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New particle formation at ground level and in the vertical column over the Barcelona area

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ABSTRACT

The vertical profiles (up to 975 m a.s.l.) of ultrafine and micronic particles across the planetary boundary layer and the free troposphere over a Mediterranean urban environment were investigated. Measurements were carried out using a tethered balloon equipped with a miniaturized condensation particle counter, a miniaturized optical particle counter, a micro-aethalometer, a rotating impactor, and meteorological instrumentation. Simultaneous ground measurements were carried out at an urban and a regional background site. New particle formation episodes initiating in the urban area were observed under high insolation conditions. The precursors were emitted by the city and urban photochemically-activated nucleation occurred both at high atmospheric levels (tens to hundreds of meters) and at ground level. The new particle formation at ground level was limited by the high particulate matter concentrations recorded during the morning traffic rush hours that increase the condensation sink and prevent new particle formation, and therefore restricted to midday and early afternoon. The aloft new particle formation occurred earlier as the thermally ascending polluted air mass was diluted. The regional background was only affected from midday and early afternoon when sea and mountain breezes transported the urban air mass after particle growth. These events are different from most new particle formation events described in literature, characterized by a regionally originated nucleation, starting early in the morning in the regional background and persisting with a subsequent growth during a long period. An idealized and simplified model of the spatial and time occurrence of these two types of new particle formation episodes into, around and over the city was elaborated.

1. Introduction

The origin and processes affecting levels and variability of ambient ultrafine particles (UFP, particles smaller than 0.1 μm in diameter) in urban areas are a hot topic in both air quality and climate sciences (Charlson et al., 1992; Spracklen et al., 2010; Makkonen et al., 2012; IPCC, 2013). The main source of UFP in urban areas is road traffic (Charron and Harrison, 2003; Pey et al., 2009; Dall'Osto et al., 2013; Kumar et al., 2014; Brines et al., 2014a), and especially diesel engines (Morawska et al., 1998; Harris and Maricq, 2001; Rose et al., 2006; Rodríguez et al., 2007). Thus, studies carried out worldwide show that UFP levels are higher close to major road traffic urban arteries compared with those recorded at urban background sites (Zhu et al., 2002; Wehner et al., 2002; Kittelson et al., 2004; Hudda et al., 2010; Padró-Martínez et al., 2012; Fujitani et al., 2012). The spatial distribution of UFP across the urban environment has been studied in detail in a few cities in Europe (Dall'Osto et al., 2011, 2013; von Bismarck-Osten et al., 2013; Salma et al., 2014; Brines et al., 2014a) finding that UFP concentrations markedly increased at road sites with respect to the urban background. In addition to the prevalent primary UFP emissions governing ambient concentrations at urban sites, nucleation, agglomeration, condensation, and evaporation processes have an effect on UFP concentrations and size (Charron and Harrison, 2003; Kulmala et al., 2004; Robinson et al., 2007; Dall'Osto et al., 2011; Harrison et al., 2012).

A number of studies in the beginning of the last decade first pointed to the occurrence of new particle formation episodes in urban environments (Woo et al., 2001; Alam et al., 2003; Stanier et al., 2004). Subsequent studies evidenced the relevance of these episodes in cities from high insolation regions such as southern Europe, California or Australia (Pey et al., 2008; Costabile et al., 2010; Reche et al., 2011; Hudda et al., 2010; Cheung et al., 2011, 2015; Brines et al., 2014b). This influence is so important that, on an annual average, maximum hourly UFP concentrations at the urban background of Barcelona (Northeast of Spain) are recorded at midday simultaneously with the lowest black carbon (BC) concentrations (Reche et al., 2011). The new

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particle formation events studied in the cities of Barcelona, Brisbane, Los Angeles, Madrid, and Rome by Brines et al. (2014b) showed nucleation bursts without important subsequent particle growth, different from the typical 'banana-like' nucleation episodes usually described in regional background environments (Kulmala et al., 2004; Kulmala and Kerminen, 2008). The lack of a relevant growing stage was attributed to the delay in the start of the new particle formation in urban environments with respect to the regional background, where new particle formation starts in the morning. This delay is due to the increase of the condensation sink (CS) during the morning and afternoon road traffic rush hours. Thus new particle formation takes place in the city only when primary pollutant concentrations are low, i.e. after the concentration of traffic primary pollutants has decreased at midday, and until the afternoon rush hour.

Two major types of new particle formation episodes in the urban area of Barcelona were identified by using four simultaneous Scanning Mobility Particle Sizer (SMPS) instruments into the city and in the regional background during the SAPUSS campaign (Dall'Osto et al., 2013). One occurs on a regional scale and hence it is simultaneously recorded at the regional background and urban sites, with the differences in growth and time extension described above. The other type starts in the city and affects the regional background in a delayed stage by the transport of the nuclei and/or the precursors. The time delay is attributed to the availability of chemical precursors in the city (Dall'Osto et al., 2013). These midday urban new particle formation episodes are favoured by the high insolation and the decrease of the CS caused by the dilution of pollutants due to reduced emissions, growth of the planetary boundary layer (PBL) and maximum sea breeze flow intensity. Furthermore, the sea breeze transports SO_2 from shipping emissions that may help to activate nucleation. Similarly to these urban new particle formation episodes, Setyan et al. (2014) reported that the occurrence of new particle formation events in the regional background of Sierra Nevada (US) was associated with the influence of the urban plumes of Sacramento.

