Responses to the Comments of Referee #1

(1) The objectives set out for the work in the last paragraph in the introduction are rather unambitious and relate to the effect of the Paris megacity on the downwind areas, and the frequency and spatial characteristics of new particle formation events. To address such objectives fully would require measurements over at least a full year but these were in fact limited to campaigns of one month in summer and one in winter, and these are not set in the context of a long-term dataset so it is not known whether they are representative or not.

A complete year of size distribution measurements (including the two intensive campaigns discussed in the present paper) has been recently presented by Dos Santos et al. (2015). These measurements took place in one site in the center of Paris (LHVP station) from July 2009 to September 2010. During this year, the highest NPF frequency in Paris was observed during July 2009 (the summer campaign examined in this work) and the lowest during the winter (which includes the winter campaign in this work). Therefore this work focuses on two extreme NPF periods in Paris. During summer under clean conditions and peak NPF frequency and during winter under polluted conditions and minimal NPF frequency. These are now explained in the revised manuscript, placing the work in the context of a longer-term dataset as the reviewer suggested.

(2) The title refers to ultrafine particle sources but the reader learns only about NPF events and nothing about the other sources of particles. Either the title needs to change or the content needs to be enhanced if possible to throw light on other sources, although the design of the experiments is not good from this perspective.

We do agree with the point of the reviewer. The title of the paper has been changed to "In situ formation and spatial variability of particle number concentration in a European Megacity", which better describes the final scope of this paper.

(3) From the section on instrumentation and the list of instruments in Table 1, it is clear that ultrafine particle measurements were made with a substantial range of different instruments using at least two different methods of drying of the air stream. For some of the instruments, the drying method is not clear and it would be useful if these were added to Table 1. Given the substantial range of instruments and at least two drying methods, it would be essential to intercompare the CPCs with one another and the SMPS/DMPS/EAS instruments with one another. This is not reported and there are consequently question marks over the comparability of measurements by the different instruments. If an intercomparison was conducted, this needs to be included and a description given of how divergences in readings were accommodated in the data analysis.

The sampling conditions (dry/ambient) are now explicitly stated in Table 1. The different instruments were intercompared during both campaigns. At least one of the mobile laboratories visited each site for several hours (5-15 h) during each campaign. The summary of these comparisons is shown in a new figure in the supplementary material (Fig. S1). During summer, the differences in number concentration between the CPC on board the visiting mobile laboratory (MOSQITA) and the aerosol sizing instrument at each of the stationary sites did not exceed 10%. During winter the discrepancies were higher mainly due to the lower detection efficiency size limit of the MoLa CPC that was used for the intercomparisons. During both campaigns the number concentrations monitored onboard MoLa and MOSQUITA were also

compared for approximately 8 hours. The two instruments were found to agree during periods without nucleation. The comparison of the CPCs in the two mobile laboratories has been presented by von der Weiden-Reinmüller et al. (2014). A brief summary of the intercomparisons together with the corresponding references to previous work have been added in the revised paper.

(4) It has been noted by a number of authors that both particle number counts and particle size distributions in urban areas changed substantially with the introduction of zero sulphur motor fuels. This effect needs to be mentioned together with information on the sulphur content of motor fuels in the Paris region at the time of these experiments. This critically affects the particle size distribution and aerosol lifetime
Most (61%) of light duty vehicles in France during the period of the measurements were using diesel fuel with 10 ppm sulfur. As the reviewer has indicated both the sulfur content and the fuel type dictate vehicle emissions. This is now discussed in the revised manuscript.

(5) Section 3.1 deals with the estimation of condensation sinks, but the method by which these were estimated is not adequately described. Section 3.3 gives adequate detail on how the humidity-adjusted size distribution was calculated but this is only part of the method
A detailed description of the condensation sink estimation, including the corresponding equations, is presented in the revised manuscript.

(6) Figure 8 shows average size distributions for each season and site and these are briefly discussed on page 5676 going into 5677. Given that the paper, judging from the title, is concerned with the sources of ultrafine particles and that other workers have sought to elicit source information from number size distributions, this section is very disappointing and gives few if any insights into the factors giving rise to these size distributions. The quite substantial differences between summer and winter are not explained other than by an indication that similar behaviour has been observed elsewhere, and the inter-site differences are described but not explained.
The discussion based upon the size distributions has been expanded focusing on the sizes of the various modes and their strength. As mentioned above, the title of the paper has been changed to avoid confusion about its focus on the primary and secondary particle number sources and not on the individual primary sources.

(7) The discussion of new particle formation in Section 6 is one of the stronger parts of the paper but the critical omission is the measurement of sulphur dioxide concentrations. Are there no useful data available from anywhere within the domain of the experiments? Without this information, the discussion is very incomplete as the authors acknowledge at the end of page 5685.
Sulfur dioxide measurements were available at GOLF which was mostly downwind of Paris during the summer campaign. However, the low sulfur content of vehicle emissions and the lack of other major sulfur sources resulted in ambient sulfur dioxide concentrations that were below the detection limit (0.5 ppb) of the instrument used most of the time. As a result, there is little useful information in these measurements. This is now mentioned in the revised paper.

(8) Page 6598, line 1 – the spelling of authors' names is incorrect.

We checked the spelling of the authors' names in the Wang et al. (2010) reference in page 5698 and it is correct.

(9) Page 5710, legend to Figure 10, 3rd line – should read exponential decrease (not decease).
Corrected

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Response to the Comments of Referee #2

(1) The paper was an enjoyable read to start with, and well written. However it became evident that the sources of the NPF events were not going to be identified, as the paper title suggested.

We do agree with the point of the reviewer. The title of the paper has been changed to "In situ formation and spatial variability of particle number concentration in a European Megacity", which better describes the final scope of this paper. The scope and analysis of this work have not changed. These include

- analysis of NPF events within, downwind and upwind of Paris that suggest that the condensational sink was the dominant factor influencing the frequency of events in this Megacity.
- effect of the Paris emissions on particle number concentrations around the Megacity.

(2) The Paris plume itself was identified by concentrations of black carbon and increased particle numbers. I wonder whether non-Paris contributions of black carbon might affect this assumption – i.e. smoke from rural grass/forest fires in summer, or suburban/rural wood burning in winter?

We have examined satellite-based products for fire identification, including small fires. No biomass burning events, significant enough to be identified by the algorithm used (Randerson et al., 2012), were observed during the two campaigns. Thus during summer biomass burning was ruled out as a potential source of error. On the other hand, during winter areas outside of the Paris plume with increased black carbon levels were identified and omitted from the analysis. The black carbon source in these cases was residential biomass burning. The particle number concentrations in these areas were relatively low though. The potential interference of these sources would have a modest to small effect on our estimates regarding the evolution of the Paris aerosol number plume. A new paragraph has been added in the revised manuscript discussing the above point.

(3) The paper explained when new particle formation takes place and whether agreeable measurements were made at other sites but does not explain the process of formation nor what the particles are composed of. I would expect that an experiment designed to investigate ultrafine particle sources would have had an aerosol speciation instrument, such as an Aerosol Mass Spectrometer or an Aerosol Chemical Speciation Monitor available.

The MEGAPOLI measurements focused on the identification of particulate matter mass sources. There were three AMS units available in the three sites and a detailed analysis of their measurements can be found in the corresponding publications (Freutel et al., 2013; Crippa et al., 2013a; 2013b; 2013c). A synthesis of all the fine PM source attribution measurements has been provided by Beekmann et al. (2015). Unfortunately, all these refer to the fine PM (PM_1 and $PM_{2.5}$) mass concentration and not to the new particles. The mass of the new particles is a very small fraction of the total and the corresponding compositions can be very different. The new particle formation events took place during periods with relative rapid photochemistry so all secondary particle components increased at the same time. We have added some text in the revised paper discussing the above points.

(4) From the list of instrumentation used in Table 1, the only coincident trace gas measurements were taken on board the aircraft at approx 600 m in height. None of

these trace gases correlated with particle number. Why were there no ground measurements of trace gases? A brief look at papers within the MEGAPOLI special issue suggests there are more measurements available, indeed the section describing the MEGAPOLI field campaign in the introduction discusses other work done to identify sources of particulate matter, but then these same measurements don't seem to be used later on to help identify the sources of these ultrafine particles.
Table 1 presents a subset of the MEGAPOLI measurements that have been used in this work. There were several additional gas-phase measurements in the ground stations (see for example Michoud et al., 2012). These measurements (OH, ROx, NO, NO_2, O_3, CO, PAN, HONO, VOCs) did not provide any additional insights about the precursors of new particles formed. We have added in the text references to the papers providing detailed information about the gas-phase measurements that took place during the MEGAPOLI campaigns.

(5) Was any modeling done across the MEGAPOLI participants to try and answer these questions? The CHIMERE model is mentioned in the introduction section as being used to decide the routes of the mobile and aircraft platforms, but could have been used to model the Paris Plume. This would then have pointed to certain emission source groups being likely candidates for the different NPF events. Even better, a model incorporating aerosol number, size and composition would aid the story.
There have been a number of modeling efforts but all of them have focused so far on particle mass and not number. The Zhang et al. (2013) study using CHIMERE and the Couvidat et al. (2013) work investigated the sources of organic aerosol in Paris. Skyllakou et al. (2014) examined the contributions of local and regional sources to fine PM mass concentrations in Paris. However, sources that contribute significantly to particle mass may contribute little to particle number or vice versa depending on the corresponding size distributions. Extrapolating from the particle mass source attribution studies to particle number is dangerous. There have been no modeling studies yet focusing on both aerosol number and mass. We do agree with the reviewer that such studies together with the MEGAPOLI measurements could provide valuable insights. References to the MEGAPOLI modeling studies have been added to the revised paper.

(6) Please explain the comment "during winter the higher condensation sink...prevented particles from growing to sizes larger than 10 nm". I would expect that high condensation would lead to an increase in the particle size either directly or via coagulation. The only other explanation is that there was a high surface area already present which caused a plateau in the particle growth, but as there were no nucleation events in winter I don't understand where this high surface area originated from.
The reviewer is correct; there was a high surface area already present resulting in the high condensation sink. The sources of these particles included long range transport, biomass burning, transportation, cooking, etc. (Crippa et al., 2013a). These sources provided plenty of aerosol surface area. This is now explained in Section 5.1 of the revised manuscript.

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Responses to the Comments of Referee #3

(1) The scope of the paper appears to be a little narrower than promised by the title.
We do agree with the point of the reviewer. The title of the paper has been changed to "In situ formation and spatial variability of particle number concentration in a European Megacity", which better describes the final scope of this paper.

(2) The ambiguity in definition of ultrafine particles (with the air pollution community, policymakers and regulators referring to traffic-dominated Aitken mode particles finer than about 100 nm as ultrafine) could be addressed with modest modification to the title and short clear description of the scope of the current study. Such a definition section within the introduction would definitely benefit the special issue. Between the first and second paragraphs of the introduction (i.e. between the PM2.5 and NPF related sections) or between the third and fourth paragraphs (linking and contextualizing in situ emission and in situ formation contributions) might be appropriate places.
The title has been changed and it does not include anymore a reference to ultrafine particles. We do agree with the reviewer that it can be confusing. The paragraph describing the scope of this study has been slightly modified stressing that the periods under investigation correspond to the two extreme conditions (frequent new particle formation-clean conditions and infrequent new particle formation-polluted conditions) encountered in the Paris region.

(3) The companion paper in the special issue from the same group (Skyllakou et al., 2014) addressing the sources of "fine" particles, defined therein as PM2.5, carries out a more conventionally defined (though quite novel) source attribution study. Challenges to performing such a comparable source attribution for the ultrafines should be discussed
This is a good point. Source attribution of particle number concentrations is challenging because particle number is not conserved due to coagulation and the particle size distribution is modified due to condensation/evaporation, nucleation, and removal. There are a few efforts in the literature trying to estimate the sources of the particle number (Wåhlin et al., 2001; Houssein et al., 2004; Zhou et al., 2005; Chan and Mozurkiewich, 2007). One method that has been applied is Positive Matrix Factorization which unfortunately cannot account for new particle formation. In order to apply such methods periods of new particle formation should be omitted (Zhou et al., 2005). This corresponds to half of the Paris summer campaign dataset. There has been an effort by our team recently (Posner and Pandis, 2015) to perform such particle number source attribution based on the results of a Chemical Transport Model. This produced encouraging results for particles smaller than 100 nm, but had weaknesses for larger particles. We have added a paragraph in the revised manuscript discussing these issues.

(4) It is difficult to consider attribution of NPF by source if there is no simultaneous source attribution of condensation sink. The authors might like to expand on the outlook for resolving NPF mechanisms and sources in complex environments, with significant mixing of air masses from different sources at a range of scales.
Our hypothesis was that we would be able to explore the spatial variability of new particle formation in the complex environment in and around a Megacity to learn more about the corresponding mechanisms. While we did observe variability in space

(please Section 7 and Figure 2), we could not relate it to any of the measured species. This does show that there are opportunities in these complex environments, but additional measurements of candidate nucleating vapors are required. The condensational sink can be viewed as an obstacle to nucleation. For these urban environments the condensational sink correlates reasonable well with PM1 or PM2.5 and the source attribution of the corresponding mass concentrations can be used as a reasonable proxy. The source contributions to fine PM for the MEGAPOLI campaigns have been discussed in detail by Beekmann et al. (2015). A brief discussion of these points has been added to the revised manuscript.

(5) I am in some agreement that broader consideration of material other than number and size measurements would provide more insight. This may be possible by reference to other papers in the special issue
Additional references to the related source apportionment work during the same field studies have been added.

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Ultrafine particle sources and in-In situ formation and spatial variability of particle number concentration in a European Megacity

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Abstract

Ambient particle number size distributions were measured in Paris, France during summer (1 - 31 July 2009) and winter (15 January – 15 February 2010) at three fixed ground sites and using two mobile laboratories and one airplane. The campaigns were part of the MEGAPOLI project. New particle formation (NPF) was observed only during summer at approximately 50% of the campaign days, assisted by the low condensation sink (about $10.7\pm5.9\times10^{-3}$ s^{-1}). NPF events inside the Paris plume were also observed at 600 m altitude onboard an aircraft simultaneously with regional events identified on the ground.

Increased particle number concentrations were measured aloft also outside of the Paris plume at the same altitude, and were attributed to NPF. The Paris plume was identified, based on increased particle number and black carbon concentration, up to 200 km away from Paris center during summer. The number concentration of particles with diameter exceeding 2.5 nm measured on the surface at Paris center was on average $6.9\pm8.7\times10^4$ cm^{-3} and $12.1\pm8.6\times10^4$ cm^{-3} during summer and winter, respectively, and was found to decrease exponentially with distance from Paris. However, further than 30 km from the city center, the particle number concentration at the surface was similar during both campaigns. During summer one suburban site in the NE was not significantly affected by Paris emissions due to higher background number concentrations, while the particle number concentration at the second suburban site in the SW increased by a factor of three when it was downwind of Paris.

