a slow-rising daytime elevation. Similar behaviour is seen also at the TC site: a broader peak between 11:00 and 19:00 peaking at 15:00 (Fig. 1). When considering the weekdays/weekend (WD–WE) variability, all the sites present a reduction for both N and BC concentrations (Table 1), implying a strong traffic influence on N and BC concentrations. The RS shows remarkably similar reduction for both N and BC (38 %). TC also presented similar reduction for N and BC (25–26 %), whereas a smaller reduction was seen for the UB site for N (18 %) than BC (25 %). The WD–WE variation at the tower TM site shows a strong reduction for the BC concentrations (35 %, similar to the road RS site) but a much smaller one for N (9 %).

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Size resolved particle number concentrations have been classically described as a function of air mass type (marine, continental, mixed) (Birmili et al., 2001; O' Dowd, 2001). Air masses of different origins have different meteorological, physical and chemical characteristics, and for example the probability of occurrence of NPF events at a given location and time depend not only on local emissions, but also on long range transport (Sogacheva et al., 2007; Tunved et al., 2006). Figure 2 shows the BC and N concentrations for different air mass types encountered during the SAPUSS study. Out of 30 days of measurements, 9 were classified as cP (continental Polar, or regional air masses scenarios) and 7 as mP (marine Polar, or Atlantic air mass scenarios) (Dall'Osto et al., 2012a).

The BC concentrations presented a similar trend for all four sites: the lowest concentration was detected under mP conditions whereas the highest were associated with the cP category (Fig. 2). It is important to remember that air arriving from the Atlantic will in most cases have traversed all of Spain (and possibly Portugal) and therefore collected significant pollution on the way. However, the main significance of mP air masses is that they are faster moving and therefore cause enhanced dilution of local pollutants. By contrast, an opposite trend was seen for N: the highest concentrations were recorded during the mP air mass type. Table 1 shows the ratio between N and the more conservative variable BC (Reche et al., 2011; Dall'Osto et al., 2011). When considering all the data, a lower N/BC ratio is seen for the RS relative to all the other three, remaining constant at about 4 under different air masses (Table 1). However, the other sites present very different ratios under different air mass scenarios, implying different evolution of particles leaving the main traffic source as well as contributions from secondary particles and from primary particles from other non-traffic sources.

3.2 Nucleation events

Newly formed nucleating particles are typically of size 1–2 nm in diameter and not easily measurable by current routine particle counting techniques. The classification of nucleation events is yet somehow subjective as a standardised method does not exist.

5 It is important to bear in mind that, within the urban atmosphere, great care has to be taken when assigning nucleation events, as UF traffic plumes or growth of existing UF can be wrongly assigned as nucleation events. In other words, primary emissions or growth of primary emitted urban UF particles cannot be classified as new nucleation events. In order to identify the nucleation events during SAPUSS, we used two different methodologies. The first method was based upon tandem particle counters with lower detection limits (50 % efficiency of detection) of 3 and 7 nm (respectively). The measured difference in N within the size range 3–7 nm gives an indication of particles recently nucleated (Alam et al., 2003). The second method was that proposed by Dal Maso et al. (2005), where SMPS plots were visually analyzed and several criteria had to be met to classify a NPF event such as: (1) a distinctly new mode of particles must appear in the size distribution; (2) the mode starts in the nucleation mode size range; (3) the mode prevails over a time period of hours, and (4) the new mode shows signs of growth. Nucleation burst particle formation events can be usually classified into two groups, with and without particle growth after the burst of nucleation mode particles observed (Vana et al., 2008; Cheung et al., 2011). During SAPUSS, only two days could be unambiguously assigned to nucleation events and are described in the following two sections. However, on at least other three days, very high values of N were measured, indicating bursts of nucleation monitored by CPCs ($D_{>5\text{nm}}$) but not by SMPSs ($D_{>10\text{nm}}$). This suggests that nucleation processes, with and without growth, were affecting the SAPUSS study period on about 20 % of the days. Indeed, the importance of nucleation events in the study area is supported by the study of Reche et al. (2011); which showed the highest annual average N in the urban background of Barcelona at midday, stressing the importance of nucleation events in the Mediterranean urban regions relative to other Northern European colder urban environments.

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3.2.1 Regional nucleation

Aerosol properties

5 A clear regional nucleation event was detected simultaneously at the four different urban sites. This event was detected across the entire city, and the SMPS data present the typical “banana” shape (Kulmala et al., 2004). This is seen clearly in the ground level data of the Fabra observatory (TCg, Fig. 3a) consistent with particle number concentrations seen at the ground level UB (Fig. 3b), and the two tower sites of TC (Fig. 3c) and TM (Fig. 3d). Unfortunately, neither particle number concentrations
10 nor aerosol size distributions were available for this day at the RB and RS sites. In summary, for this day SMPS systems were running optimally only at TCg, whereas CPCs were running well at the TC, TM and UB (Fig. 3). The event covers an area of at least 10 km² in a column of air at least 525 m high. This nucleation event was found to occur simultaneously in the city centre as well as in the urban background locations, and was homogeneously distributed across the vertical profile of the urban boundary layer. Whilst the maximum N at both sites above ground (TM, TC) occurs at 10:00–11:00, the maximum N at both sites at ground-level (TCg, UB) occurs at 11:00–12:00; suggesting this nucleation event may have initiated above the urban surface first.

