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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ultrafine particle sources and in-situ formation in a European megacity" *by* M. Pikridas et al.

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(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, RO_x, 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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