1. Introduction

Urban areas in the developed and developing world have been growing annually by 0.7% in population since 2005 and comprised approximately 54% of the total population of the planet in 2014 (United Nations, 2014). In this work, following the definition of the Organization for Economic Co-operation and Development (OECD), urban areas are defined as corresponding to a population density greater than 1500 inhabitants per km^2 (OECD, 2013). Several of these urban areas have increased in size to mega-centers, attracting more than 10 million inhabitants. This has led to an increasing demand for transportation, energy and industrial activity, which resulted in concentrated emission of gases and particulate matter (PM) impacting local air quality (Molina and Molina, 2004; Molina et al., 2004; Lawrence et al., 2007; Gurjar et al., 2008). Several epidemiological studies suggest that the risk of cancer, particularly lung cancer, is increased for people residing in areas affected by urban air pollution (Barbone et al., 1995; Beeson et al., 1998; Laden et al., 20012006; Nyberg et al., 2000; Pope et al., 2002; Nafstad et al., 2003). Pope et al. (2009) and Wang et al. (2008) showed that fine particles with diameter smaller than 2.5 μm ($PM_{2.5}$) are related to increased mortality.

Aerosol particles can change climate patterns and the hydrological cycle on regional and global scales (Chung et al., 2005; Lohmann and Feichter, 2005; IPCC, 2007). Submicrometer particles, down to 100 nm, are the most effective ones in scattering solar radiation. The uncertainties in the primary emission rates of these pollutants and in their

formation from gaseous precursors are still large. On a global scale new particle formation (NPF), that is nucleation of low volatility vapors and subsequent condensational growth to larger sizes, is the major reason for high particle number concentrations (Kulmala et al., 2004). The mechanism behind this major particle formation process is still not completely understood (Riccobono et al., 2014). This uncertainty has a direct impact on our understanding of the role of nucleated particles in climate change (Pierce and Adams, 2009). NPF is often a regional phenomenon covering areas of several hundred square kilometers (Vana et al., 2004; Stanier et al., 2004a; Komppula et al., 2006; Crumeyrolle et al., 2010) but it can be space-restricted when the source of one of the nucleating vapors is space limited, as it has been observed in coastal sites (Wen et al., 2006).

During the past decade a number of studies reported ambient particle number concentrations in urban areas. The measurement period spanned from a few months (Hering et al., 2007; Wang et al., 2010; Dunn et al., 2004; Baltensperger et al., 2002; McMurry et al., 2005), to one or more years (Woo et al., 2001; Alam et al., 2003; Shi, 2003; Wehner and Wiedensohler, 2003; Stanier et al., 2004b; Wehner et al., 2004; Wu et al., 2007; Rodriguez et al., 2005; Watson et al., 2006; Wåhlin, 2009). The majority of studies are based on observations from one or at most two stationary sites, assuming that these stations are representative of the area under investigation. Most of these studies have found higher concentrations during winter due to both increased emissions caused by higher energy demand, and lower boundary layer height. Also, typically a diurnal pattern has been found that shows peaks due to morning rush hour traffic during weekdays but not on weekends.

NPF has often been observed in urban areas (Woo et al., 2001; Baltensperger et al., 2002; Laakso et al., 2003; Tuch et al., 2003; Stanier et al., 2004a; Watson et al., 2006; Wu et al., 2007), but growth and nucleation rates are rarely reported in these studies (Birmili and Wiedensohler, 2000; McMurry, 2000; Shi et al., 2007; Wehner et al., 2007; Manninen et al., 2010).

During the "Megacities: Emissions, urban, regional and Global Atmospheric POLlution and climate effects, and Integrated tools for assessment and mitigation" (MEGAPOLI) project (Baklanov et al., 2010), measurements were conducted in and around the megacity of Paris. Gas and particulate phase measurements from three fixed ground sites, two mobile laboratories, and one airplane were collected for both summer 2009 and winter 2010. The residence time of the air mass over land was found to influence PM mass levels, with longer residence times leading to higher PM levels mass

concentrations (Freutel et al., 2013). As a result airAir masses from the Atlantic, which were dominating during the summer campaign, led to relatively clean conditions (Freutel et al., 2013; Freney et al., 2014). Cooking was identified as a significant local organic aerosol source within Paris during summer with vehicular traffic being second (Crippa et al., 2013b). During winter residential wood burning for residential purposes was found to be a major source of primary particulate matterorganic aerosol (Crippa et al., 2013a). During both MEGAPOLI campaigns, the contribution of primary transportation emissions to submicrometer organic aerosol (OA) was around 6% (Crippa et al., 2013b). In the year of the MEGAPOLI campaigns, 61% of the light duty vehicles in France were powered by diesel engines and 72% of the consumed fuel was diesel (World Bank, 2012). The sulfur content of diesel in France at that time was 10 ppm compared for example to 500 ppm in 1998. The sulfur content of fuel affects both the total particle emissions but also the shape of the corresponding aerosol distribution (Platt et al., 2013; Bermúdez et al., 2015).

Beekmann et al. (2015) have presented a synthesis of the MEGAPOLI PM mass source attribution efforts based on the corresponding field measurements. In parallel, several modeling efforts have been also conducted examining the contribution of regional sources to fine PM (Skyllakou et al., 2014) and investigating the organic aerosol sources in Paris (Couvidat et al., 2013; Zhang et al., 2013). All of these studies focused on PM mass concentration and not on particle number. The different size distributions of the aerosol emitted by different sources usually result in very different source contributions to particle number and mass (Zhou et al., 2004). There have been a number of studies that tried to quantify the particle number sources using available size distribution measurements (Wåhlin et al., 2001; Hussein et al., 2004; Zhou et al., 2004; Chan and Mozurkewich, 2007). However, the changes of these distributions due to new particle formation and growth or other dynamic changes seriously limit the applicability of techniques like Positive Matrix Factorization (PMF). Zhou et al. (2004) excluded the corresponding new particle formation periods from their dataset to overcome this problem.

In this work we focus on the particle number concentrations in Paris and its surroundings during both (summer and winter) campaigns. The effect of the Paris megacity on the downwind areas will beis assessed together with the spatial extent of its influence. The frequency and spatial characteristics of new particle formation events are also investigated.

2. Sampling sites

Month long campaigns were conducted in the Parisian region during summer (1 July to 31 July 2009) and winter (15 January to 15 February 2010). They included monitoring of the aerosol size distribution along with composition, coupled with gas phase and meteorological monitoring.

The city of Paris is an urbanized area covering about 3000 km^2 with 2.2 million inhabitants. The greater Paris area, called Île de France (IDF), is one of the largest metropolitan areas in Europe including more than 12 million inhabitants. The administrative boundaries of Paris and IDF are shown in Fig. 1 along with the population density map of the area.

Detailed aerosol particle measurements were conducted at an urban and two sub-urban sites (Fig. 1). The Site Instrumental de Recherche par Télédétection Atmosphérique (SIRTA, 48° 43' 5" N and 2° 12' 26" E) is located on the campus of Ecole Polytechnique (Palaiseau), 20 km southwest of Paris center in a semi-urban environment inside the campus of Ecole Polytechnique. This site is surrounded by highways at 3-6 km distance in all wind directions. Measurements in the Laboratoire d'Hygiène de la Ville de Paris (LHVP, 48° 49' 11" N and 2° 21' 35'' E), inside of Paris, were performed on a terraced roof 14 m above ground level and on the ground inside a research container. This site includes a station of the AIRPARIF air quality monitoring network and is representative of the Paris urban background air pollution (Sciare et al., 2010; Favez et al., 2007). Finally the sub-urban station at Golf de la Poudrerie (GOLF, 48° 56' 2'' N and 2° 32' 49'' E**)** was located 20 km northeast of Paris center near a golf course and a forested park.

Two mobile platforms, named "MoLa" (Mobile Laboratory) and "MOSQUITA" (Measurements Of Spatial QUantitative Immissions of Trace gases and Aerosols), were operated by the Max Planck Institute for Chemistry (Drewnick et al., 2012; von der Weiden-Reinmüller et al., 2014a) and the Paul Scherrer Institute (Bukowiecki et al., 2002; Weimer et al., 2009), respectively. The measurement path of both mobile platforms was decided based on forecasts of the chemical transport model CHIMERE (Rouil et al., 2009; Menut and Bessagnet, 2010; Menut et al., 2013). Three measurement strategies were employed during both campaigns: stationary, axial and cross sectional measurements (von der Weiden-Reinmüller et al., 2014a; 2014b). Cross sectional (mobile) measurements were carried out by maintaining approximately constant distance from Paris center while varying the cardinal directions, allowing distinction between background concentrations and Paris emissions. Axial (mobile) measurements were conducted by maintaining approximately the same cardinal direction while varying the distance with respect to Paris

center, thus monitoring the evolution of the plume. Stationary measurements were conducted when the direction of the Paris emissions, based on the CHIMERE model, were not stable enough to allow cross sectional or axial measurements. Stationary measurements were conducted only by MoLa either downwind of Paris, or upwind to assess background aerosol loadings.

The airborne measurements were performed by an ATR-42 and a Piper Aztec aircraft during summer and winter, respectively, operated by the French Service des Avions Français Instrumentés pour la Recherche en Environnement (SAFIRE). Each flight included a circle around IDF followed by crossing the expected Paris plume multiple times, at a constant altitude of 600 and 500 m above sea level for the summer and winter campaigns, respectively. During July 1 the flight path was kept at a constant altitude of approximately 800 m. Flights were performed on 11 out of the 31 days of the summer campaign. Fig. 2 shows the flight patterns and sampling days of the ATR-42 during summer. Flight days were selected based on CHIMERE predictions. Higher PM concentration days were favored, thus the observed aerosol properties are expected to be biased toward more polluted conditions. During winter two flights per sampling day were conducted for four days (January 27 and 31, February 14 and 15). The first flight included a survey of the aerosol properties around IDF and the second monitored the Paris plume, following a flight path similar to the summer one.

2.1 Instrumentation

The MEGAPOLI project focused on the properties of ambient aerosol, including both mass and number concentration measurements. This work examines the particle number concentration N during both MEGAPOLI campaigns; the instruments and measurements relevant for this purpose are summarized in Table 1. A number of additional measurements of concentrations of gas-phase pollutants, radicals, etc., were conducted during the campaigns (Michoud et al., 2012), but are not used in the present work because they did not provide any additional insights.

At SIRTA, threetwo instruments were used to monitor the ambient particle number distribution. A Scanning Mobility Particle Sizer (SMPS; TSI Model 3936) sampled aerosol particles from 10 to 500 nm in diameter through an inlet located approximately at 4 m above ground. The particles were actively dried using a Nafion dryer. An Air Ion

Spectrometer (AIS; Mirme et al., 2007) monitored the size distribution of ambient (not dried) positive and negative air ions of mobility diameters ranging from 0.8 to 40 nm. To minimize particle losses the sampling line length of the AIS was 30 cm. A Differential Mobility Particle Sizer (DMPS, Aalto et al., 2001) also monitored, close to the AIS, ambient number size distributions ranging from 6 to 800 nm during summer. At LHVP, the sampling inlet was located 6 m above ground and the aerosol sample was dried using a diffusion dryer as described in Tuch et al. (2009) before entering a mobility particle size spectrometer TROPOS-type TDMPS (Twin Differential Mobility Particle Sizer; Birmili et al., 1999), which monitored the aerosol size distribution from 3 to 630 nm. At the same site, an Air Ion Spectrometer (AIS; Mirme et al., 2007) monitored the size distribution of ambient (not dried) positive and negative air ions of mobility diameters ranging from 0.8 to 40 nm. To minimize particle losses the sampling line length of the AIS was 30 cm. At GOLF, the particle size distribution between 5 nm and 1 µm was monitored with an Electrical Aerosol Spectrometer (EAS, Airel Ltd.) and sampling was conducted 8 m above ground. Because the three aerosol size distribution instruments (SMPS, TDMPS, EAS) used for the stationary ground measurements during both campaigns overlaps between 10 nm and 500 nm (mobility diameter), our analysis will focus on this size range, denoted as $N_{10\text{-}500}$.

MoLa, which was based at GOLF, monitored the total particle number concentration via an Ultrafine Water Condensation Particle Counter (UWCPC, TSI Model 3786) with 50% detection efficiency at 2.5 nm, which will be denoted as $N_{2.5}$. The aerosol inlet during stationary measurements was located at approximately the same height as the stationary measurements at GOLF (8 m above ground). During mobile measurements, sampling occurred at about 2.4 m above ground level. MOSQUITA monitored the total particle number concentration via a butanol-based Condensation Particle Counter (CPC; TSI Model 3010, 50% detection efficiency at 10 nm) during summer, further denoted as N_{10}, and via an Ultra High Sensitivity Aerosol Spectrometer (UHSAS; DMT Model A) during winter. The UHSAS monitored the size distribution, with respect to the optical diameter, ranging from 60 nm to 1 µm.

On-board the METEO-FRANCE aircraft (ATR-42), aerosols were sampled, under dry conditions, through the community aerosol inlet (Gomes et al., 2014) and delivered to a comprehensive suite of aerosol instruments. This isokinetic and isoaxial inlet is based on the University of Hawaii shrouded solid diffuser designed by A. Clarke and had been modified by Meteo France (McNaughton et al., 2007). Particle number concentration was

monitored directly during summer and winter flights using a CPC with 10 nm (TSI Model 3010) and 2.5 nm (TSI Model 3025) lower cutoff, respectively. Because the CPCs used during the summer and winter campaigns had different lower detection limits, the corresponding number concentrations will be denoted as N_{10} and $N_{2.5}$, respectively.

In order to quantify potential differences between instruments, at least one of the mobile laboratories visited each site for 5-15 hours during each campaign. During summer, the differences in number concentration between the CPC on board the visiting mobile laboratory (MOSQUITA) and the aerosol sizing instrument at each of the stationary sites did not exceed 10% (Figure S1 in the Supplementary Information). The CPC on board MOSQUITA had a detection size limit equal to approximately 10 nm. During winter the MoLa CPC, with a lower detection size limit of 2.5 nm, was employed for the intercomparisons. In this case, the differences were higher and equal to 30%, 18%, and 19% at SIRTA, LHVP, and GOLF, respectively. Taking into account that particles below 10 nm were typically present at SIRTA during winter the corresponding discrepancy can be partially explained by the different detection limits of the two instruments (10 nm for the SMPS at SIRTA and 2.5 nm for the MoLa CPC). During both campaigns the number concentrations monitored onboard MoLa and MOSQUITA were also compared for approximately 8 hours. The two instruments were found to agree when the concentrations of the nucleation mode particles were moderate or low. This is expected due to their different size detection limits. The results of this intercomparison have been presented by von der Weiden-Reinmüller et al. (2014a).