20 Meteorological parameters

The variations of the meteorological parameters for this unique regional nucleation event were compared with the other days of the field study. Very low values of RH were found during the regional nucleation event at all monitoring sites (RH < 40 %
25 between 11:00 and 15:00). This is also seen in the driest air column encountered during the 32 daily radio soundings recorded during SAPUSS (Fig. S4). A unique feature was also the maximum hourly radiation intensity value during this day (781 W m⁻² at 13:00), the second highest value recorded during the whole study.

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It has been observed at different continental locations that new particle formation (NPF) takes place preferentially at low RH (Laaksonen et al., 2008). Hamed et al. (2011) summarised that the increase of coagulation scavenging plays a relatively small role in the inhibition of nucleation at high RH. On the other hand according to Hamed et al. (2011), the low OH concentrations at high RH limits H₂SO₄ levels in the air, concluding that high new particle formation rates rarely occur above 80 % RH. Although solar radiation is likely the trigger for this nucleation event, other meteorological conditions may also influence the new particle formation process. For example, Peter et al. (2010) reported enhanced NPF events associated with mixing between air masses of distinct thermodynamic properties, suggesting that large gradients in temperature and relative humidity may have promoted new particle formation via turbulent mixing. This regional nucleation event occurred after a persistent typical summer recirculation pattern, with low pressure gradients over the Western Mediterranean basin (19–22 September) and a second brief period (23–24 September) characterized by the advection of southern air from North Africa, with intrusion of Saharan Dust. During the evening of the 24 September 2010, air masses changed to a marine Polar trajectory originated from the North Atlantic (Fig. S5). This was associated with the strongest wind speed recorded during SAPUSS on the 25 September, coming from the North and recorded homogeneously across the air column (see radio sounding profile of Fig. S4). The boundary layer was found to grow convectively at about 8:30, reaching a maximum height of 1050 m at about 13:30 (Fig. S5a) and then progressively decreasing. The boundary layer height after 9:00 was found to be 970 ± 250 m, well above all monitoring sites (Fig. S4).

3.2.2 Urban nucleation events

Aerosol properties

Based on CPC measurements, Reche et al. (2011) recently reported annual mean maximum N levels at midday in Barcelona, indicating that nucleation events occur

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all year round. However, in the absence of SMPS measurements it is difficult to confirm if the particles grow to larger sizes. When measuring size resolved particle concentrations larger than 13 nm across a whole year near the SAPUSS UB site, photochemically induced nucleation particles make only a very small contribution to the total particle number concentration (2–3% of the total) (Dall'Osto et al., 2012b). The work of Reche et al. (2011) shows that the city is a source of new secondary particles, but these particles often fail to grow above the SMPS detection limit of about 10 nm (Dall'Osto et al., 2012b). This only implies that nucleating particles cannot easily grow within the urban agglomerate of Barcelona, but it is not known if such particles are able to grow while being transported away from the city centre with the afternoon sea breeze typical of Mediterranean coastal urban agglomerates. However, our study seems to support such a hypothesis. Indeed, a second type of nucleation event, initiating only in the urban centre of Barcelona and not across the whole region, was observed during the SAPUSS campaign, Fig. 4a. Afternoon values for N are higher than those in the morning at the ground sites UB and RS, as reported in the annual mean (Reche et al., 2011). The tower site within the city (TM) is surprisingly found to possess even higher afternoon N concentrations relative to the ground sites. The TC also shows an increased N concentration in the afternoon period, but peaking later at about 17:00. This is in contrast to the more general behaviour encountered during stagnant polluted days, Fig. 4b (29 September 2011), where a strong morning rush hour traffic peak is seen for both ground sites RS and UB. Furthermore, higher particle number concentrations can be seen for both ground sites (RS, UB) relative to the tower sites (TM, TC) at all hours of the day. A slight increase in N is seen for the TM at 10:00 and in the afternoon, whereas a more constant pattern can be seen for the TC site.

By deploying simultaneously four SMPS instruments, a clear evolution of the nucleation event can be better seen in Fig. 5: nucleating particles increase in concentration in the city centre (RS and UB) at around 14:00, and are subsequently transported by the sea breeze to the background tower (TC) and to the regional background site

(RB) in the evening. This strongly suggests that this type of nucleation event begins in the urban areas of Barcelona and its growth continues as the plume travels away from the city. When calculating the growth rate, a constant pool of nanoparticles was seen at both the ground sites RS and UB, with a constant unimodal distribution centred at about 16 ± 2 nm between 14:00 and 17:00 at the UB. In contrast, a growth rate of about 2.8 nm h^{-1} can be seen at the TC site from 14:00 (23 ± 7 nm) to 19:00 (37 ± 2 nm). A lower growth rate was seen at the regional site RB: 2.1 nm h^{-1} from 14:00 (37 ± 2 nm) to 19:00 (48 ± 2 nm), indicating a decrease of the growth rate with the distance from the city centre.

