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The paper *Vertical and horizontal distribution of regional new particle formation events in Madrid* from C. Carnerero *et al.* presents an interesting dataset about new particle formation in the metropolitan area of Madrid.

A proper understanding of NPF in urban environments is still missing despite the potentially high impact that ultrafine particles could have on the total particle number concentration and their health effects. To address this issue field and laboratory experiments are needed and the results presented in this work provide new insights in this direction. By providing a horizontal and vertical distribution of new particle formation episodes, the authors clearly show that these events are occurring over a large part of the metropolitan area underlining their impact on a large region. In my opinion this paper falls within the scope of ACP but there are several issues that need to be addressed before publication.

Major comments

- Despite the interesting dataset this paper fails in conveying a clear message due to its structure and the way the results are presented. In particular the authors mix the main results (horizontal and vertical characterization of NPF in Madrid) with episodes (e.g. the UFP peaks at night) and phenomena (e.g. shrinkage of particle size) that are not strictly related with them. To address this issue I suggest to the authors to focus more on the important results and emphasize them in a clearer way as well as to restructure the results section in order to make a clear separation between these results and all the minor observations.

- The authors state several times that NPF is dominating the total particle concentration in Madrid, however it is not clear how they separate between newly formed particles and ultra fine particles directly emitted from cars. I assume that the formation rate calculation is biased by the fact that primary UFP were not taken into consideration and this would explain why formation rates at the urban stations are higher than those measured in the suburban whereas the growth rates are smaller. The authors need to quantitatively estimate the source of UFP and revise the formation rates excluding primary UFP from their results. Doing this would also allow to compare directly the Madrid case with the other locations reported in the introduction. Probably the simplest way to discriminate between primary and secondary UFP would be to use the data of the particle concentrations below 9 nm, that should be available at all the measurement sites.

- In section 2.2 no details are provided about the sampling conditions. The authors should give a short description of the inlets and explain for example if losses were measured and/or calculated, if any intercomparison between the different particle counters was performed, etc... Moreover, a big part of the work is based on the measurements performed with the Hy-SMPS but no proper characterization is provided (figure S2 is not really useful to evaluate the performances of this instrument) and the only cited paper is written in Korean (I'm also not sure whether this is a peer reviewed journal or not). For these reasons a more complete characterization of this instrument is required, for example it could be useful to compare the Hy-SMPS with a reference SMPS while looking at separate size bins and not only at the total concentration.

- In section 2.3 it is explained how formation and growth rates are calculated, however no explanation is provided about the decision of using the 9-25 nm range, despite the fact that measurements of particle concentrations down to about 1nm were performed. I would argue that this is not the best choice, in particular for the formation rates that could be highly biased by primary emissions as previously explained. Moreover, using a smaller reference diameter for the formation rate would permit to better compare with measurements performed in other locations and/or in chamber studies. For these reasons I would suggest to calculate formation rates for a more meaningful size (below 5 nm) and eventually to calculate growth rates at two different sizes (for example keep the 9-25 nm range for comparability with the vertical sounding and add a lower size GR depending on the availability of the existing data). Finally, uncertainties for all the growth and formation rates should be estimated in order to make a proper comparison between different locations and days.

- Section 3.2.2 reports a comparison of growth rates and formation rates at the different sites but without including the vertical profiles (in this case growth rates are provided in section 3.3). I think that the paper would benefit by having all the growth rates presented together, this would improve the readability and the clarity of the paper.

- Section 3.2.3 reports PTR measurements of 3 ions that show some correlation with the particle growth, however this section is not adding any valuable information to the overall picture of the paper. For this reason I would suggest to either remove it or expand it with more detailed analysis. For example it would be worth investigating the possible precursors for HOMs formation. It may be possible to say something about the origin of the condensable vapours by looking at the concentration and diurnal profiles of biogenic vs. anthropogenic

VOCs.

- Section 3.2.4 should be written in a more consistent way, in particular the authors first speak of sub-25 nm particles but then they only shows bivariate plots of sub-4nm particle concentration, what is the reason for this? Moreover to better support the *airport hypothesis* it would be useful to show a time series of UFP for the period of interest together with wind speed and wind direction (extrapolating these data from the supplementary is difficult due to the long time period reported there). I'm asking this because the bivariate plots only show that UFP particle concentration is higher when the wind is coming from NE but to support the authors hypothesis it would be important to check if there are periods with low UFP concentration under the same wind conditions. If this is the case then I wound find the hypothesis less convincing due to the fact that the airport should be a more or less stationary source of UFP.