3. Methods

3.1 Particle formation event categorization

Particle formation events have been categorized in the past based on the concentration of 1.6 – 7.5 nm air ions (Hiirsiko et al., 2007; Vana et al., 2008) and on the concentration of total ambient particles below 25 nm (Stanier et al., 2004a; Dal Maso et al., 2005). At SIRTA LHVP both air ions and ambient particles were measured and therefore we used two classification schemes, one based solely on ambient particles following Dal Maso et al. (2005) and one that includes air ions, following Hirsikko et al. (2007). In both cases, the observation period was divided into particle formation event days, non-event days and undefined days. In general, a day is classified as event day if a nucleation mode (particles with sizes smaller than 10 nm) is present for several hours and grows

continuously during the course of the day. If no traces of a nucleation mode are seen, a day is classified as a non-event day. Days that did not clearly belong to either of the aforementioned categories were classified as undefined. Examples of event, undefined and non-event days are shown in Figs. 3, 4 and 5, respectively.

During July 12, a nucleation mode appeared at 14:00 LST (local standard time) simultaneously at all ground sites (Fig. 3). During this cloudy day, nucleation was observed approximately one hour after the solar intensity increased by a factor of three (from 300 to 1070 W m^{-2}). This day was consequently classified as event day. During July 10, an increase in the number concentration of particles above 10 nm in diameter was measured simultaneously at LHVP and SIRTA at 14:00 LST (Fig. 4). It was unclear whether the mode also appeared at GOLF due to interferences by local sources. Particle growth was not continuous and the mode disappeared abruptly after approximately three hours, even though the direction of the wind did not change at this time. At SIRTALHVP air ion bursts in the size range between 1.6 – 7.5 nm did not follow a distinct pattern but were random. As a result it was unclear whether NPF occurred and the day was classified as undefined for all sites. During July 29, no nucleation event was observed, and the day was consequently classified as non-event day. During this day, the condensation sink (CS) was rather high ($9.0\pm1.7\times10^{-3}$ s^{-1}, $20.3\pm9.7\times10^{-3}$ s^{-1} and $14.4\pm4.1\times10^{-3}$ s^{-1} at SIRTA, LHVP and GOLF respectively) from 08:00 to 16:00 LST, when NPF was expected to occur. These sink values were above the summer average for all sites (see Section 3.3) and contributed to the lack of a nucleation mode at all sites (Fig. 5).

3.2 Duration of nucleation events

The duration of nucleation events at SIRTALHVP was calculated based on AIS measurements following the procedure described by Hirsikko et al. (2005) and Pikridas et al. (2012). In brief, a normal distribution was fitted to the time series of concentration of air ions with diameters between 2 and 5 nm. The beginning of the event was determined by the initial increase of the air ion concentration (assuming a stable air ion concentration before the event) and the end by the peak of the normal distribution. A decrease of the number concentration implies that the rate of particle production is lower than the combined rates of coagulation and particle growth to diameters greater than 5 nm, or that the air mass is getting diluted; it does not necessarily imply that the rate of production is zero. Our calculated event-end is thus a lower bound estimate.

3.3 Condensation sink

The condensation sink (CS) is defined as the condensational loss rate constant of vapors (Kulmala et al., 2001; Dal Maso et al., 2002). The CS values were calculated based on the aerosol number size distribution using:

$$CS = 2\pi D \int_0^\infty D_p \beta_m(D_p) n(D_p) dD_p = 2\pi D \sum_i D_{pi} \beta_{mi} N_i \quad (1)$$

where D is the diffusion coefficient of the condensing vapor, D_{pi} is the diameter and N_i the particle number concentration in size class. The term β_{mi} corresponds to the transition regime correction factor for the size class i, which was calculated based on Fuchs and Sutugin (1971). The properties of the condensable vapors are assumed to be similar to those of sulfuric acid, without accounting for hydration, leading to an upper limit estimate. If the aerosol sample was dried prior to the measurement, the diameter reduction due to water loss was estimated using the Extended Aerosol Inorganic Model II (E-AIM, http://www.aim.env.uea.ac.uk/_aim/aim.php; Carslaw et al., 1995; Clegg et al., 1998; Massucci et al., 1999). The hourly averaged inorganic concentrations for sulfate, ammonium and nitrate measured by the aerosol mass spectrometer (AMS; Jayne et al., 2000; Jimenez et al., 2003) and ambient RH measured at each site, were used as inputs to the model, neglecting any contribution of organics to the aerosol water content. The volume growth factor was determined following the method of Engelhart et al. (2011) which assumes that all submicrometer particles grow similarly by neglecting the Kelvin effects. The diameter growth factor was calculated as the cubic root of the volume growth factor and was applied to the whole particle distribution.

3.4 Mobile measurements

Due to the high frequency of local contamination events, mobile data was post-processed by examining video footage recorded at the driver's cabin of the mobile laboratory, based on Drewnick et al. (2012). Measurement periods were omitted from analysis if traffic was identified less than 150 m from the platform; if human activities (e.g. cooking, heating) were spotted; when driving at low speed caused a possible contamination by the vehicle's own exhaust; and when travelling inside tunnels. In order to reduce the amount of contaminated data major roads were avoided. More details concerning the conditioning of mobile measurements presented in this study can be found in von der Weiden-Reinmüller et al. (2014a). Further analysis of the mobile dataset was conducted based on results from

the particle dispersion model FLEXPART performed in forward mode (Stohl et al., 2005). Particles were released from an area whose borders were determined by the population density map presented on Fig. 1 and included Paris. Based on these modeling results and the respective measurement tracks, mobile measurements were attributed as influenced or not by Paris emissions.

4. Meteorology

During summer, the lowest ambient temperature was 12°C, observed at SIRTA and GOLF, and the highest of 33°C was measured at LHVP. Campaign average temperatures during summer were 19.7, 21.1 and 18.7 °C at GOLF, LHVP and SIRTA, respectively. In general, the temperature was higher inside the city center by 1°C at least, compared to the suburban sites. Diurnal variations of RH (ranging from 35% to 90%) and temperature were similar at all sites during summer. There were several cloudy periods and cloud coverage was geographically dependent. During summer at all ground sites, solar radiation reached a maximum of 900 W m^{-2} while the presence of clouds could reduce it by a factor of three. Precipitation as monitored at SIRTA occurred on 8 of the 31 days of the campaign (July 8, 16-18, 22, 23, 27 and 30). Maximum observed precipitation rate during the summer campaign was 0.5 mm min^{-1}; however it rarely exceeded 0.1 mm min^{-1}.

During winter the campaign average ambient temperatures were 2.6, 3.3 and 1.2 °C at GOLF, LHVP, and SIRTA, respectively. RH varied from 40% to 90% and exceeded 95% on several occasions at all sites. Hourly average global solar irradiance did not exceed 400 W m^{-2} during the winter campaign and did not exceed 100 W m^{-2} on 14 of the 32 days of observations. Precipitation occurred during winter on two thirdthirds (21 of 32 days) of the campaign days and the average precipitation rate was approximately 0.15 mm min^{-1} .

Figure 6 shows the wind direction distribution at all sites, for each campaign. Wind direction, measured at 10 m above ground, during summer was predominantly SW at LHVP and GOLF and W at SIRTA (Fig. 6) indicating that air masses often crossed the city center before reaching GOLF and that SIRTA was mostly upwind of the city. During winter wind directions were more variable with the wind equally coming from both NE and W (Fig 6). During the winter campaign SIRTA was more often than GOLF influenced by air masses that crossed the urban area before reaching the site.

5 Particle number concentrations and size distributions

5.1 Stationary measurements

Average number concentrations of particles with diameters between 10 and 500 nm ($N_{10\text{-}500}$), for all ground sites during both campaigns, are summarized in Table 2. On average, the $N_{10\text{-}500}$ concentrations during winter were higher than during summer by a factor of two at SIRTA and GOLF, and by 35% at LHVP. The highest hourly averaged concentrations were observed at GOLF (54.1×10^3 cm^{-3} and 72.2×10^3 cm^{-3} during summer and winter, respectively) followed by the urban center station LHVP (34.4×10^3 cm^{-3} and 45.5×10^3 cm^{-3} during summer and winter, respectively). The average ratio of the aerosol number concentration observed at LHVP to the one observed at GOLF was 0.86 and 0.62 during summer and winter, respectively. The average ratio of the aerosol number concentration observed at LHVP to the one observed at SIRTA was 2.1 and 1.5 during summer and winter, respectively.

The particle number concentration at all sites followed the same diurnal pattern during both seasons (Fig. 7). Regardless of site and season, minimum concentrations were observed between 3:00 and 4:00 LST, when anthropogenic activities are expected to be minimal. The concentration exhibited two maxima: during morning traffic hours, peaking between 7:00 and 10:00 LST, and during nighttime, between 8:00 and 9:00 LST. These diurnal profiles are typical of urban areas (Ruuskanen et al., 2001; Woo et al., 2001; Watson et al., 2006).) and can be explained based on the evolution of the mixing layer (Bukowiecki et al., 2005). In the afternoon atmospheric mixing reaches its maximum and primary pollutant concentrations decrease due to dilution. The mixing height remains fairly constant till nighttime when it decreases resulting in increasing primary pollutant levels. Boundary layer measurements using a Cloud and Aerosol Micro Lidar (Cimel model CE-370) at 355 nm that were performed at SIRTA support this explanation. The magnitude and time of the peaks varied depending on site and season. By comparing these maxima, which correspond to the peak of anthropogenic activity, against the minimum of the diurnal cycle, a rough estimate of the $N_{10\text{-}500}$ anthropogenic contribution can be made for each site. During summer the increase was 84%, 79%, and 21% at GOLF, LHVP, and SIRTA respectively, and during winter and 153%, 133% and 141%.

Average distributions for each season and site are shown in Fig. 8. During summer, particles with diameter ranging from 30 to 100 nm dominated the $N_{10\text{-}500}$ at SIRTA, accounting on average for 53%, followed by particles with diameters ranging from 10 to 30 nm which accounted for 30% (Fig. 8). Similar behavior was observed at LHVP during summer, where particles with diameter ranging from 30 to 100 nm accounted for 47% and particles with diameters ranging from 10 to 30 nm for 40% of the $N_{10\text{-}500}$. However, $N_{10\text{-}500}$

409 measured at GOLF was dominated by particles with diameter ranging from 10 to 30 nm,
410 which accounted for 50% of the $N_{10\text{-}500}$, followed by particles with diameter ranging from
411 30 to 100 nm that accounted for 42%.
412 During winter the contribution of particles with diameter from 10 to 30 nm to $N_{10\text{-}500}$
413 was almost equal to that from particles with diameters 30 to 100 nm at SIRTA (42% and
414 39%, respectively) and LHVP (44% and 40%, respectively). At GOLF the contribution of
415 particles with diameters between 10 to 30 nm increased even further (compared to
416 summer) reaching 56%, and the contribution of particles with diameters between 30 to 100
417 nm decreased to 34%. These differences are due to the shift of the Aitken mode of the
418 distributions to lower sizes during the winter.Average size distributions for each site are
419 shown in Fig. 8, along with the corresponding lognormal modes. During summer, an
420 Aitken mode centered approximately at 35 nm was dominating the number distributions at
421 LHVP and SIRTA. Nucleated particles grew to approximately this size during summer
422 (see Fig. 3 and 4) and could be identified for several hours after each event. The average
423 number size distribution in LHVP and SIRTA usually had two more modes centered at 15
424 and 115 nm respectively. The summertime number distribution at GOLF was characterized
425 by two modes centered at approximately 15 and 80 nm. Unlike SIRTA and LHVP the 15
426 nm mode dominated the aerosol number distribution at GOLF.
427 During winter the contribution of particles with diameter from 10 to 30 nm to $N_{10\text{-}500}$
428 was almost equal to that from particles with diameters 30 to 100 nm at SIRTA (42% and
429 39%, respectively) and LHVP (44% and 40%, respectively). At GOLF the contribution of
430 particles with diameters between 10 to 30 nm increased even further (compared to
431 summer) reaching 56%, and the contribution of particles with diameters between 30 and
432 100 nm decreased to 34%. The average size distribution, shown in Fig. 8, indicates a
433 dominating mode centered below 20 nm at all sites and a smaller second mode at 60, 80,
434 and 50 nm at SIRTA, LHVP and GOLF, respectively. Similar behaviorshifts of the aerosol
435 distribution to lower sizes during winter has been observed elsewhere (Bukowiecki et al.,
436 2003) where an inverse temperature dependence of the particle number concentration was
437 reported. Particles larger than 100 nm accounted for less than 20% of $N_{10\text{-}500}$ during both
438 campaigns at all sites.
439 Taking into account the location of each site, the contribution of small particles
440 (diameters 10-30 nm) to $N_{10\text{-}500}$ increases when moving from the SW (SIRTA) to the NE of
441 Paris (GOLF). Consequently, the contribution of particles with sizes 30-100 nm to the $N_{10\text{-}500}$ exhibits a decreasing (opposite) trend from the SW to the NE of Paris. Both trends were

observed during both seasons and indicate a persistent source of particles with diameters smaller than 30 nm NE of Paris, where GOLF was located. This conclusion is further supported by mobile measurements (Section 5.3) that showed that the background $N_{2.5}$ was relatively stable in the area further than GOLF during summer.

5.2 Impact of Paris on its surroundings

To investigate the impact of the emissions from the city center on number concentrations at the two satellite sites (GOLF, LHVP) the measurements were separated with respect to wind direction, excluding periods when the wind speed was below 1 m s^{-1} (Fig. 9). Taking into account that the area is relatively flat, it was assumed that the urban center influences each of the satellite sites at certain wind directions (215±30º and 65±30º for GOLF and SIRTA, respectively), noted with red on Fig. 9. This analysis is complicated by the variability in aerosol load due to air mass origin difference. During most of the summer campaign clean air masses from the Atlantic were reaching Paris (Freutel et al., 2013). Air masses of different origin, which accounted for only two consequent days during the summer campaign were omitted to minimize any discrepancy. During winter air mass origin was more variable and a common background could not be ensured, limiting this analysis only to the summer campaign.