Meteorological properties

The urban nucleation event on 5 October 2010 was associated with the strongest radiation intensity during the entire SAPUSS study. However, it presented dry intermediate RH conditions (52 %) and average temperatures (19.8°C). The radio sounding measurements taken during this day also present average conditions with no peculiar feature in any of the parameters measured (Fig. S4). The beginning of the nucleation event was recorded at about 15:00, concomitant to the maximum hourly average temperature of the day whereas the maximum solar radiation intensity (860 W m^{-2}) was found at 13:00. Like the regional nucleation event, this urban nucleation event also occurred after a persistent typical summer recirculation pattern (29 September–2 October) and a second brief period of southern air from North Africa (2–4 October). During the evening of the 4 October 2010, the air masses changed to a marine Polar trajectory originating from the North Atlantic (Fig. S5). With regard to the boundary layer structure, a marked difference relative to the regional nucleation event can be seen: some light cloud present till 11:30 limits the convective growth of the PBL (Fig. S4). Boundary layer conditions for this day were found similar in the afternoon, well above all the monitoring sites (>1200 m, Fig. S4).

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4 Analysis and discussion

4.1 Origin of the nucleation events

The first step of a nucleation event is the formation of an initial nucleus, followed by growth of the particles to larger sizes. In the continental boundary layer so-called “regional nucleation events” are common. Observed aerosol formation events very often, if not always, occur during daytime, as photochemistry plays a central role in this process. Such events can cover large spatial scales. For example, Wehner et al. (2007) found that new particle formation events vertically well mixed within the entire boundary layer occurred at all monitoring sites located over more than 400 km apart. However, the nucleation phenomenon could be interrupted when clouds were limiting the incoming solar radiation. Other factors that favour atmospheric aerosol formation include a sufficiently low pre-existing aerosol concentration, low relative humidity, and high vapour source production rate. The most important of these factors seem to be the high vapour source rate and the low sink due to pre-existing particles (Kulmala and Kerminen, 2009).

The unique conditions of the regional nucleation event detected during SAPUSS centre upon the incursion of cool and very dry air masses (Figs. S4, S5) associated with the passage of a cold front leading to stronger winds and probably strong turbulent mixing. Numerical calculations have shown that the mixing of two air parcels with different properties such as precursor concentration, temperature and relative humidity generates more particles through nucleation than would be the case for the two air parcels separately (Nilsson and Kulmala, 1998; Jaenisch et al., 1997; Khosrawi and Konopka, 2003).

As regards the time of nucleation, this event was observed in the late morning (09:00 to 12:00), and is coupled to a clear transition from an atmosphere with low turbulence intensity and weak vertical velocities, to much a weaker vertical gradient of wind speed, increased turbulence intensity and stronger downwards vertical velocities, consistent

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with growth of the mixed layer and entrainment of air from the residual layer, as recently summarised by Pryor et al. (2011).

By contrast, whilst the urban nucleation event was also associated with mP air mass back trajectories, it did not present any unique meteorological features like the regional event. The urban nucleation event occurs in the afternoon, at about 15:00, and is not linked with the morning urban boundary layer dynamics. The only meteorological variable peaking at 15:00 was found to be the temperature, with wind speed being at maximum level 15:00–17:00. In this regard, it is important to remember that when turbulence weakens in the afternoon, due to the decreasing sensible heat flux, the temperature inversion builds up again and the depth over which mixing occurs becomes shallower. In these afternoons and after-sunset conditions, the strongest aerosol gradient corresponds to a residual aerosol layer aloft the actual surface mixed layer (Haeffelin et al., 2012). Indeed, Fig. S4 shows a lower boundary layer at 14:30–15:00 which may have affected the nucleation event.

However, meteorological conditions are not the only key parameters affecting the nucleation events. Chemical compounds are needed in order to form a nucleus and contribute to subsequent growth of particles. Sulfuric acid is a key component in particle formation processes (Sipila et al., 2010). As regards SO₂ concentrations, both regional and urban nucleation events at all monitoring sites (excluding the RS) exhibited low concentrations (<1 ppb) suggesting the anthropogenic emissions affecting the study area were diluted by the windy Atlantic air mass scenarios (Fig. S4). Although such low concentrations could be sufficient to initiate nucleation, it is likely that other chemical components supported the new particle formation and growth. Among the most cited mechanisms are the binary (Vehkamaki et al., 2002) and ternary (Napari et al., 2002) nucleation. Laaksonen et al. (2008) reported that VOC oxidation products may also have a key role in determining the spatial and temporal features of nucleation events. The conclusion was supported by aircraft measurements, which showed that the nucleation even, started in the surface layer, i.e. near the VOC source, and evolved together with the mixed layer. Lately, the role of organics in new particle formation has

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gained greater interest. Metzger et al. (2010) showed that the role of organics is important for the growth of the nucleated particles and for the nucleation itself. Riipinen et al. (2011) reported that the condensation of organic vapours is a crucial factor governing the lifetimes and the climatic importance of the smallest atmospheric particles.

