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Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the spatial distribution and evolution of ultrafine aerosols in urban air" *by* M. Dall'Osto et al.

Anonymous Referee #1

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The manuscript presents results from several different stations in and around the metropolitan area of Barcelona, Spain over a period of about one month. The main data presented are total particle number concentrations, number size distributions and black carbon concentrations, which are used to estimate the main sources of particles in the area, distinguishing between regional and urban/traffic production.

The extent of the concurrent measurements is very impressive, and the data would merit publication in ACP. However, I have a few major concerns relating to the interpretation and presentation of the results that the authors need to address first. If they can do this, the manuscript can be considered for ACP. Detailed comments below.

Major comments

Data interpretation:

While the analysis of the presented data set is certainly challenging, I find that the authors have in some cases oversimplified and overinterpreted the data. My main concerns are listed below.

1. Event analysis.

While it is a pity that only one station had available SMPS data for the reported regional event, the data seem convincing. I would like to see a plot of the condensation sink (CS) during the measurement period. By looking at Fig. S3, the CS seems to be lower than normal during both the reported regional and urban events. Currently the CS is only discussed in a qualitative way.

The latter event which is reported to be an "urban“ event raises many questions for me. It is stated in the manuscript that in the afternoon the sea breeze causes the air to first pass over TM. Presumably the sampling there is then less influenced by the urban emissions than RS and UB. At least according to Fig. 1b, this seems to be the case. For the "urban“ event, the authors write that the event measured at TC and RB are transport from the urban area of Barcelona. If the wind direction is such that the air from Barcelona travels to RB, then I would expect that TM should not see much nucleation at all if the particles are formed in the city. Especially in the afternoon, the sea breeze should be causing cleaner marine air to impact TM, however, the highest particle number is seen at that time. How is this situation possible if the particles are formed in the city? Are there WD data available to show in which direction the plume would be expected to move during the event?

Even more importantly, the authors suggest nucleation occurring in the city, which would function as a point source. The non-growing modes at UB and RS would agree with this, but the growing modes at TC and RB suggest to me that something else is causing the particle production and growth. But to keep with the idea of urban nucleation, the growth of the mode should be related to changes in available condensable

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vapors between nucleation (e.g. RS) and detection (e.g. RB), though this might be challenging. The change of the modal size at TC and RB should definitely not be directly interpreted as the GR of the nucleating particles, as done now, if the NPF is not assumed to be a regional event. In summary, if the authors wish to show that this event was an urban event, they need to provide a self-consistent explanation of the area where particles would be produced, in which direction the wind transported the particles, and what do the growth rates need to be to explain the particle sizes observed at the different locations. E.g. why is the difference between UB and TC of the same order as the difference between TC and RB if the growth is 2-3 nm/h at both TC and RB? And how can there still be (growing) new particles visible at 23:00 in TC? That suggests to me that the particle production during this day was occurring in a larger area than just the city.

2. Vertical extent of particle formation.

The authors need to provide more evidence that the height of the TM and TC sites are more important than the geographical locations. Currently, small changes between TM/TC and RS/UB are interpreted to show changes in the vertical extent of particle production. Perhaps the authors have data to back these claims, but based only on the data shown, I would expect a much larger impact from the different locations in the inhomogeneous metropolitan area. Also used to show how particles are formed at higher levels, on page 16624, lines 10-16, the authors state that the points under discussion in Fig. 6 occurred on 3 days with possible NPF, and that particles were seen at the tower sites but not at ground level (UB and RS). When I look at the plots in the supplementary material, for RS CPC data is lacking for all three days, SMPS data for two of them. For the UB site, SMPS data is also lacking for the first two days, and for the third the SMPS shows a concentration and distribution similar to the "urban" event. Furthermore, the CPC counts at UB according to Fig. S2 are the same or higher than for both TC and TM on all the three mentioned days! The authors need to clearly show how they came to the conclusion that NPF is only seen at tower level and not

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at ground level, as this would have been a very surprising observation since the tower sites themselves are not very high above ground and the boundary layer is expected to be relatively well-mixed.

3. Interpretation of TM in Fig. 8.

The authors state in the text "leading us to suggest some particles may evaporate (leading to a lower N/BC) when the atmosphere starts to heat up in the morning. In other words, a reduction of the ratio N/BC (07:00–10:00 at TC and TM, Fig. 8) can be explained by some semi-volatile particles evaporating when the urban heat begins to develop. During afternoon, more particles are seen in both towers, higher than at the ground site. This is an indication that nucleation processes are taking place above the city in the afternoon at higher intensity than ground level." If I compare Fig. 8 to Fig. 1, the above text makes no sense. The reason for the large variation in N/BC is clearly due to changes in BC and not N, and there the BC trend seems consistent with the seabreeze behavior, decreasing in the afternoon. The particle number at TM never exceeds UB or RS in Fig. 1, yet the authors write that more particles are seen at TM and use this as an indication that particles are produced above the city!

Structure of the paper:

There are a lot references to other work in all sections of the manuscript. For introduction, methodology and discussion sections this is of course fine, but also the results and conclusions sections bring up results from other work, sometimes with unclear references, which makes it very confusing for the reader to be clear about which results are new and which old. As an example, while explaining results in the abstract, it is stated that "This is also evident in the urban background annual mean diurnal trend of N/BC, showing a midday peak in all seasons", making it sound like a result from this study, although the reported measurements only cover one month. The text should be clarified throughout the manuscript, so the reader can easily distinguish which results are from this study, and which are from already published work.

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Other comments:

Grammar: There were sections of the text that would benefit from a read-through of one of the native English speaking co-authors, and I recommend that the authors do this for any future versions of the manuscript.

Abstract: I feel the abstract is longer than necessary. Some better restructuring of the paper and the main results could also lead to a more condensed abstract. Additionally, the abstract starts by saying that <100 nm particles were investigated, but later on concentrations up to 500nm are discussed.

Page 16606, line 15 and page 16607, line 1: "Little is known about the vertical distribution of primary and secondary UF particles in the urban atmosphere“ and later "Even less is known about the vertical distribution of secondary new UF particles, especially in the urban atmosphere.“

Title: I would recommend adding "in Barcelona, Spain“ to the end of the current manuscript title.

16612, 2: Although certainly within the error bars, do not use "same order“ when the order is not the same.

Fig. 6. There is speculation about the temperature effect on N/BC, but I would have assumed that if the higher N/BC ratio is due to nucleation, then radiation would be more important. The RS data set is the one that shows the best rainbow in Fig. 6C, but there is not much of a rainbow in Fig. 6d for RS, which is surprising. However, when I look more closely at Fig. 6d, the coloring of the points in RS seems to be completely different from the other three plots in 6d. Perhaps there was some mistake in the plotting?

Fig. 7: It is stated on page 16624, line 1 that the intercept is interpreted as the regional contribution, but as TC is much lower than the others, this cannot be true.

Table 2: The table says "cP air masses“ twice.

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Interactive Comment

Fig 5.: The layout of this graph could probably be improved.

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