During summer, the highest $N_{10\text{-}500}$ measured at SIRTA was observed when the air masses crossed the city center ($9.8\pm3.5\times10^3$ cm^{-3} and the lowest when the wind originated from the opposite direction ($4.2\pm2.3\times10^3$ cm^{-3}) considered later on as the background concentration. The urban emissions led thus to an increase of the number concentration by a factor of two at SIRTA. On the contrary, at GOLF the $N_{10\text{-}500}$ was not clearly affected by the wind direction during July 2009. $N_{10\text{-}500}$ measurements at GOLF were higher than at SIRTA, located at the same distance from Paris but on the opposite direction, by a factor of three when either site was not influenced by Paris. These results do not imply that Paris did not affect its surroundings during summer, but rather that the effect of the city was not large enough to be observed due to higher background concentrations at the GOLF site in the NE of Paris with respect to those at the SIRTA site in the SW. Mobile measurements that covered mainly the N-NE area with respect to Paris support this result (see Section 5.3). The possibility that these observations were due to temperature changes (Bukowiecki et al., 2003) was also investigated. However, no clear dependence between temperature

and $N_{10\text{-}500}$ was established. As an example, at SIRTA the lowest temperatures (around 17 °C on average) were observed both when air masses wherewere influenced by Paris and when the wind came from the opposite direction.

On July 21, MoLa performed stationary measurements 38 km north of Paris, which is almost twice the distance of each of the stationary sites (20 km) from the city center. Initially, air masses reaching MoLa were influenced by Paris emissions, based on FLEXPART simulations, and $N_{2.5}$ was equal to 14.1×10^3 cm^{-3}. However, the wind direction shifted while sampling and the $N_{2.5}$ decreased by 40% reaching approximately 8.5×10^3 cm^{-3}.

5.3 Spatial evolution of particle numbers in Paris and its surroundings

The majority of mobile measurements were conducted downwind of Paris in order to characterize its effect on its surroundings (von der Weiden-Reinmüller et al., 2014a; 2014b). These measurements were conducted in different distances from the center of Paris, under various meteorological conditions, different air mass origin (marine, continental) and were affected by the diurnal pattern (Fig. 7) of Paris emissions. The mobile measurements were further affected by wind direction shifts which resulted in monitoring of background concentrations instead of Paris emissions.

Paris emission measurements were identified during data analysis using FLEXPART in forward mode (Section 3.4). During summer, marine air masses were predominantly resulting in a relatively stable and low PM background. During winter air mass origin was not as stable as during summer, yet Paris emissions were also higher, thus facilitating the analysis. Variations in the number concentration due to meteorology effects or Paris emissions fluctuations can be dealt with by examining short case-study periods when these variables are relatively stable. However because such periods span a few hours only, the measurement sample is small. If measurements throughout each campaign are considered the sample size is satisfactory, yet it reflects the different conditions mentioned above. In this work both approaches were considered and are presented to quantify the behavior of the Paris plume downwind of the city.

Mobile measurements were separated, based on location, into concentric rings with borders at 0.15, 0.25, 0.4, 0.6, 0.8 and 1° (16.7, 27.8, 44.4, 66.7, 88.9, and 111.1 km) radius centered at kilometer zero of Paris (the official Paris center) as shown in Fig. 1. The first ring includes Paris and highly populated areas surrounding it, while the second one includes the greater Paris area where the two stationary sites (GOLF, SIRTA) are located.

During summer, when SW winds were predominant, the majority of the mobile measurements took place N-NE of Paris. The $N_{2.5}$ decreased exponentially with distance reaching 1.3±1.6×10^4 cm^{-3} approximately 100 km away from Paris center (Fig. 10), which is not statistically different at the 95% confidence interval from the average background (not influenced by Paris emissions) concentration (1.4±1.6×10^4 cm^{-3}) measured during summer upwind at distances greater than 30 km from the city center by MoLa. However, at distances shorter than 30 km, where GOLF is located, the background $N_{2.5}$ was almost twice as large (2.5±1.1×10^4 cm^{-3}) indicating a significant regional number source affecting this area. During 13 July 2009, axial measurements with respect to Paris were performed under relatively stable meteorological conditions and the results, shown as black dots in Fig. 10, are in good agreement with the campaign average values, following the same exponential decrease. Similar behavior in that area was observed for other pollutants during the same period (von der Weiden-Reinmüller et al., 2014b).

During winter, $N_{2.5}$ exhibited an exponential decrease with increasing distance from Paris center similar to summer. However, at the center $N_{2.5}$ was 75% higher than during summer. This difference was diminished in the Paris suburbs (second bar in Fig. 10), reaching 20%. At distances greater than 30 km from the Paris center, no statistical difference at the 95% confidence interval between $N_{2.5}$ measured during summer and winter was observed. Measured $N_{2.5}$ further than 70 km away from Paris remained stable (≈1.4±1.9×10^4) and was not statistically different from the background $N_{2.5}$ concentrations measured during winter (1.1±1.4×10^4 cm^{-3}) or summer (1.4±1.6×10^4 cm^{-3}). During 19 January 2010, axial measurements were performed and the results (shown as green triangles in Fig. 10) are also in good agreement with the winter campaign averages.

6. New particle formation at ground level

A summary of the particle formation categorization for both campaigns can be found in Fig. 11. During the summer campaign air ion bursts (of both polarities) for particles of sizes between 2 and5 nm were picked up by the AIS at SIRTALHVP on a daily basis (Fig. 11) with the exception of July 29. Concentrations of negatively charged particles between 2 and 10 nm were higher by one order of magnitude compared to positively charged. In Fig. 11 we present the NPF categorization based on the negative ions which provided a more sensitive way of identifying nucleation events.

During the summer campaign we observed 14 events at SIRTA, 14 at LHVP and 7 at GOLF based on SMPS, DMPS and EAS measurements, respectively. When NPF was identified at SIRTA it also took place at the city center (Fig. 11) with one exception (July 7). Due to technical issues of the DMPS, data for five days are not available at the LHVP site. Nucleation events, if identified at two or more of the ground sites, always occurred practically simultaneously (<10 min difference). $N_{10\text{-}500}$ typically doubled at GOLF due to NPF. At LHVP, an increase of $N_{10\text{-}500}$ ranging between 50% and 150% was observed due to NPF. The greatest increase in $N_{10\text{-}500}$, often exceeding 100%, due to NPF was observed at SIRTA.

The highest particle growth rate (17.6 nm h^{-1}), based on SMPS measurements, was observed at SIRTA on July 4 during a regional event observed at all ground sites while the lowest growth rate (1.6 nm h^{-1}) was observed on July 15 at LHVP, where typically lower daily growth rates compared to the two satellite sites were observed. The average growth rates were 6.1 ± 1.8 nm h^{-1}, 4.6±1.9 nm h^{-1} and 5.5±4.1 nm h^{-1}, at GOLF, LHVP and SIRTA, respectively, during the summer campaign (Table 2). Growth rates for events that occurred on all sites on the same day were 5.9± 2.4 nm h^{-1}, 4.5±2.0 nm h^{-1} and 8.3±5.6 nm h^{-1}, at GOLF, LHVP and SIRTA, respectively.

During July 28 nocturnal particle formation was observed at SIRTALHVP, which was identified by an increase of the ion number concentration within the 1.2–1.7 nm size range. An apparent growth of cluster ions to larger diameters than the upper limit of the preexisting ion pool was evident but air ions did not grow above 2 nm. Nocturnal cluster growth has been observed in remote areas (Junninen et al., 2008; Kalivitis et al., 2012; Hirsikko et al., 2012) and has been linked to the presence of monoterpenes (Ortega et al., 2012). Sulfuric acid generation due to nighttime oxidation processes has also been observed before (Mauldin et al., 2003).

The CS during the summer campaign for all sites is shown in Fig. S1S2 of the Supplementary Information, where event and undefined days are marked with blue and light blue labels, respectively. During summer the CS was half the value than in winter at GOLF (11.7±11.6×$10^{-3}$$s^{-1}$ in summer compared to 21.5±14.4×$10^{-3}$$s^{-1}$ in winter) and SIRTA (5.7±3.5×$10^{-3}$$s^{-1}$ compared to 12.3±6.8×$10^{-3}$$s^{-1}$) and 30% lower at LHVP (12.8±7.5×$10^{-3}$$s^{-1}$ compared to 17.0±8.6×$10^{-3}$$s^{-1}$). During summer at SIRTA and LHVP, NPF events occurred when the CS was lower than the seasonal average by 45% and 25%, respectively. Undefined events at both sites were associated with CS similar to the seasonal average value and non-event days with 25-30% higher CS compared to the seasonal average. In

winter, the high CS values in conjunction with the low solar intensity (see Section 4) most likely prevented nanoparticle growth and resulted in only five events without significant growth, identified only by the AIS at SIRTALHVP.

The solar intensity influence on NPF event occurrence was evident at SIRTA and LHVP. During NPF events at these two sites solar intensity was on average 30% and 20% higher, respectively, compared to non-event days. At GOLF, solar intensity during non-event days was found to be higher by 8% compared to actual event periods.

At GOLF, seven NPF events were identified, corresponding to a monthly frequency of 23%. The event frequency difference between GOLF and the other two ground stations was partially due to a higher frequency (23%) of undefined days (Fig. 11) caused by interferences of nearby traffic. When no event was identified at all sites the CS at GOLF was double ($14.7\pm4.5\times10^{-3}$ s^{-1}) compared to event days ($7.3\pm0.8\times10^{-3}$ s^{-1}), indicating that, similarly to the other sites, the CS was contributing to the inhibition of NPF occurrence. On several occasions (July 2, 6, 8, 23, and 28), NPF events were identified at LHVP and SIRTA (on July 8 it was not clear if NPF at SIRTA occurred) but not at GOLF (Suppl. Fig. S2S3). During these days CS values at GOLF were similar to event days and lower by 30% compared to the campaign average, indicating that at least the CS was not suppressing NPF. On two occasions (July 6 and 8) the observations show a continuous mode below 30 nm, either due to electrometer noise or local interferences, which prevented identification of NPF. Both days were listed as non-event days but NPF may have occurred. During July 2, a nucleation mode was observed at LHVP for more than an hour but nucleated particles did not grow above 20 nm (Class II events based on Dal Maso et al., 2005). During the same time, an air ion burst between 2 and 5 nm particle diameter was picked up by the AIS at SIRTA (the same site, but at GOLF the nucleation mode was not observed. The size distribution of particles above 40 nmat SIRTA was not monitored), but at GOLF the nucleation mode was not observed. It is uncertain if nucleation occurred and ions did not grow to detectable size, thus this day was listed as non-event. On July 23 NPF was identified at SIRTA, but not at LHVP only the size distribution below 40 nm was monitored by AIS, due to technical issues. Air masses crossed SIRTA before reaching GOLF and a fresh Aitken mode appeared at GOLF three hours later. Wind direction was constant during that period and the lag was consistent with the time needed for an air mass to travel between the two sites at the observed wind speeds (3 m s^{-1}). Similarly to July 23, on July 28 an NPF event was identified at SIRTA and LHVP, while at GOLF a new Aitken mode appeared after approximately three hours. From all this, it can be concluded that the

612 event frequency difference between GOLF and the other two sites can be explained to a
613 large extent by local interferences and uncertainty in identifying nucleation events.
614 Inhomogeneities with respect to the extent of NPF between locations a few tens of
615 kilometers away, similar to this work, have been reported before (Wehner et al., 2007) and
616 were attributed to cloud cover in combination with a boundary layer evolution scheme that
617 allowed such behavior. However, in the cases investigated in this work, cloud cover did
618 not appear to dictate non-event days at GOLF. Additionally, the beginning of events at all
619 sites always coincided, unlike the cases reported by Wehner et al. (2007). Despite these
620 differences, that work also noted the importance of CS in urban areas.
621
622 **7. Airborne Measurements**
623 Airborne measurements of N_{10} during summer and winter showed increased number
624 concentrations downwind of Paris accompanied by increases in light absorption measured
625 by the PSAP (Fig. 12). These results were attributed to PM emissions of Paris and are
626 referred henceforth to as the "Paris plume". This plume identification method assumes that
627 the only black carbon source in the area under investigation is the greater Paris region.
628 However, local sources of black carbon, such as wildfires during summer or domestic
629 heating during winter could interfere. To investigate the validity of our assumption, fire
630 maps derived from satellite information, utilizing a detection algorithm that includes small
631 fires (Randerson et al., 2012), were examined for the two periods (summer and winter)
632 under investigation. During both periods no biomass burning activity was identified ruling
633 out interferences due to this source. During winter, areas where simultaneous increases in
634 absorption and number concentration were identified and attributed to local sources and
635 not the Paris plume. The particle number concentrations in these areas were relatively low
636 though. The potential interference of these sources has a modest to small effect on our
637 estimates regarding the evolution of the Paris aerosol number plume. A similar method of
638 plume identification that involves aerosol absorption was also implemented by Freney et
639 al. (2014) for the same campaign. Increased concentrations of toluene and benzene, both of
640 which are anthropogenic, were also encountered in these plumes.
641 Due to air traffic restrictions, the ATR-42 was not allowed to get closer than 30 km
642 to the Paris center, but the Paris plume could be identified as far as 200 km away from the
643 city. As stated earlier, airborne measurements were conducted on days when pollution
644 levels were above average and the flight paths were determined a priori based on
645 forecasted values of the numerical model CHIMERE, thus the sample is positively biased.

Mobile laboratories on the ground sampled closer to Paris during the whole campaign, but separating the plume from the background was cumbersome (von der Weiden-Reinmüller et al., 2014a).

During summer the averaged aircraft measured N_{10} within the Paris plume was $10.1\pm5.6\times10^3$ cm^{-3}, which was 14% higher than the concentrations observed outside of the Paris plume ($8.8\pm6.5\times10^3$ cm^{-3}), defining the background concentrations. The high background number concentrations in this N to E quadrant where all of the summer flights but one took place (grey, blue and green lines in Fig. 2) are consistent with the ground (stationary and mobile) observations.

During all summer flights, with the exception of July 25, "hot spots" outside of the Paris plume where particle number concentrations similar to or higher than those of the Paris plume were identified without increase in black carbon or anthropogenic volatile organic compounds (VOCs; benzene, toluene). The "hot spots" where the particle number increase occurred were separated into three groups based on their location with respect to the Paris plume as "upwind", "alongside" and "local".

The "upwind" events were identified upwind of Paris four times, always near IDF (Fig. 12b) and simultaneously with regional nucleation events observed at least at two of the ground sites. The number concentration increases were thus attributed to NPF. Assessment of the spatial extension of these events was complicated by the presence of the plume and limited by the designated flight paths (Fig. 2). In general, the N_{10} measured upwind was 40% higher than that measured in the plume during these "upwind" NPF events.