5 Figures 1 and 4 are in line with the work of Reche et al. (2011) reporting UB ground level annual maximum N at midday attributed to new particle formation. Additionally, our study suggests that the formation and/or growth of UF in urban nucleation events is enhanced in the vertical column above the city centre relative to the urban surface and the air column above the urban background. In other words, when considering all
10 monitoring sites, the highest UF concentrations are found above the city centre, in our case at 150 m a.s.l. The lifetimes of such particles depend on the competition between their condensational growth and coagulative scavenging (CS) (Kerminen and Kulmala, 2002). Boulon et al. (2010) found a positive correlation between the NPF event frequency and the CS at elevated altitudes (Jungfraujoch, 3580 m a.s.l.), indicating that
15 the vapour source of NPF is likely linked to polluted conditions and that at this site the presence of condensing vapours is a more important parameter for NPF to occur than a low CS. Nishita et al. (2008) also found nucleation mode particles in the mixed layer air, but never in the free tropospheric air, suggesting that new particle formation occurred in the air transported from the mixed layer. Because CS was consistently higher
20 in the mixed layer air than in the free tropospheric air, the source rate of condensable vapors in the mixed layer air must be higher than that in the free tropospheric air, overcoming the inhibitory effects on NPF attributable to high CS.

25 In urban and suburban vertical air columns, traffic emissions are the dominant source of most volatile organic compounds (VOCs) to the atmosphere, with alkanes accounting for the largest contribution (Langford et al., 2010). Flocas et al. (2003) reported that at altitudes of 300–400 m, the VOC concentration levels remained high, suggesting the influence of surface sources. Additionally, it was found that the aromatic fraction gains significance with height as compared with the ground. By contrast, no differences were

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observed in the VOCs concentrations within the 200 m layer above the ground due to the efficient vertical mixing (Velasco et al., 2008).

The relevance of organic compounds in aerosol formation was confirmed by both observation and simulated vertical profile of particle number concentrations during nucleation event days which showed a highly dynamical picture (Lauros et al., 2011). Furthermore, it was shown that deposition affects noticeably only the smallest particles in the lowest part of the atmospheric boundary layer. Primary aerosol sources can suppress nucleation through providing a condensation sink for sulfuric acid vapor and scavenging newly formed particles through increasing the coagulation sink.

If traffic related particles evaporate (Dall'Osto et al., 2011), then a source of VOC is provided in the urban air column, whilst the CS decreases with height (evaporation of primary particles and dispersion of other sources i.e. terrestrial dust) as does temperature. Our study suggests that the lower CS found above the city as well as the additional availability of VOC possibly coming from traffic particles evaporation make an environment which favours the nucleation of particles.

4.2 N/BC in relation to different variables

Correlations between BC and N are repeatedly observed in urban environments owing to the impact of vehicle exhaust emissions (Zhu et al., 2002; Rodriguez and Cuevas, 2007). However, whilst BC is an appropriate tracer for the intensity and proximity of traffic emissions (Turpin and Huntzicker, 1995), N can also be affected by the occurrence of nucleation processes (Reche et al., 2011). Figure 6 investigates the correlations of N versus BC at four different monitoring sites. It is important to bear in mind that the two nucleation days were excluded from the current analysis, where such data would be confined in the top left part of the plots (high N and low BC). The variation of N/BC with the following parameters is investigated; NO_x (Fig. 6a), RH (Fig. 6b), temperature (Fig. 6c) and solar radiation (Fig. 6d). Figure 6a shows a good linear correlation ($R^2 = 0.7\text{--}0.75$) between N and BC at the ground RS and UB sites, further supported by the correlation with NO_x presented in the colour plot and implying a direct traffic

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source of N. In contrast, the N/BC plots for the two tower sites (TM and TC) suggest a linear correlation between N and BC only where a NO_x dependency is also found. In other words, there is a large ensemble of points to the top left part of the N/BC diagram for TC and TM, indicating higher N/BC ratios not correlated with NO_x (blue points) and suggesting an additional source of UF particles at the tower sites (TM, TC) but not at the ground sites (UB, RS). Further differences between the ground sites (RS, UB) and the tower ones (TC, TM) should be noted in Fig. 6b. Whilst the ground sites do not have a clear RH dependency, the tower sites possess the highest N/BC ratios (top left part of the plot) associated with the driest conditions (blue and green points). An even clearer gradient is seen at all sites when temperature values are considered (Fig. 6c), with higher N/BC recorded with the highest temperatures. However, the correlation between the N/BC plot and temperature at the road site is surprising. Previous studies (Olivares et al., 2007) reported an increase in N with decreasing temperature in a street canyon in Stockholm. Bigi and Harrison (2010) also found N showing a minimum corresponding with maximum air temperatures in August in London, consistent with loss of nanoparticle by evaporation during atmospheric transport. In this work we find opposite trends: for a given BC concentration, higher N are recorded with higher temperature, implying that other sources of UF particles are impacting the RS monitoring site. Finally, Fig. 6d shows the N/BC plot as function of solar radiation: still an unclear dependence for the ground sites UB and RS, but a strong gradient for the two tower sites (TM, TC) with high solar radiation values found above the intercept N/BC.