- In section 3.3 the authors often speak of "bottom-up flux" for the UFP however, what vertical profiles show is only that the UFP concentration is homogeneous inside the mixing layer. For this reason one should avoid using this terminology and the authors should correct all the corresponding parts in the paper. Moreover, I found that the interpretation of the vertical profiles graphs is complicated by the absence of a direct measurement of the mixing layer height. Referring to the "twin paper" [1], this information should be available for all the soundings (for example the potential temperature or total particle concentration can be used) and it would be a really nice addition to the graphs.

- The conclusions are written as a summary of the paper but here the author should focus more on the significance of their findings compared with existing observations. This section should be rewritten in order to convey a clearer message, so I suggest to the authors to delete all the unnecessary parts focusing more on the important results of the paper.

Minor comments

- Page2 line 3: "The NPF events extend over the full vertical extension of the mixed layer reaching as high as 3000 m." But the maximum height of the sounding is 2000 m, so this should be corrected.

- Page 2 line 4: "This can have consequences in the radiative balance of the atmosphere and affect the climate", the climatological effect of NPF in a polluted environment as Madrid is questionable and not supported by any evidence in this paper, for this reason remove or rephrase this sentence.

- Page 2 line 5: As previously stated, a proper estimation of NPF over primary UFP should be given here.
- Page 2 line 25: There is no need to cite 25 papers, this can be reduced.
- Page 3 line 13: The sentence seems to contradict the cited paper of Querol et al.[1] where it is said that "Relatively low concentrations of ultrafine particles (UFPs) were found during the study, and nucleation episodes were only detected in the boundary layer."
- Page 4 line 13: It would be useful to add a scale to the map in figure S1.
- Page 4 line 19: It is not clear if the PSM was operated in scanning mode or fixed mode, it would be good to specify this.
- Page 6 line 14: It is said that NPF was identified on 12 days but table 1 only reports 7 days so this should be made consistent. Moreover, figure S8 shows that there were 2 nucleation events at CIEMAT on the 13th and 14th of July that are not mentioned in the table. In addition, it would be useful to add also formation rates to the table together with the GR values.
- Page 6 line 27: It's almost impossible to see the early morning UFP concentration just by looking at the full time series. Thus a dedicated plot should be made either by plotting sub-10nm particles on top of figure 1 or by plotting a diurnal profile for this size range.
- Page 7 line 1: Particle size shrinking is an interesting phenomenon but it doesn't fit nicely in this part of the text. For this reason describe it in a separate (small) section. It would also be nice to plot the wind speed specifically for the shrinking phase because from the overall plot it is difficult to see a trend. In some cases also a rapid change in number concentrations is observed pointing to a change in air mass. In these cases it does not make sense to speak about a shrinking of aerosols because the aerosol population changes. The authors should demonstrate clearly, that the same population of aerosols is shrinking. Otherwise, it is not shrinking and they should delete this.
- Page 7 line 14: Here a reference to figure 5 would be useful. Moreover, I think that associating UFP with particles in the range 9-25 nm is misleading and it would be better to plot the diurnal profile for all particles below 25 nm and for the total particle concentration.

- Page 8 line 5: "above" should be replaced by "below".

- Page 8 line 11: "The fact that J9 is higher at the urban stations is probably linked to higher traffic emissions [...] in the city, and not related with higher nucleation rates, since PSM measurements indicate lower concentrations of 1.2-4 nm particles". Here the authors confirm my hypothesis that primary particles affect formation rates, underlining the necessity to take this process into account in their calculation.

- Page 8 line 14: This is not figure S4 but figure S7.

- Page 8 line 14: "The calculated formation rates agree with those reported in other studies, ranging 0.01-10 $cm^{-3}s^{-1}$ during regional events around the world." I don't see any reason for reporting an agreement within 4 orders of magnitudes. The formation rates measured during this campaign should be compared in a more targeted way with other locations around the world.

- Page 8 line 30: "Thus, the particles growth appears to be driven by the uptake of secondary organic compounds." This is a reasonable assumption but cannot be proven by the PTR measurements presented in this work. Try to support this assumption with additional information, for example one can try to check if the growth rates can be explained by sulfuric acid alone (assuming a reasonable range of values for sulfuric acid concentration) or not.[2, 3]

- Page 10 line 10: "the mode slightly decreases its size when the sounding ascends above the mixed layer limit", as already written above the mixing layer height should be plotted together with the particle size distribution to better visualize these changes.