Studies carried out in London at different heights suggested that during the upwards convective transport of urban air pollutants as the PBL grows, important volatilization processes occur affecting the external shells of semi-volatile organic matter of UFP (Dall'Osto et al., 2011; Harrison et al., 2012). These volatilized organic substances are very reactive and they may yield to the formation of a large amount of newly formed particulate matter (Robinson et al., 2007). In any case, studies on the urban vertical distribution of UFP are very scarce, especially those covering the whole PBL, given that special instrumentation is required to measure UFP concentrations using balloons, or other airborne settings, throughout the vertical profile of the PBL (Ahn and Eun, 2013). Wehner et al. (2007) designed a study with four ground sites in Leipzig, Germany, separated up to 50 km, and a tethered balloon equipped with a combination of two Condensation Particle Counters (CPC) with different lower size detection limits so that the particle number concentration between 5 and 10 nm was deduced. They found two different nucleation scenarios, namely (i) homogeneous case, during which new particle formation was measured at the different ground sites nearly in parallel with subsequent particle growth and the UFP were found to be well mixed within the entire PBL; and (ii) inhomogeneous case, during which new particle formation was observed at one to three ground sites irregularly and subsequent particle growth was often interrupted. In this latter case, the new particle formation was found to depend mainly on the incoming solar radiation, but other factors must account for the inhomogeneous occurrence of the event in the study area. Some other studies on the vertical distribution of aerosols with in-height and/or tethered balloon borne instrumentation exist but they do not include the UFP size range (e.g. Matsuki et al., 2005; Greenberg et al., 2009; Ferrero et al., 2010, 2011; Babu et al., 2011; Moroni et al., 2012, 2013; Hara et al., 2013, 2014) and therefore the new particle formation processes could not be investigated.

The vertical profiles of aerosol properties have been determined using different instrumentation. Thus, Ma and Yu (2014) measured the aerosol extinction up to 5 km and found that it is mainly located below 1 km, with larger percentages in winter seasons (62–79%) and smaller percentages in summer seasons (44–57%), and attribute these differences to the strength of vertical transport. Moreover, they link the aerosol extinction to secondary particles whereas that attributed to other aerosol species is relatively small and only limited near the surface. Fernández-Gálvez et al. (2013) combined ground and column measurements, finding that anthropogenic aerosol in the atmosphere in the absence of a well-developed mixing layer is not exponentially distributed with height, and concluded that estimates of columnar aerosol optical and microphysical properties from ground measurements are not straightforward.

The main objective of this study is to analyze how the new particle formation takes place over an urban Mediterranean environment, by investigating the vertical distribution of UFP throughout the PBL height (PBLH) and the free troposphere. To this end measurements were carried out using a tethered balloon equipped with miniaturized CPC and optical particle counters (OPC) (Lee et al., 2014), a microaethalometer, a rotating impactor, instrumentation to measure meteorological variables (wind speed and direction, temperature and relative humidity) and a GPS. Vertical profiles up to 975 m a.s.l. were studied from 13 to 15 May 2014 between 8:00 and 16:00 local time with different sampling strategies regarding the ascending and descending velocity and the time spent at maximum height. To support interpretations, simultaneous measurements of aerosol parameters (concentration, size and composition), gaseous pollutants ambient concentrations, and meteorological parameters were carried out at two ground monitoring stations (one urban and one regional background).

2. Methodology

2.1. Measurement area, sites, and schedule

The measurements were carried out in the Northeast of Spain, in the Western Mediterranean Basin (WMB). This area is characterized by mild winters, warm summers and prevalent clear sky conditions all year round. Solar radiation is thus intense, its maximum values ranging from 450 to 900 W/m^2 at midday. Precipitations are scarce and usually concentrated in spring and autumn. More characteristics of the WMB can be found in Millán et al. (2000). The city of Barcelona is geographically constrained by the coastal range of Collserola to the North and the Mediterranean Sea to the Southeast, thus being influenced by the sea-breeze dynamics. This results in a dominant Northwest wind component during the night and the development of sea breezes during the day. The diurnal breezes turn progressively from Southeast to Southwest and the gradually increasing wind speed reaches its maximum levels around noon when the boundary layer is fully developed (Pérez et al., 2004). Moreover, different meteorological scenarios can have an impact on the air pollution of the city, ranging from stagnant anticyclonic conditions, recirculation of air masses or Atlantic air mass advection to African dust outbreaks. The main source of atmospheric pollution in Barcelona is road traffic (e.g. Amato et al., 2009; Pérez et al., 2010; Reche et al., 2011), although emissions from industry, power generation, construction, domestic and residential sectors, and harbor activities are also relevant.

The balloon measurements location was situated in the city of Barcelona, in an open area from the Real Club de Polo de Barcelona. The balloon flights were carried out during 13 to 15 May 2014 according to the schedule in Table 1. Local time in Barcelona is GMT + 2. All reported times in this study correspond to local time. One flight up to 800 m a.s.l. from 8:29 to 10:34 was carried out on the 13 May. Two flights were performed on the 14 May, from 8:12 to 10:35 (up to 800 m a.s.l.) and from 12:01 to 15:52 (up to 975 m a.s.l.). On 15 May the measurements were carried out during an ascending flight from 8:00 to 9:30 up to 900 m a.s.l. and a short descending flight down to 770 m a.s.l. (until 10:28). Subsequently the measurements were carried out at a constant height (about 700 m a.s.l.) until 16:00.