The "alongside" events occurred at an average distance of 40 km from the plume edge and were attributed to NPF (Fig. 12d). The average number concentration increased by 47% in comparison to the concentration within the Paris plume. The area in between the Paris plume and the hot spot area always exhibited at least 20% lower concentrations than the latter two (Fig. 12d shows the number concentration with respect to cardinal coordinates and Suppl. Fig. 34 as a time-series). The alongside events occurred during four flights (July 1, 15, 21, and 28), two of which were non-event days for all ground sites and two when NPF was identified at SIRTA and LHVP, but not at GOLF. The high N_{10} areas covered approximately 3,000 km^2 along the plume.

In order to investigate why the alongside events occurred only on one side of the Paris plume during these flights, each flight path was separated into three areas: (1) the area with high N_{10} outside of the plume, (2) the plume area and (3) the area on the other

side of the plume, where no increase in particle number was observed. The observed differences between both sides of the Paris plume with respect to the CS, solar intensity and isoprene concentration, which has been reported as a potential inhibitor of NPF in forested areas (Kiendler-Scharr et al., 2009; Kanawade et al., 2011), were 12%, 5% and 6%, respectively (Suppl. Fig. 45). These relatively small differences probably cannot explain the observed phenomenom.phenomenon. Other pollutants such as benzene, toluene, monoterpenes, methacrolein, methyl vinyl ketone, O_3, CO, but also meteorological parameters such as temperature and RH were investigated in order to identify potential reasons for the different particle number concentrations between both sides of the plume. Differences in all the investigated parameters were less than 10%. These events clearly require more investigation with instrumentation that can sample particles smaller than 10 nm in combination with trace gas measurements relevant to NPF (e.g. SO_2). Unfortunately, there were no ground measurements in the areas in which the alongside events were identified.

The "local" events were the most frequent (6 out of the 11 study cases) and occurred either at the north coast of France downwind of the city of Fecamp (4 events) and were associated with high or medium tide height (indicating influence of ship emissions?), or near the city of Aulnoye-Aymeries (4 events). On two occasions these events were identified on both locations during the same flight. Because the local events were always associated with specific areas, the particle number concentration increase was attributed to local sources.

During the three winter flights, the Paris plume $N_{2.5}$ was 45% higher than the background and no "hot spots" were identified, consistent with ground measurements where no NPF was identified.

8. Summary and conclusions

Ambient aerosol number concentrations were monitored at the center of Paris (LHVP) along with two satellite suburban stations (SIRTA, SW and GOLF, NE). Mobile measurements were performed by two mobile laboratories and the SAFIRE aircrafts during July 2009 (summer, ATR-42) and January - February 2010 (winter, Piper-Aztec).

During summer, $N_{10\text{-}500}$ (number concentration for particles between 10 and 500 nm diameter) at the city center was lower by 14% than at the downwind (GOLF) and 54% higher than at the upwind (SIRTA) suburban site, respectively. The contribution of particles with diameters between 10 and 30 nm to $N_{10\text{-}500}$ increased from the mostly upwind

714 suburban site (30% at SIRTA) over the city center (40% at LHVP) to the mostly
715 downwind suburban site (50% at GOLF). The contribution of particles with diameters
716 between 30 and 100 nm ranged between 40-50% and followed the opposite trend (highest
717 upwind, lowest downwind).

718 During summer at SIRTA, $N_{10\text{-}500}$ increased to $9.9\pm2.4\times10^3$ cm^{-3} when the site was
719 downwind of Paris and decreased to $4.2\ \pm2.5\times10^3$ cm^{-3} when the site was upwind. At
720 GOLF, located at approximately the same distance from the city center as SIRTA but in
721 the opposite direction (NE), the effect of Paris emissions was not clear, suggesting a high
722 background $N_{10\text{-}500}$ at the measurement location for all wind directions.

723 NPF events were observed at all sites during summer. At SIRTA and LHVP, events
724 were identified every second day and at GOLF once every four days on average. The lower
725 frequency of NPF events at GOLF was mainly due to interferences from nearby traffic and
726 instrumental limitations which did not allow clear event identification. NPF occurred
727 during periods when the CS was lower by 45%, 25% and 50% at SIRTA, LHVP and
728 GOLF, respectively, in comparison to each site's average value, indicating that the CS may
729 have been a controlling factor for the frequency of events. Solar intensity was higher by
730 30% and 20% on event days compared to non-event days at SIRTA and LHVP,
731 respectively. At GOLF, solar intensity was higher by 8% during non-event days compared
732 to event days. On average, NPF events caused $N_{10\text{-}500}$ to double at all stationary
733 measurement sites.

734 Average particle growth rates were 5.5, 4.6 and 6.1 nm h^{-1} at SIRTA, LHVP and
735 GOLF, respectively. The differences between these average growth rates were not
736 statistically significant.

737 The particle number concentration within the Paris emission plume was found to
738 decrease exponentially on the ground with distance from the Paris center during both
739 campaigns. At distances from the city center greater than 70 km, $N_{2.5}$ was approximately
740 1.4×10^4 cm^{-3} regardless of season or whether the measurements were affected by the Paris
741 plume. However during summer background conditions (not affected by Paris), $N_{2.5}$ close
742 to GOLF (second circle in Fig. 1) was approximately a factor of two higher, in agreement
743 with $N_{10\text{-}500}$ measurements at GOLF that indicated a higher background in the region NE of
744 Paris.

745 The Paris plume was identified by aircraft measurements at an altitude of 600 m,
746 using black carbon as a tracer, as far as 200 km away from the city center. Averaged N_{10}
747 outside and within the Paris plume were $8.8\pm6.5\times10^3$ and $10.1\pm5.6\times10^3$ cm^{-3}, respectively

748 which corresponds to a 33% increase. During summer, "hot spots" of high particle number
749 concentrations were identified outside of the Paris plume at 600 m altitude. On four
750 occasions the particle number concentration increase was located upwind of the ground
751 stations simultaneously with regional NPF observed on the ground at least at two of the
752 sites. These increases therefore were attributed to NPF. Increased particle number
753 concentrations were also identified along one side of the plume on four occasions. A
754 number of parameters were investigated including CS, solar irradiance, anthropogenic and
755 biogenic VOC concentrations among others, as possible explanations for this asymmetry.
756 All differences observed between both sides of the Paris plume were approximately 10%
757 or lower, so none of these could explain the observations.
758 During winter the absolute $N_{10\text{-}500}$ was higher by a factor of two at both suburban
759 sites and by 36% at the city center compared to summer. At LHVP particles from 10 to 30
760 nm accounted for 44% of the $N_{10\text{-}500}$ on average and those from 30 to 100 nm for 40%. At
761 GOLF, similar to summer, the $N_{10\text{-}500}$ was dominated by particles with diameters between
762 10 and 30 nm which accounted for 56%, followed by particles from 30 to 100 nm (33%),
763 following the same trends as during summer. At SIRTA the contribution of particles from
764 10 to 30 nm and from 30 to 100 nm to the $N_{10\text{-}500}$ was 42% and 39%, respectively.
765 Regardless of site or season a mode, centered at a diameter below 20 nm, was always
766 present and was dominating during winter at all sites. During winter the higher CS and
767 lower solar intensity compared to summer prevented particles from growing to sizes larger
768 than 10 nm.
769 A complete year of air ion measurements (including the two intensive campaigns
770 discussed in the present paper) has been recently presented by Dos Santos et al. (2015).
771 These measurements took place in the MEGAPOLI site in the center of Paris (LHVP
772 station) from July 2009 to September 2010. During this year, the highest NPF frequency in
773 Paris was observed during July 2009 (the summer campaign examined in this work) and
774 the lowest during the winter (which includes the winter campaign in this work). Therefore
775 our analysis above focused on two extreme NPF periods in Paris. During summer under
776 clean conditions and peak NPF frequency and during winter under polluted conditions and
777 minimal NPF frequency.
778
779
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Table 1. Summary of main MEGAPOLI measurements used in this study.

Variable	Instrument	Group	Time Resoluti	Sample Condition
ATR-42				
Absorption (summer)	PSAP[a]	LaMP[j]	1 sec	dry
Trace Gas Concentration	HS PTR-QMS [b]	CNRS[k]	1 sec	dry
Aerosol Number Concentration	TSI 3025 CPC[c]	CNR [l]	1 sec	dry
Aerosol Number Concentration	TSI 3010 CPC[c]	LaMP[j]	1 sec	dry
Absorption (winter)	PSAP[a]	CNR [l]	1 sec	dry
MoLa				
Aerosol Number Concentration	TSI 3786 UWCPC[d]	MPIC[m]	1 sec	ambient
MOSQUITA				
Aerosol Number Concentration	TSI 3010 CPC[c]	PSI[n]	1sec	ambient
Aerosol Number Concentration	UHSAS[e]	PSI[n]	1 sec	ambient
SIRTA				
Aerosol Number Size Distribution (10–500 nm)	SMPS[f]	CNRS[k]	10 min	ambient
Aerosol Number Size Distribution (6–800 nm)	DMPS[g]	UoH[p]	9 min	ambient
Positive/Negative Ion Size Distribution (0.8–40 nm)	AIS[h]	UoH[p]	3 min	ambient
LHVP				
Aerosol Number Size Distribution (3–620 nm)	DMPS[g]	IfT[q]	10 min	ambient
GOLF				
Aerosol Number Size Distribution (5 nm–1 μm)	EAS[i]	MPIC[m]	1 min	ambient

Aerosol Number Concentration	UHSAS[e]	PSI[n]	1 sec	ambient
SIRTA				
Aerosol Number Size Distribution	SMPS[f]		10 min	dry
Aerosol Number Size Distribution (6–	DMPS[g]	UoH[p]	9 min	ambient
LHVP				
Aerosol Number Size Distribution (3–	DMPS[g]	IfT[q]	10 min	dry
Positive/Negative Ion Size Distribution (0.8–40 nm)	AIS[h]	UoH[p]	3 min	ambient
GOLF				
Aerosol Number Size Distribution (5	EAS[i]	MPIC[m]	1 min	ambient

[a]PSAP: Particle Soot Absorption Photometer; [b]HS PTR-QMS: High Sensitivity Proton Transfer Reaction-Quadrupole Mass Spectrometer; [c]CPC: Condensation Particle Counter; [d]UWCPC: Ultrafind Water Condensation Particle Counter; [e]UHSAS: Ultra High Sensitivity Aerosol Spectrometer; [f]SMPS: Scanning Mobility Particle Sizer; [g]DMPS: Differential Mobility Particle Sizer; [h]AIS: Air Ion Spectrometer; [i]EAS: Electrical Aerosol Spectrometer; [j]LaMP: Laboratoire Meteorologie Physique; [k]CNRS: Centre national de la recherche scientifique; [l]CNRM: Centre National de Recherches Météorologiques; [m]MPIC: Max Planck Institute for Chemistry; [n]PSI: Paul Scherrer Institute; [o]CMU: Carnegie Mellon University; [p]UoH: University of Helsinki; [q]IfT: Leibniz Institute for Tropospheric Research.

Table 2. Aerosol number concentrations during the summer and winter campaigns and characteristics of NPF during summer. σ is the standard deviation.

	Average ± 1σ Number Concentration (10 - 500 nm) 1000/cm^3		Average increase±1σ in Number Concentration due to NPF (%)	Growth Rate ±1σ (nm h^{-1})
Site	*Summer*	*Winter*	*Summer*	*Summer*
GOLF	13.3±6.8	25.3±15.1	127±110	6.1±1.8
LHVP	11.4±5.1	15.6±7.1	100±50	4.6±1.9
SIRTA	5.3±3.1	10.1±5.7	129±59	5.5±4.1

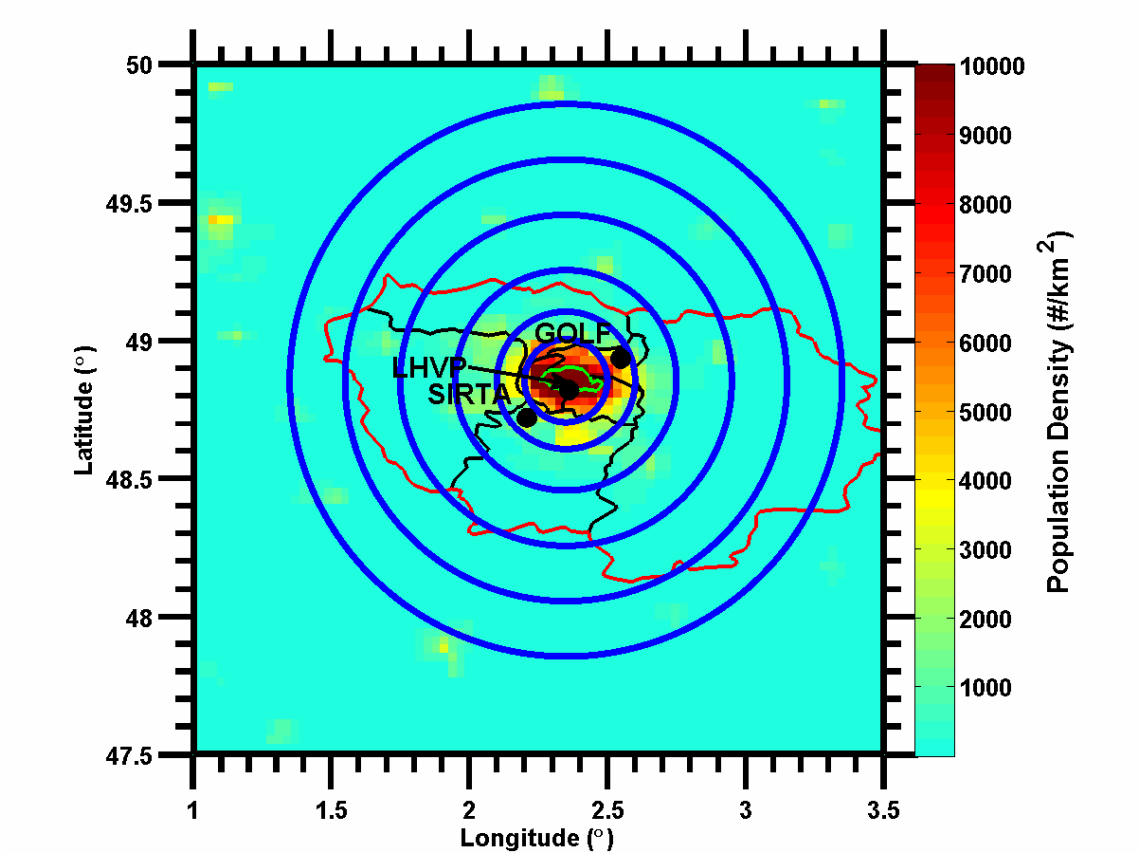

Fig. 1. Population density and administrative map of Paris. Outlined in red is Île de France and in green Paris. The three ground stations (SIRTA, LHVP and GOLF) are depicted with black dots. The map is separated into sectors depicted by blue lines, formed by concentric circles centered at kilometer zero of Paris (48.8534 °N 2.3488 °E). The radius of the circles is 0.15, 0.25, 0.4, 0.6, 0.8 and 1 °, which corresponds to 16.7, 27.8, 44.4, 66.7, 88.9 and 111.1 km.

