

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 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

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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.

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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 percent. 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 MOSQITA 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 percent) 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

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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

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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.

References

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von der Weiden-Reinmüller, S.-L., Drewnick, F., Crippa, M., Prévôt, A. S. H., Meleux, F., Baltensperger, U., Beekmann, M., and Borrmann, S.: Application of mobile aerosol and trace gas measurements for the investigation of megacity air pollution emissions: the Paris metropolitan area, *Atmos. Meas. Tech.*, 7, 279-299, 2014a.

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