In summary, N and BC seem linearly correlated at all sites only when there is also a NO_x correlation with them. However, we find that the N/BC plots diverge from linearity towards higher values (top left part of the diagram, Fig. 6) when primary traffic conditions are not the only dominating factor. A strong dependence upon RH and radiation values for the two towers and strong temperature dependence for all ground and tower sites should also be kept in mind. Figure 7 shows the regression lines for the four sites, excluding the regional and urban nucleation events (25 September and 5 October 2010). Fairly good linear correlations are found ($R^2 = 0.41\text{--}0.77$). The intercept

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gives an indication of the regional contribution, whereas the gradient reflects a contribution from local sources. The highest intercept is found for the urban tower TM (6700 cm^{-3}) whereas the lowest one is associated with the urban background tower TC (3700 cm^{-3}). This indicates that high background particle number concentrations are seen above the urban tower (TM) in the city centre. However, those concentrations become lower when sampling aerosols at tower level but away from the urban city agglomerate (TC). Whilst N correlates linearly very well with BC at the RS site, the correlations for the other three sites away from traffic hot spots do not. This is particularly well seen for the two tower sites (TM, TC), where a number of points are found at low BC concentrations ($<4\text{ }\mu\text{g m}^{-3}$) but high N ($>10\,000\text{ cm}^{-3}$). By individually inspecting the occurrence of such hourly data points, we found that those are mainly associated with afternoon values of 24, 26 and 27 September 2010. Such days are characterised by similar marine Polar conditions found for the regional nucleation events (25 September 2010), implying that a burst of UF particles was recorded at higher level (TC, TM) but could not be clearly monitored by the CPC or SMPS instruments deployed at ground level (RS, UB). Given the high variability of the scatter plot seen in Fig. 7, the regression lines were calculated separately for each hour of the day for each sampling site (96 regression lines, Fig. S6). Fig. S7 shows the results grouped in 4 time intervals: 00:00–06:00, 06:00–12:00, 12:00–18:00 and 18:00–00:00.

The RS gives an intercept at about $6000\text{ particles cm}^{-3}$, with a reduction of about 25% during morning rush hour. This could be interpreted as higher CS due to fresh primary particles emitted and reducing the background concentrations. The TM and UB site seems to follow a similar pattern, with higher N concentrations during day time. However, it is important to note that all sites present roughly the same regional N concentration at about 6000 cm^{-3} during afternoon time when the boundary layer is fully developed. Much more difficult is to interpret the gradient, which should give a contribution of the primary particles linked to the BC, i.e. non volatile EC core particles linked with traffic emissions. With this assumption, Table 2 shows that under clean conditions (mP air masses), at the road site RS 28% are primary non-volatile (but they

reach 37 % during rush hours), whereas during polluted conditions (mC air masses) higher values are seen (47 %, up to 58 % during morning rush hours). By contrast, the urban tower TM sees similar conditions for the UB site: more primary particles during night time but the least during morning rush hours. The overall resulting scenario is a large proportion of particles not related to non-volatile EC traffic particles (seen also in Fig. S8).

Figure 8 summarises the overall structure of N versus BC at the 4 monitoring sites. The road site shows a flat line at about 4 for most of the day, with a slightly increase at 14:00–15:00, indicating a contribution of new particle formation (higher N/BC) during the warmer part of the day. The UB site shows a similar feature, but a much enhanced N/BC ratio in the afternoon. The two towers show a similar trend, but different from the ground sites. A lower ratio is seen at TC and TM for the morning rush hour. This cannot be explained by dilution as when the boundary layer develops, both BC and N should dilute, leading us to suggest some particles may evaporate (leading to a lower N/BC) when the atmosphere starts to heat up in the morning. In other words, a reduction of the ratio N/BC (07:00–10:00 at TC and TM, Fig. 8) can be explained by some semi-volatile particles evaporating when the urban heat begins to develop. During afternoon, more particles are seen in both towers, higher than at the ground site. This is an indication that nucleation processes are taking place above the city in the afternoon at higher intensity than ground level. However, this process is confined above the city centre as when we move away from the TM and arrive at the urban background tower TC, this is reduced (Fig. 8).

5 Conclusions

Within the SAPUSS project, the sources and the evolutions of UF particles were investigated both horizontally and vertically in the large urban agglomerate of Barcelona, Spain. Ground level measurements were taken in the city centre hot spot as well as in the urban and regional backgrounds. Furthermore, two towers were also used as

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monitoring stations: Tower Mapfre (TM, 150 m above ground, 150 m a.s.l.) located in the city centre, and Tower Collserola (TC, 80 m above ground, 525 m a.s.l.), located in the hills delimiting the urban agglomeration. Our results shows that, under stagnant polluted atmospheric conditions, primary traffic particles are the main source of N and BC, with clear vertical and horizontal concentrations gradients decreasing with the distance from the traffic city centre hot spot. By contrast, under clean conditions (marine Polar air masses, high wind speed and low condensation sinks), the sources and evolutions of UF particles become more complex:

- Regional nucleation events can impact the whole region, including the city centre. Such nucleation events begin in the morning at about 09:00–11:00 and are found with the passage of a new cold dry Atlantic front.
- A second more city-localised type of nucleation event can originate in the city centre. In this case, N is found to have the highest concentrations at 14:00–15:00 at all monitoring sites, where the BC is at its lowest levels. Additionally, we find higher N above the city centre vertical column rather than on the ground. This type of nucleation event is associated with particle growth while being transported away towards the regional background, reaching CCN sizes.