- Page 10 line 12: It would be good if the authors could specify how they calculated the growth rate in the residual layer. The impressions from the graphs is that there are really few points inside the residual layer and it is not clear whether there is any growth at all inside this layer.

- Page 10 line 15: I'm not really convinced by the presence of a 10 nm mode in the first sounding, Maybe there is an over-fitting issue with the mode fitting algorithm. For this reason I think calculation of the growth rate is questionable and should be avoided.

- Page 10 line 23: "The accumulation mode grows from 156 nm at 07:00 UTC to 200 nm at 10:00 UTC", also in this case I think the accumulation mode is over-fitted, the authors should either revise their fitting algorithm or prove that I'm wrong by reporting in the SI a single SMPS scan plot with the fitted modes on top of it.

- Page10 line 26: "Another mode starting roughly at 40 nm at 09:00 UTC" I guess this should be 07:00 UTC.
- Page 10 line 30: I don't see any nucleation mode earlier than 09:30-10.00 UTC. Correct this sentence and eventually revise the calculated growth rate.
- Page 10 line 38: "As the insolation increased, so did the altitude of the mixing layer, until it reached the altitude at which the balloons were positioned." By looking at the plot it seems more likely that the balloon height decreased until reaching the mixing layer.
- Page 10 line 40: As previously explained avoid speaking of particles flowing upward, measurements are just showing that UFP are homogeneous inside the mixed layer.
- Page 11 line 5: I do not see a growth of 40 nm particles in the residual layer. The size of this mode is the same at the 9 and 11 UTC sounding.
- Page 11 line 6: how do you know that you observed these particles already the previous day? Moreover, also here I think that the Aitken mode is over-fitted.
- Page 11 line 16 and following lines: I would avoid speaking of the accumulation mode. I'm really sceptical about the presence of this mode in the measurements presented here and, even if it is present, then it is above the detection limit for most of the time. Moreover, it is said that the accumulation mode grew faster than the other modes and "this phenomenon has been rarely reported in ambient air." I think the data do not support this conclusion. If I'm not mistaken the fitted accumulation mode shows a growth only for a couple of hours on a specific day and the data are quite scattered so it doesn't seem like the growth is significantly higher compared with the Aitken mode. If the authors want to support this observation then they should try to look if anything similar is present in the SMPS ground measurements.
- Page 12 line 13,14: As already explained the vertical profiles do not show a clear accumulation mode and this is particularly true for the residual layer, so I would remove this sentence.
- Page 12 line 18: the authors don't need a miniaturized instrument with "greater resolution" but an instrument able to measure smaller particles.

- Figure 1: I would greatly recommend to avoid using jet colormap (*i.e.* rainbow colormap) for surface plots. This colormap is not perceptually uniform and this can create several kinds of issue as widely documented elsewhere (e.g. https://www.ncbi.nlm.nih.gov/pubmed/22034369 and http://ieeexplore.ieee.org/document/4118486/?reload=true)

- In figure S8 the authors report the total size distribution for the three measurement sites. This is a useful supplementary information but the readability of the graph should be improved. In particular a logarithmic color scale should be used as well as a higher image resolution. I also suggest to extrapolate the total particle number concentration for the 3 sites in the same size bins for better comparability rather than using different sizes for each site. Finally I noticed that there are some mismatches in the merged size distributions measured at CIEMAT (*e.g.* 8/7/2016). This would indicate that one of the instruments was not working properly. Please comment on this. How would this affect the presented results?

References

[1] X. Querol et al. "Phenomenology of summer ozone episodes over the Madrid Metropolitan Area, central Spain". In: *Atmospheric Chemistry and Physics Discussions* 2017 (2017), pp. 1–38. DOI: `10.5194/acp-2017-1014`. URL: `https://www.atmos-chem-phys-discuss.net/acp-2017-1014/` (cit. on pp. 3, 4).

[2] T. Nieminen et al. "Sub-10 nm particle growth by vapor condensation-effects of vapor molecule size and particle thermal speed". In: *Atmospheric Chemistry and Physics* 10.20 (2010), pp. 9773–9779. ISSN: 16807316. DOI: `10.5194/acp-10-9773-2010` (cit. on p. 5).

[3] Jasmin Tröstl et al. "The role of low-volatility organic compounds in initial particle growth in the atmosphere". In: *Nature* 533.7604 (2016), pp. 527–531. ISSN: 0028-0836. DOI: `10.1038/nature18271`. URL: `http://www.nature.com/doifinder/10.1038/nature18271` (cit. on p. 5).