

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 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 sec-*

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tion within the introduction would definitely benefit the special issue. Between the first and second paragraphs of the introduction (i.e. between the PM_{2.5} 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 (Skylakou et al., 2014) addressing the sources of “fine” particles, defined therein as PM_{2.5}, 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, removal. There are a few efforts in the literature trying to estimate the sources of the particle number (Wählén et al., 2001; Houssein et al., 2004; Zhou et al., 2005; Chan and Mozurkewich, 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

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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 see 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 has been added.

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