This study suggests that, under sunny, dry and low CS atmospheric conditions, volatility of traffic related particles is driving not only the loss of themselves (by evaporation) but at the same time it could provide additional VOC material in the atmosphere. As pointed out by Kulmala and Kerminen (2009), high vapour source rate and the low CS due to pre-existing particles are the two most important parameters governing new particle formation. We speculate that organic compounds evaporating from freshly emitted traffic particles as possible candidates for new particle formation above the urban surface. In other words, our study suggests that the column of air above the city ground level possesses the best compromise between low CS and high vapour source, hence enhancing the concentrations of freshly nucleated particles. Model simulations indicate that nucleation is a significant source of aerosol particles in the global

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atmosphere (Spracklen et al., 2008). The importance of secondary particle formation on a global scale is uncertain, though it may make a significant contribution to both global condensation nuclei (CN) and cloud condensation nuclei (CCN) concentrations (Spracklen et al., 2008; Spracklen et al., 2010). Reddington et al. (2011) recently quantified how primary particle emissions and secondary particle formation influence size-resolved particle number concentrations in the boundary layer by using a global aerosol microphysics model as well as aircraft and ground site observations. It was found that the major source of uncertainty in CCN-sized particles in polluted European air is the emitted size of primary carbonaceous particles. It is important to understand the relative contribution of primary and secondary particles to regional and global aerosol so that models can attribute aerosol radiative forcing to different sources. This study suggests that, within the urban atmosphere, freshly emitted primary traffic particles can convert into freshly nucleated secondary particles under certain conditions, leading to very complex aerosol dynamics.

Supplementary material related to this article is available online at:
<http://www.atmos-chem-phys-discuss.net/12/16603/2012/acpd-12-16603-2012-supplement.pdf>.

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Table 1. Average N and BC concentrations for weekdays (WD) and weekend (WE) days and relative difference (WD–WE) at four different monitoring sites; and ratios of N/BC for different air mass scenarios.

Site	N (particle cm^{-3})			BC (ng m^{-3})			N/BC (particle cm^{-3} /ng m^{-3})		
	WD	WE	% WD–WE	WD	WE	% WD–WE	All air masses	Marine (mP)	Continental (cP)
RS	15 124	9355	38	3650	2245	38	4	4	4
UB	13 058	9722	26	1394	1182	22	8	10	7
TM	9856	9005	9	1583	1009	36	8	14	8
TC	6190	4613	25	748	547	27	7	11	5

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Table 2. Percentage (%) of primary non volatile traffic particles (EC related) at different time of the day under polluted conditions (cP air masses) and diluted Atlantic conditions (mP air masses) at four different monitoring sites.

hour of the day	RS	UB	TM	TC
% Traffic Primary particles (cP air masses)				
0	20	20	41	50
6	37	18	24	42
12	30	11	35	9
18	24	13	20	15
% Traffic Primary particles (mP air masses)				
0	37	51	51	71
6	58	46	33	64
12	50	32	45	20
18	42	38	48	30

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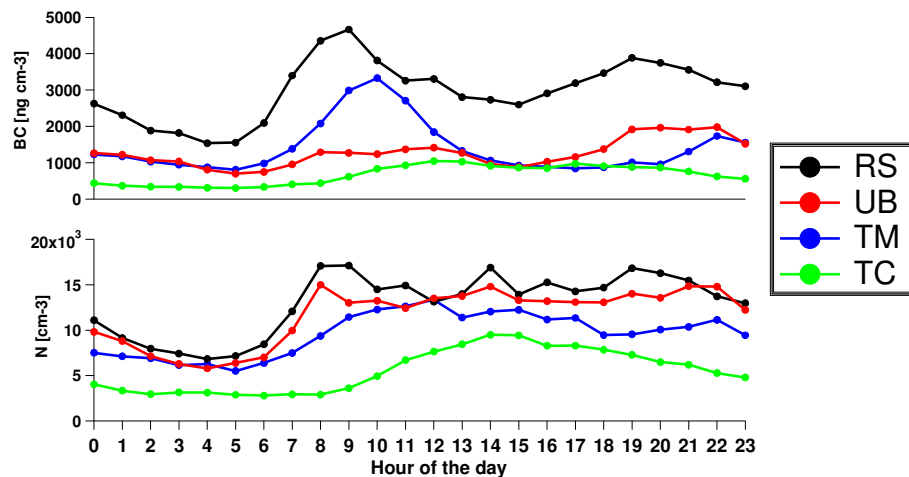


Fig. 1. Diurnal profile of BC and N at four different monitoring sites.

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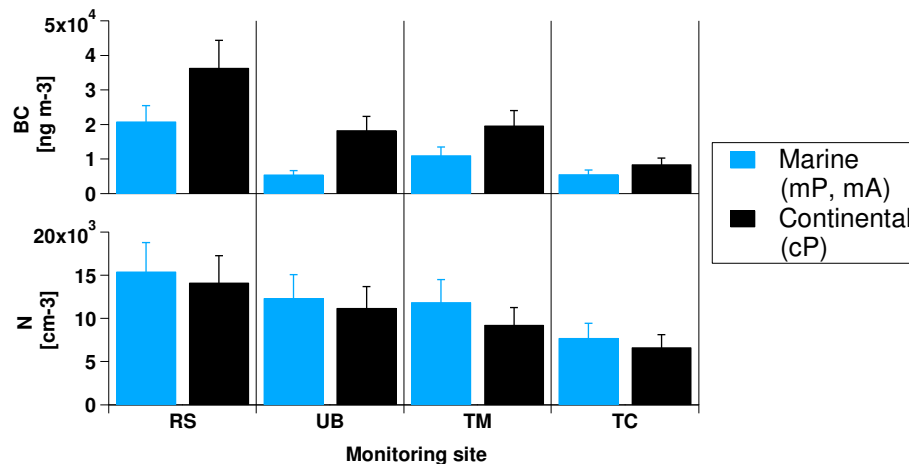


Fig. 2. Average BC and N concentrations under different air mass back trajectories scenarios at four different monitoring sites.

