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

Anonymous Referee #3

Received and published: 24 May 2015

The paper describes a specific subset of the science from the MEGAPOLI project; namely in situ observations of freshly formed particles of secondary origin of a few nm in diameter that the authors define as ultrafine. Notwithstanding the possible ambiguity with such a definition when applied to urban pollution (as raised by the other reviewers), the paper provides a useful addition to the literature in characterising new particle formation events in a megacity. Such studies have been rendered much more insightful in recent years by the development of instrumentation to better characterise behaviour of the smallest particles much sooner after formation than previously accessible. The description and characterisation of new particle events across the various static locations and mobile platforms in terms of number concentrations and size distributions alone make this a useful paper. The core and ancillary measurements appear to have been

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appropriately carried out and the data robustly categorised. The text is well-written and the figures are clear. I think the paper content and quality is appropriate for ACP.

The scope of the paper appears to be a little narrower than promised by the title. I have few specific suggestions beyond those raised by the other reviews (many of which appear quite valid), but the authors might like to consider the following to better manage the expectation of a reader.

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 PM_{2.5} and NPF related sections) or between the third and fourth paragraphs (linking and contextualising in situ emission and in situ formation contributions) might be appropriate places.

It is indeed particularly challenging to provide quantitative insight into the sources and mechanisms of NPF. The paper does a good job at reporting the observations and phenomenologically describing the contrast in frequency of NPF events between summer and winter and broadly characterising the geographical behaviour of the resulting enhancements in ultrafine particles. However, as the authors clearly describe, there is little clear quantitative conclusion that is drawn from the measurements and certainly little insight into specific sources of the ultrafine particles. The identification of in-plume and out-of-plume events illustrates the difficulty in evaluating the relative contributions to number and the sectorisation into Paris and non-Paris contributions is a rather coarse attribution of sources. 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. It is difficult to consider attribution of NPF

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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.

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.

Addressing my comments may require relatively minor revision, addressing the valid comments from the other referees more major.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 5663, 2015.

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