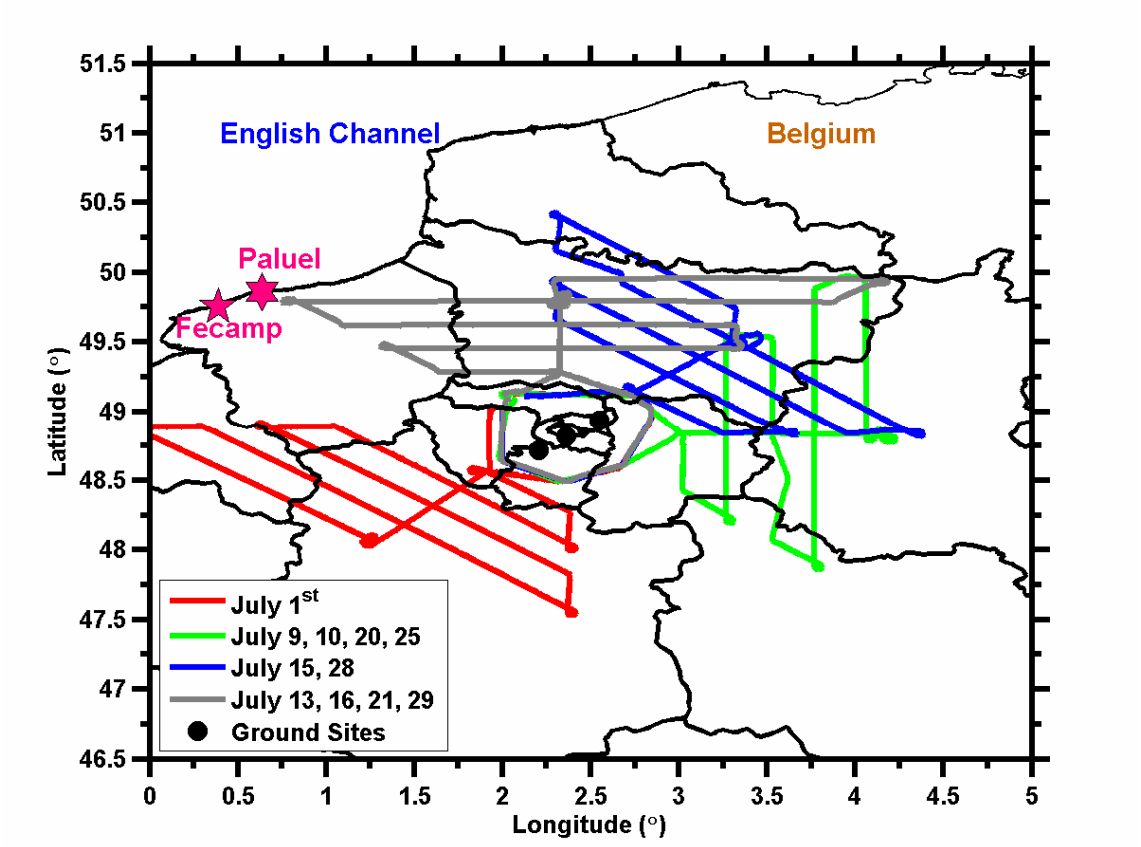

Fig. 2. Flight paths of the ATR-42 aircraft during the summer campaign. Different colors correspond to different flight routes. The cities of Fecamp and Paluel are also depicted in the map.

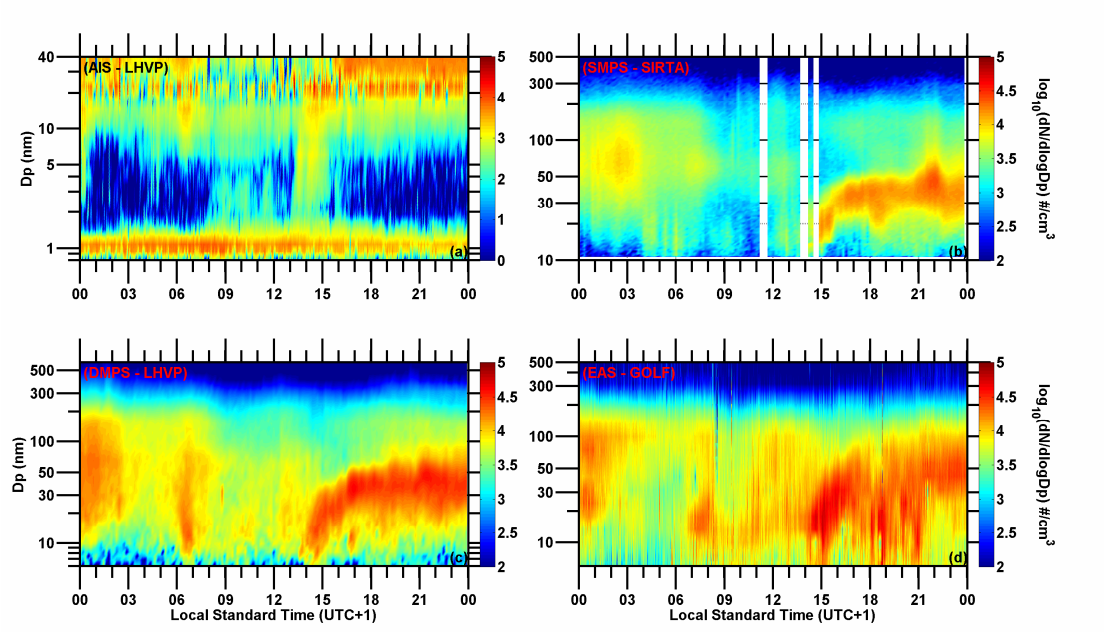

Fig. 3. Size distribution measurements during a nucleation event day (12 July 2009) at all ground sites. (a) AIS measurements at SIRTALHVP, (b) SMPS measurements at SIRTA, (c) DMPS measurements at LHVP, (d) EAS measurements at GOLF. Time of day corresponds to local standard time (UTC+1). D_p is the particle diameter.

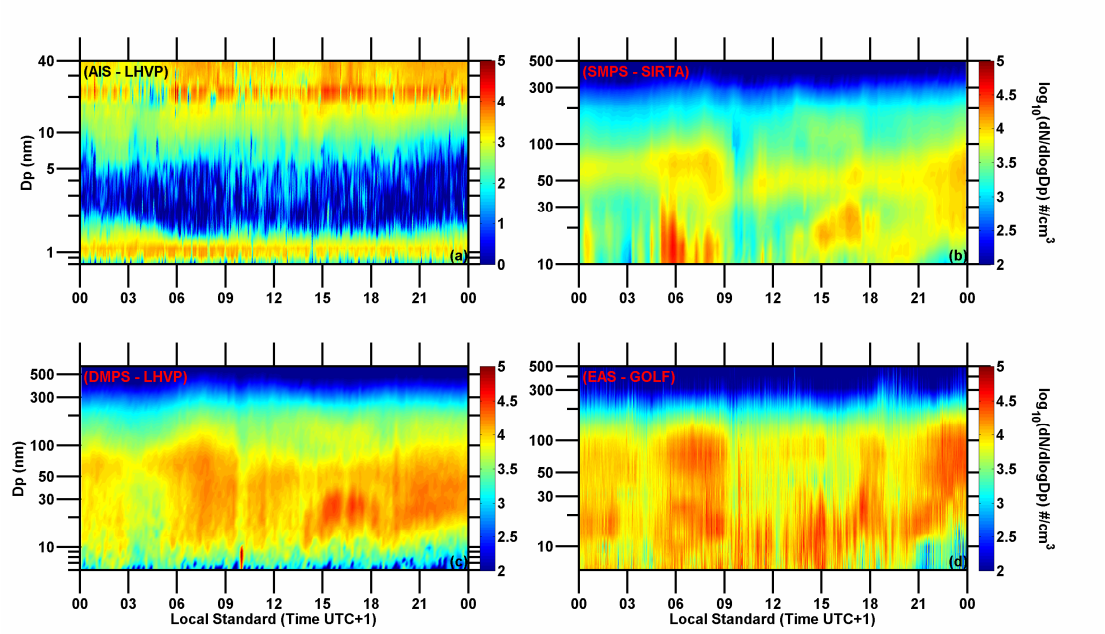

Fig. 4. Size distribution measurements during an undefined event day (10 July 2009): (a) AIS measurements at SIRTALHVP, (b) SMPS measurements at SIRTA, (c) DMPS measurements at LHVP, (d) EAS measurements at GOLF. Time of day corresponds to local standard time (UTC+1). D_p is the particle diameter.

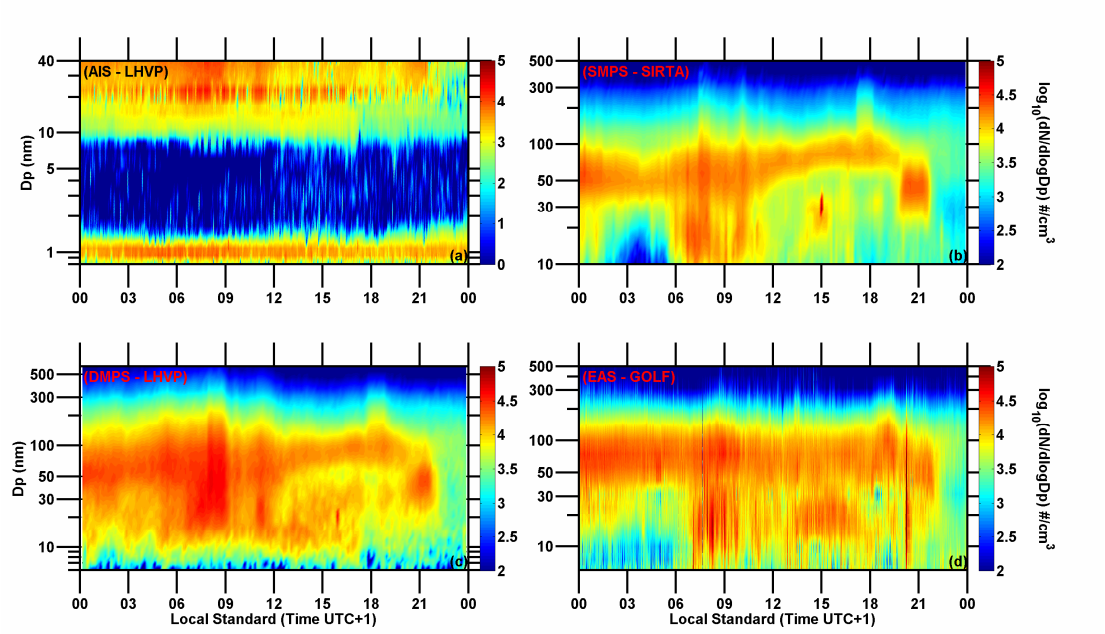

Fig. 5. Size distribution measurements during a non-event day (29 July 2009): (a) AIS measurements at SIRTALHVP, (b) SMPS measurements at SIRTA, (c) DMPS measurements at LHVP, (d) EAS measurements at GOLF. Time of day corresponds to local standard time (UTC+1). D_p is the particle diameter.

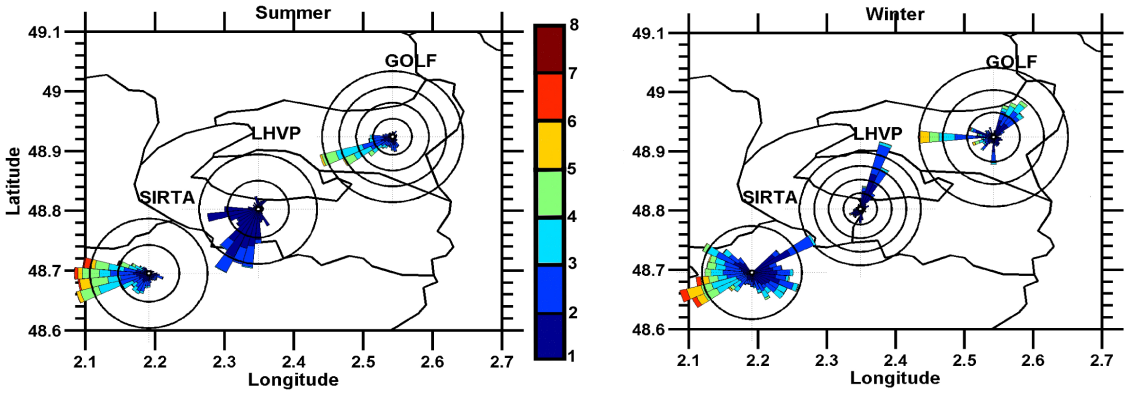

Fig. 6. Wind direction rose plots during the summer and winter campaigns at each of the ground sites. Each rose segment corresponds to an angle bin of π/18 (i.e. 20°) and concentric circles at each site correspond to 5% relative frequency. Wind speed, in m s^{-1}, corresponding to each size bin is color coded inside each rose. Wind speeds below 1 m s^{-1} have been omitted from the graph.