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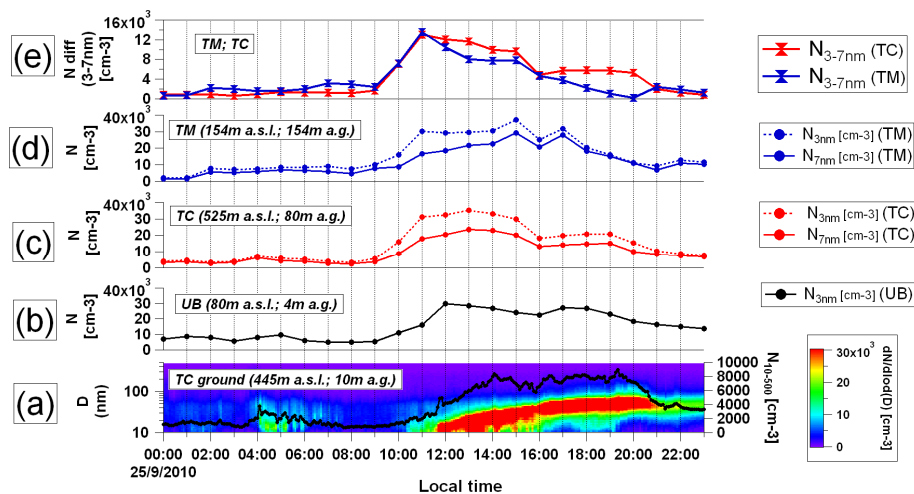


Fig. 3. (a–e) Regional nucleation event observed on 25 September 2011.

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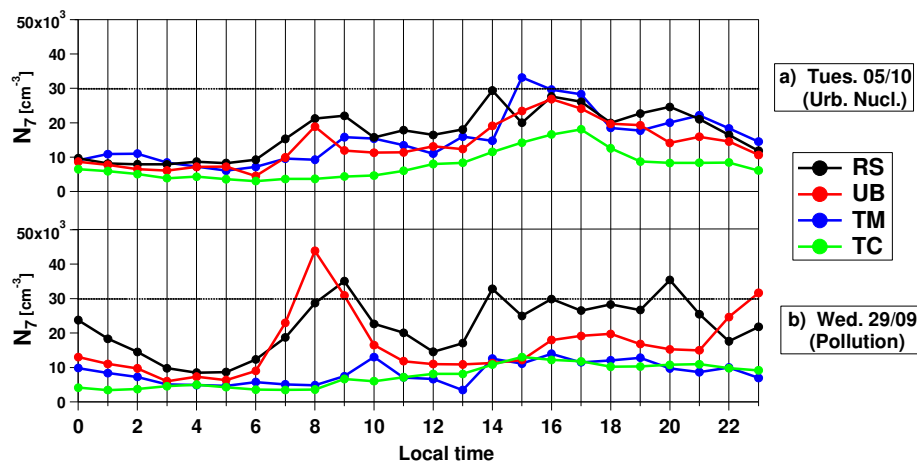


Fig. 4. Diurnal variations of the particle number concentrations for **(a)** urban nucleation event day on 5 October 2010 and **(b)** typical polluted day (29 September 2010) at four different monitoring sites.

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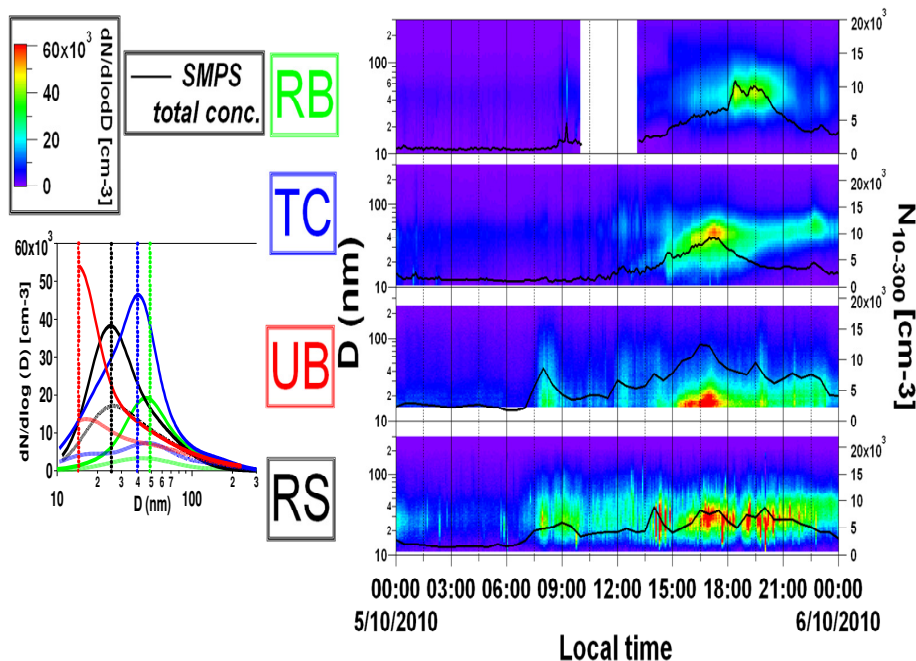


Fig. 5. Aerosol size-resolved particle number concentrations at four different monitoring sites during the urban nucleation event on 5 October 2010. Aerosol size distributions (coloured as the monitoring site names) are also shown on the left panel before (dashed lines, 10:00–14:00) and during the nucleation event (solid lines, 14:00–18:00).

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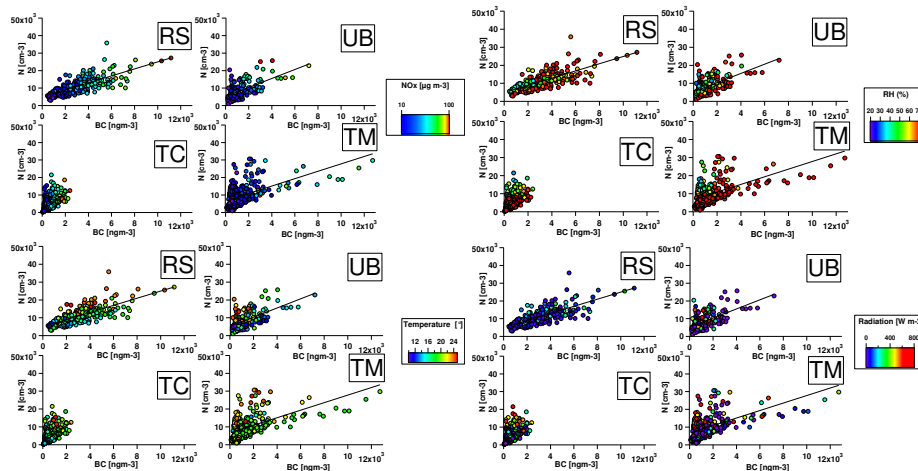


Fig. 6. Hourly black carbon (BC) and particle number concentrations (N) at four different monitoring sites. In contour plots are shown (a) NO_x , (b) RH, (c) temperature and (d) radiation values.

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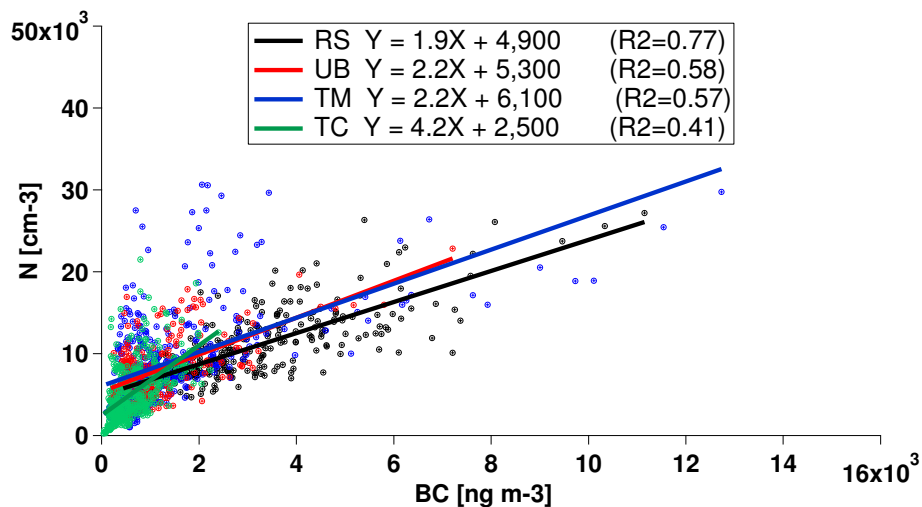


Fig. 7. Linear correlations between BC and N at four different monitoring sites. Note that data of the two nucleation event days (regional and urban) are not included in the chart.

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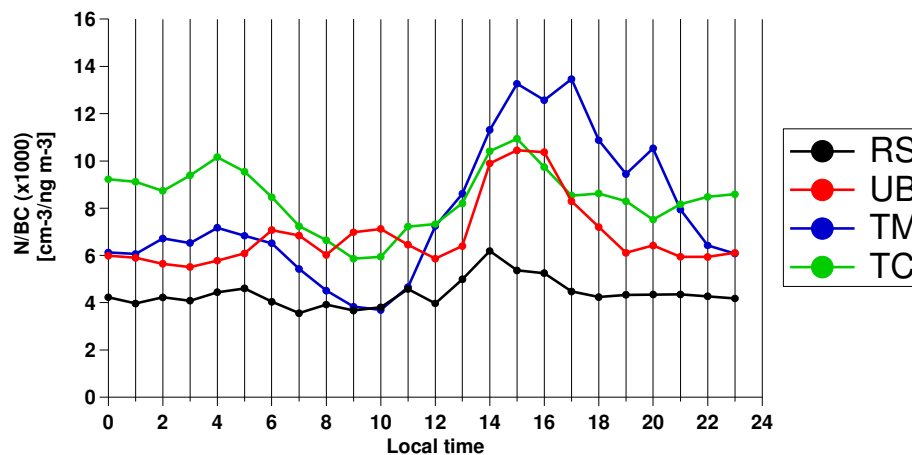


Fig. 8. Diurnal variation of the N/BC ratio at four different monitoring sites.

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