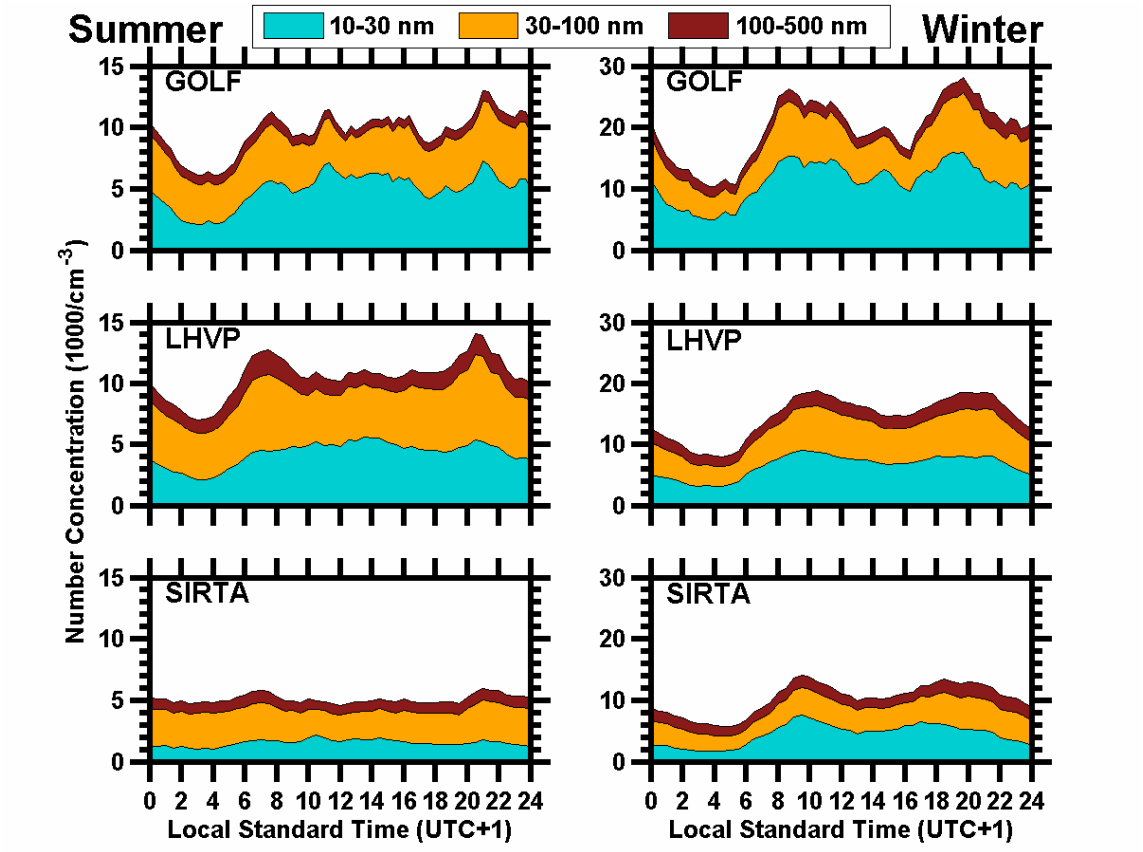

Fig. 7. Number concentration diurnal profiles of summer (left) and winter (right) campaigns for size ranges from 10 to 30 nm, 30 to 100 nm, and 100 to 500 nm, respectively. Different scales are used for each season.

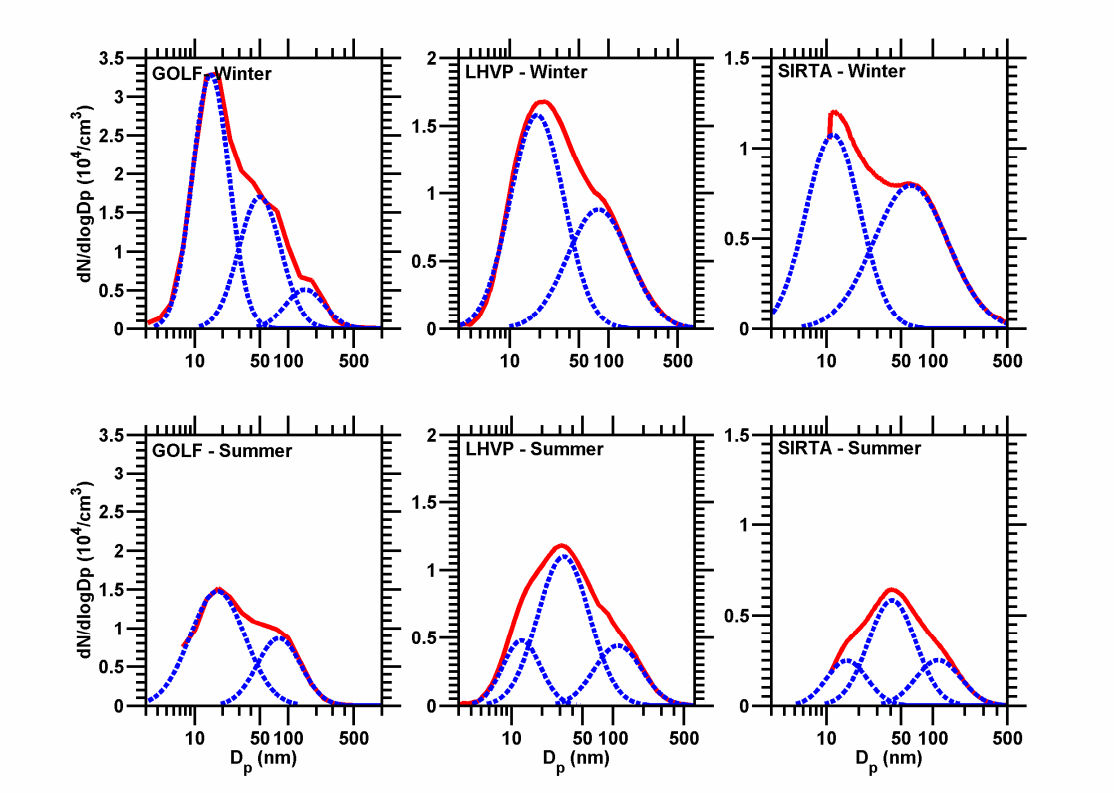

Fig. 8. Campaign average particle number size distributions for winter (top) and summer (red) and winter (black) of allbottom) for the three ground sites based on measurements of EAS at GOLF, DMPS at LHVP and SMPS at SIRTA. Each average size distribution (solid red line) is deconvoluted to lognormal modes (dashed blue lines). Note the different scaling of the y-axes. between sites.

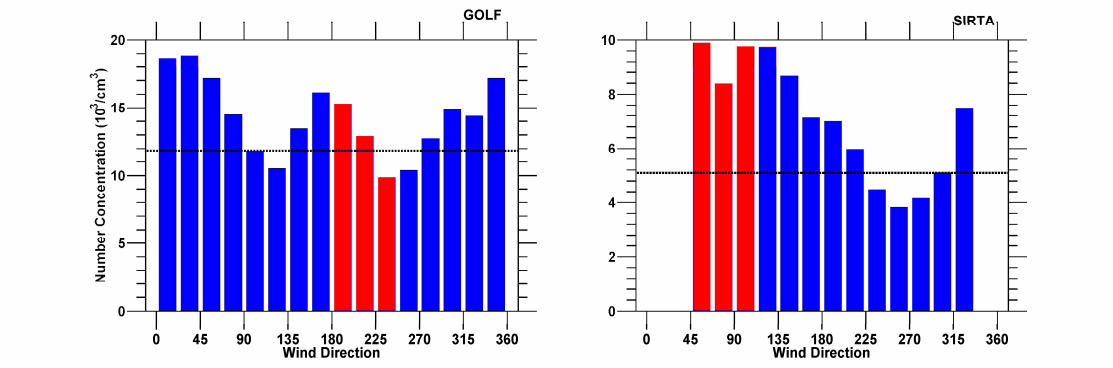

Fig. 9. Number concentrations measured at the two satellite sites during summer with respect to wind direction / air mass transport direction measured at the respective site. The angles which indicate that the air mass traveled through the city center prior to reaching the site are depicted in red. The horizontal dashed black line corresponds to the campaign average for each site. Periods with wind speed below 1 m s^{-1} were omitted from the analysis.

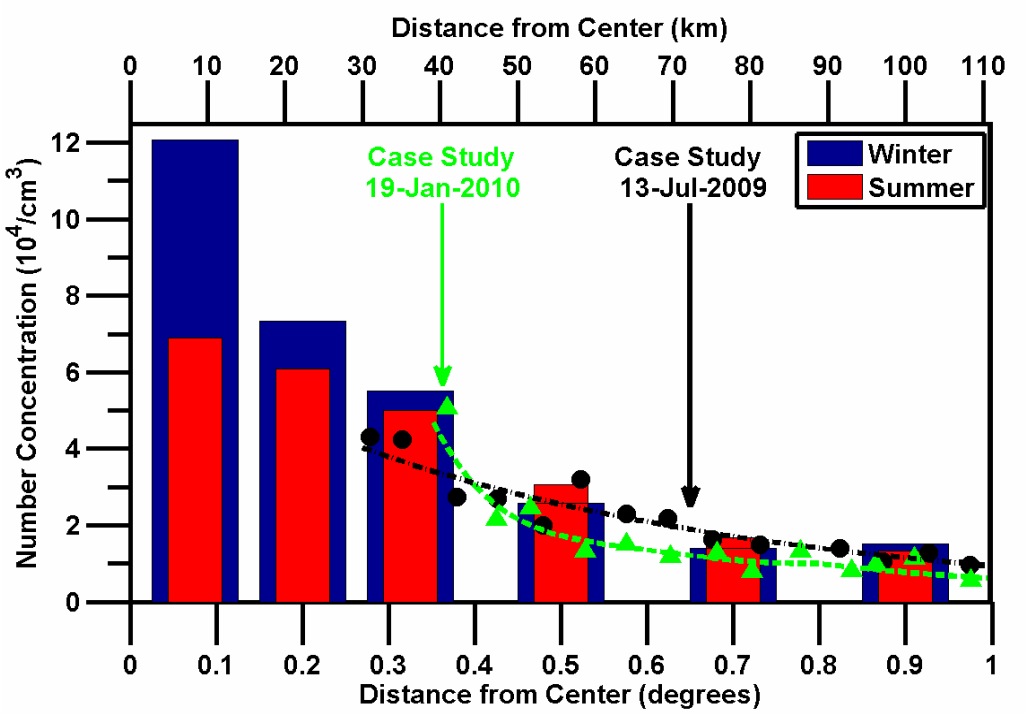

Fig. 10. Average number concentration ($N_{2.5}$) with respect to distance from the city center measured by the mobile platforms during summer (red) and winter (blue). During both campaigns an exponential decreasedecrease of the number concentration with respect to distance was observed. The number concentration measured in an axial measurement on a case study day is also depicted in the graph for summer (black dots) and winter (green triangles).

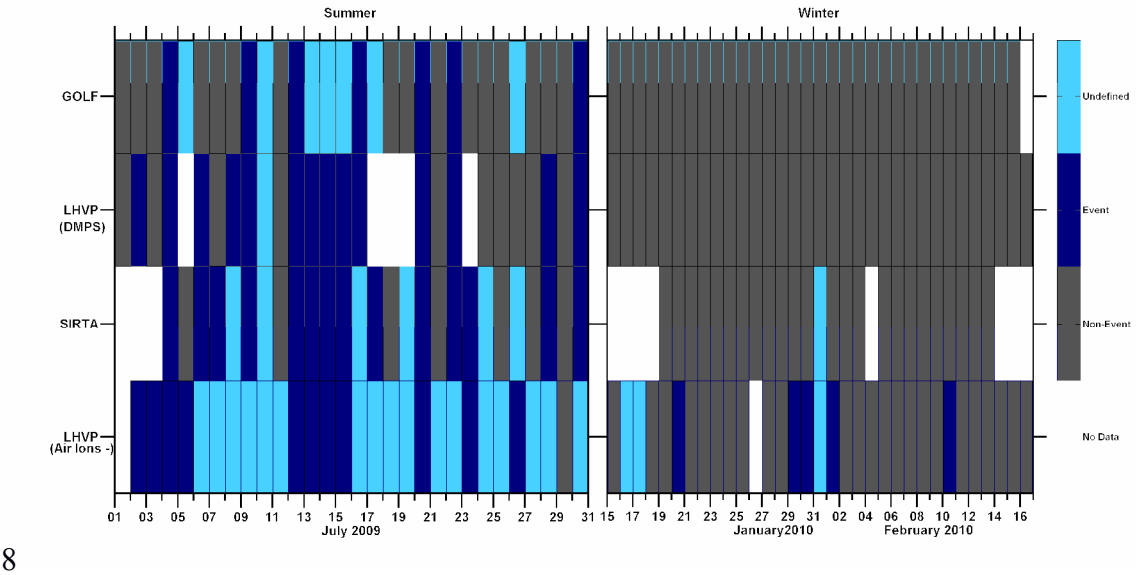

Fig. 11. Nucleation analysis results during summer and winter for all ground sites. Events, non-events, undefined and lack of data are depicted in blue, grey, light blue and white, respectively.

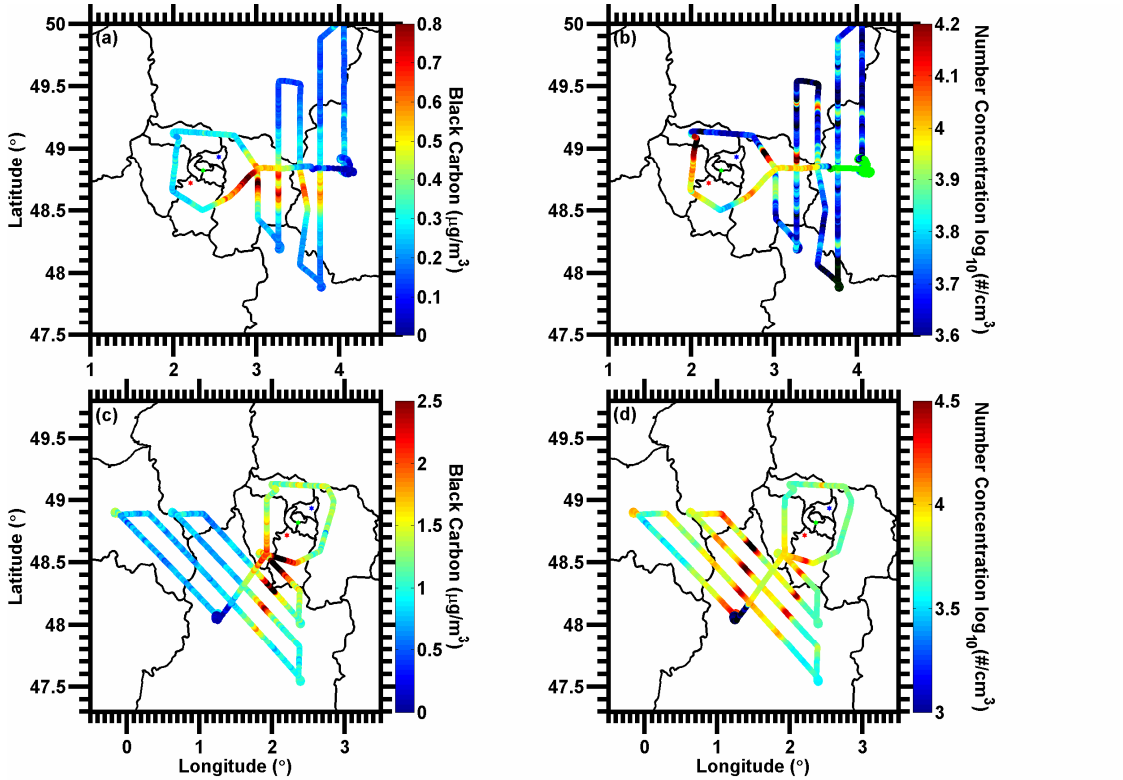

Fig. 12. Flight trajectories for 9th (a, b) and 1st (c, d) July 2009, color coded for black carbon and number concentrations (N_{10}), respectively. Black carbon concentrations are used as tracers of the Paris plume (a, c); its direction relative to the city center indicates wind direction. Red, green and black dots within the figure correspond to the locations of SIRTA, LHVP and GOLF, respectively. Increased number concentrations were observed outside of the plume. During July 9 (b) the area where the number concentration increased was located upwind of the city center and NPF was identified at all ground sites. During July 1 (d) the particle number increase was observed along the plume. The number and black carbon concentration corresponding to c and d are also shown with respect to time in Suppl. Fig. 3.

Supplementary material to

Ultrafine particle sources and in-situ formation in a European Megacity

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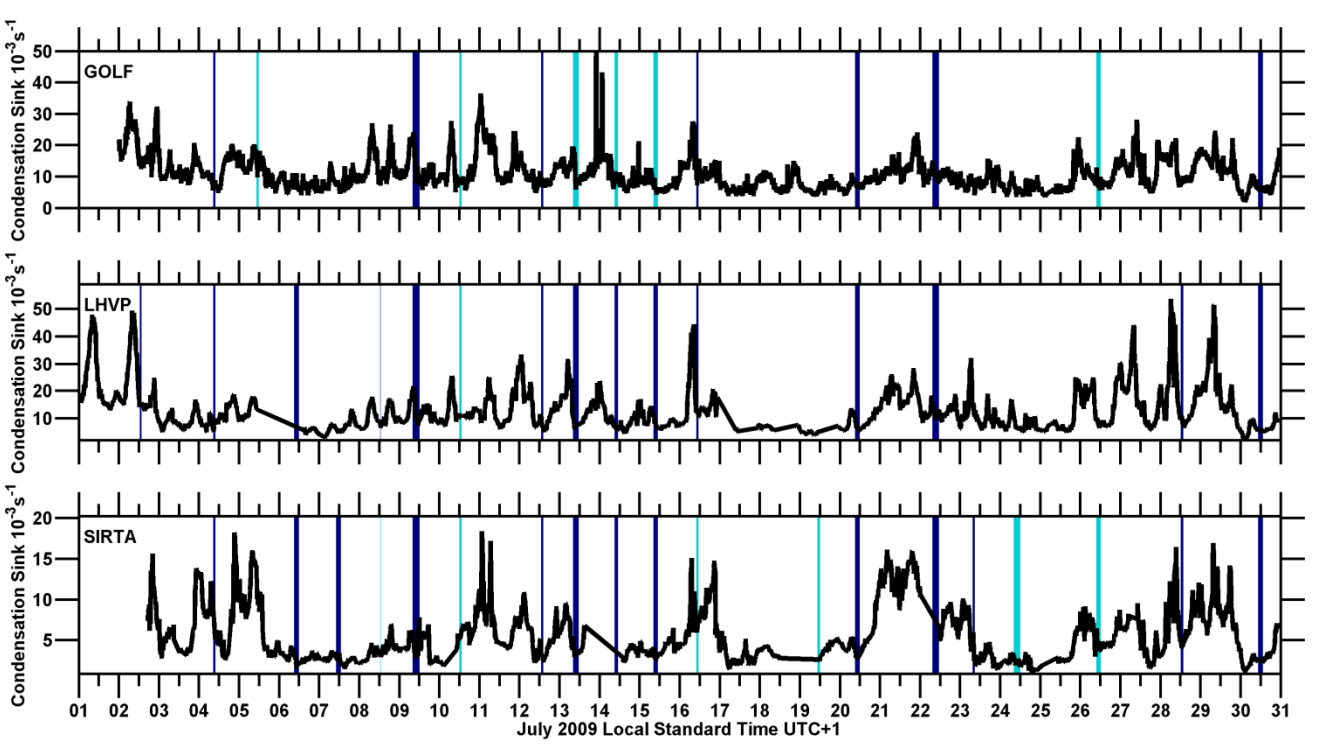
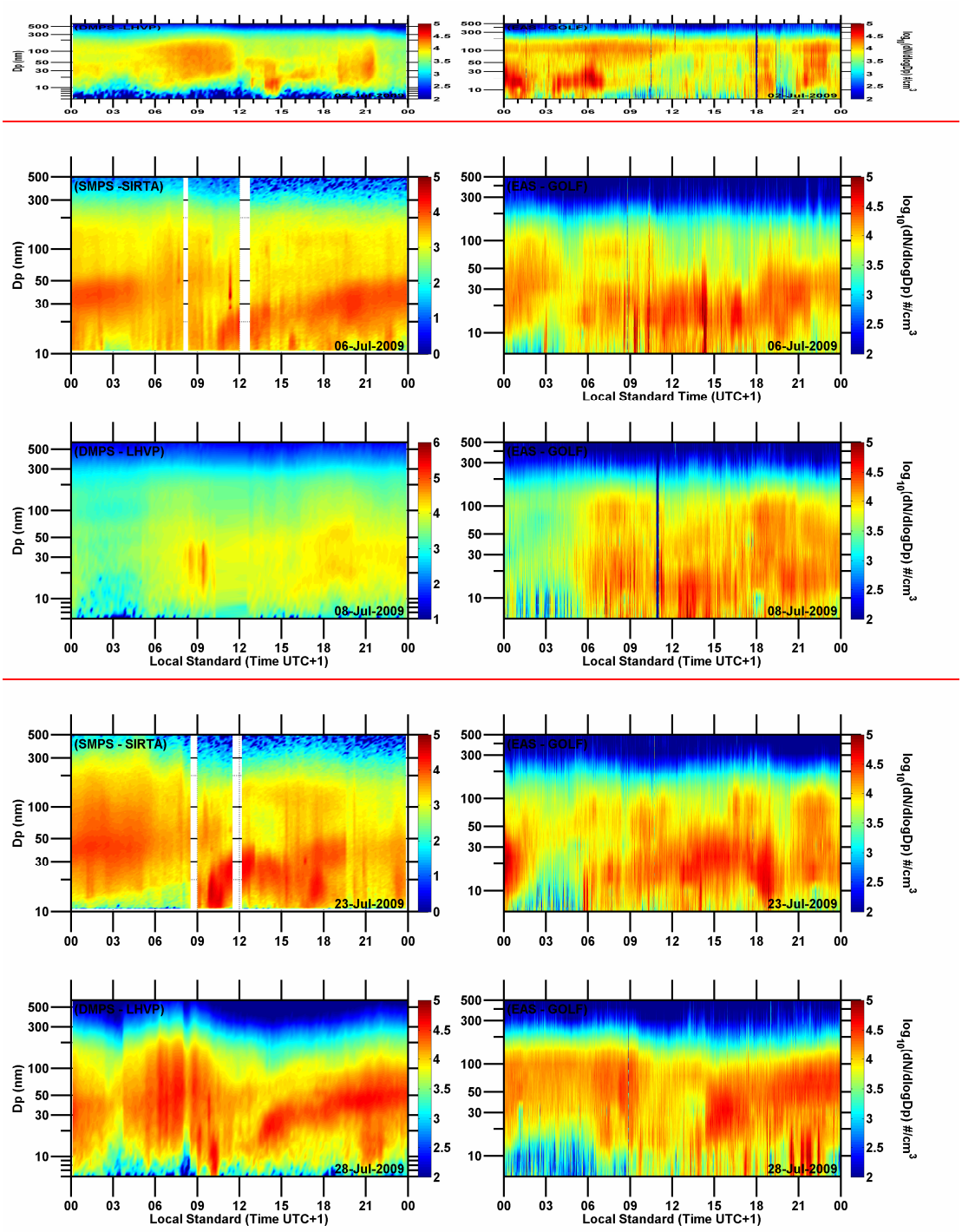
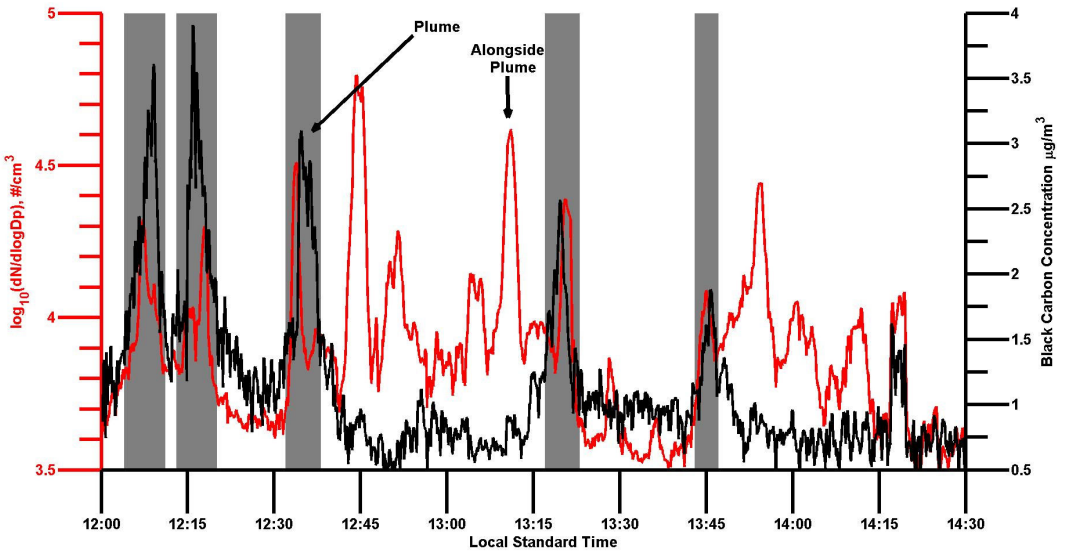


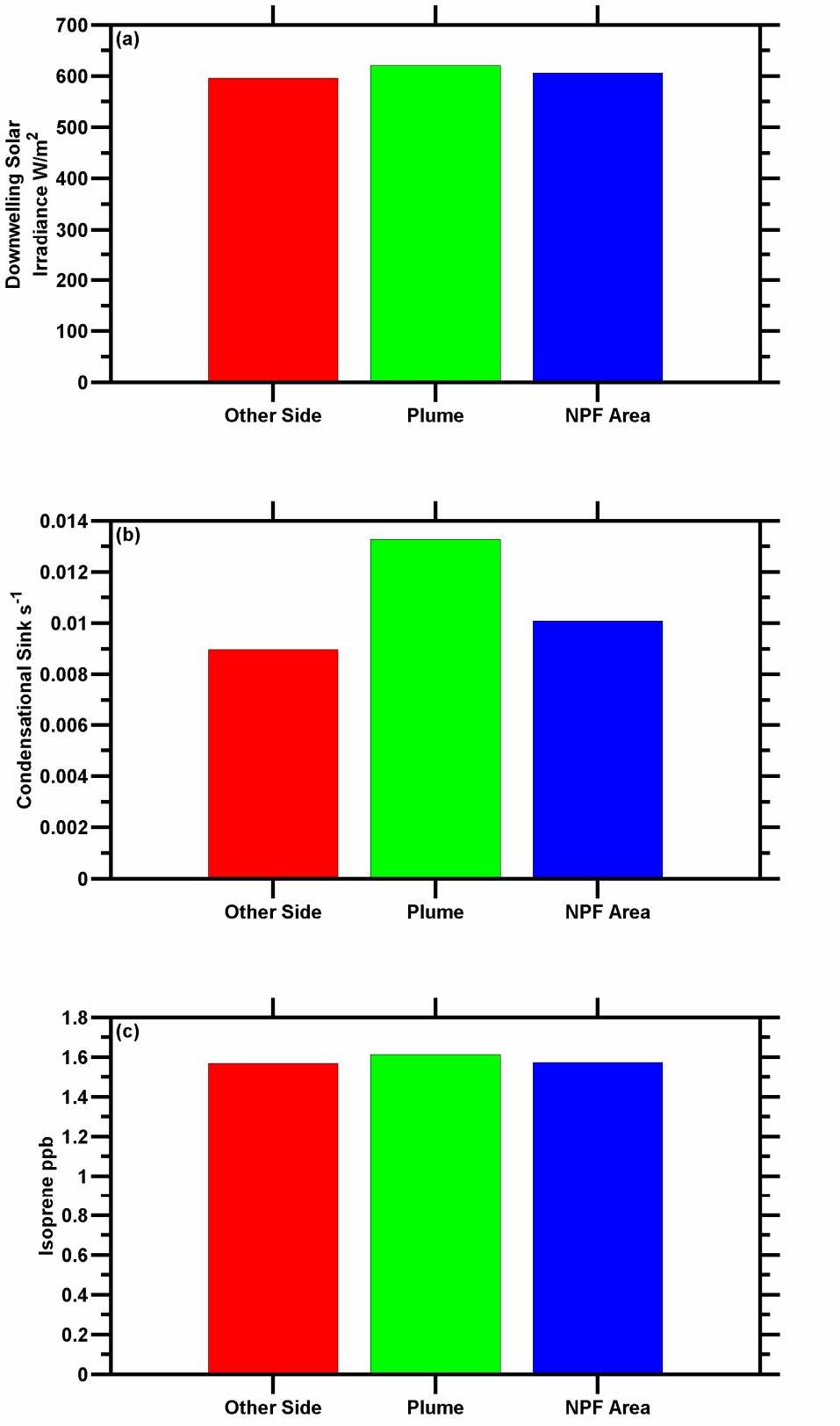
Suppl. Fig.1. Condensation sink measuredat the three ground sites during July 2009. Dark and light blue bars indicate the event and undefined periods, respectively.



Suppl. Fig. 2. Number size distribution time series when a nucleation event was identified at SIRTA and/or LHVP but not at GOLF. D_p is the particle diameter.



Suppl. Fig. 3. Number (red line) and black carbon (black line) concentrations during airborne measurements on July 1st 2009. Number concentration increases observed simultaneously with increases in black carbon mass concentration (grey areas) were attributed to the Paris plume. Number concentration increases were also observed along the plume.



Suppl. Fig. 4. Downwelling solar irradiance (top), condensation sink (middle) and isoprene concentration (bottom) comparison of the Paris plume with areas on either side of the plume when high particle concentrations were observed at one side outside of the plume. Significant differences among these areas were not observed with respect to condensation sink, isoprene and solar irradiance.