Review of P. Crippa, et al. "Evidence of an elevated source of nucleation based on model simulations and data from the NIFTy experiment"

The present study reports measurement and model results pertaining to new particle formation at a forested site. The focus of the work is a two week period during which 6 'Class A' nucleation events (i.e., strong nucleation and clear particle growth) were observed. The Class A events exhibit similarities in meteorological characteristics, highlighted by the breakup of a nocturnal inversion prior to the detection of the newly formed particles. The measurements are combined with box modeling results to infer that new particle formation during Class A events occurs at some altitude above the nocturnal inversion - likely in the residual layer. Vertical mixing driven by solar radiation causes the inversion to break up and the new particles to mix downward where they are then detected by instrumentation positioned near the surface.

The work is original and is within the scope of ACP. If true, the observation of nucleation aloft would be a meaningful addition to the large body of work on atmospheric new particle formation. The authors state clearly that their hypothesis of new particle formation occurring at an altitude above the nocturnal inversion is only an inference based on surface measurements and box modeling results. However, I think that there are weaknesses in both the interpretation of the measurement data and in the model results. In my opinion, further analysis is needed to justify publishing this hypothesis.

General Comments:

One of the major pieces of support for the hypothesis of nucleation aloft comes from the modeling results. Specifically, the base model simulations systematically underpredict both the nucleation rate and the growth rate. Model simulations improve when the initial particle size distribution is set to clear atmospheric conditions rather than the observed surface particle size distribution. However, I worry and suspect that a potentially major source of model error stems from the inaccurate treatment of organics. Even using observed [H_2SO_4], the model systematically underpredicts particle growth rates. Table 2 shows that all organics are treated with a common rate constant for reaction with OH: 1.53×10^{-1} ppb^{-1} sec^{-1}. This is an appropriate rate constant for OH reactions with the anthropogenic species as it is approximately equal to the rate constants for OH reaction with toluene and ethyl benzene. It is at least an order of magnitude too low for the rates of OH reaction with the biogenic species. From the Pryor et al. (2011) work that is cited, it appears that the concentration of biogenic species is at least a factor of ~2 higher than the anthropogenic compounds (this makes sense at a forested location). Thus, with a reaction rate constant that is ~10-20 times too slow, it appears the model is significantly underestimating the potential importance of condensable organics to new particle formation and growth at this location. Also, from the Prior et al. (2011) work, it appears that α-pinene was the most abundant individual organic compound measured: the α-pinene-O_3 reaction should also be

treated in the model since α-pinene has a similar lifetime due to reaction with O_3 and OH at typical atmospheric levels (Atkinson and Arey, 2003). Organics have shown to be important contributors to new particle formation and growth at many locations (e.g., Laaksonen et al., 2008; Smith et al., 2008; Riipinen et al., 2011). In particular, the Laaksonen et al. (2008) study showed that a nucleation event strongly influenced by organics began at the surface and the newly formed particles subsequently mixed upward with the breakup of the inversion. On Pg. 11984, ln. 22-26, the authors acknowledge the limitations of the model treatment of organics. I think that this is a significant shortcoming that has potentially influenced fundamental results in the paper and thus further analysis is required. Perhaps a more accurate treatment of the organics (more accurate OH-organic rate constant and inclusion of the α-pinene-O_3 reaction) will better reconcile the observed nucleation rates and growth rates – even with the observed near-ground particle size distribution. If not, then the argument of a nucleation source aloft is made even stronger.

The comment posted by Kari Lehtinen and Ari Laaksonen should be directly addressed. If the results presented in Fig. 7 have been wrongly interpreted, this may affect the hypothesis as well, since this was presented as evidence from the measurements that nucleation occurred aloft (pg. 11996, ln. 5-8).

Specific Comments:

- In general, significantly more detail is needed in the experimental section: i.e., how was the SMPS operated? What were the size ranges of the SMPS and FMPS systems? There is no detail at all given on the VOC measurements. One should not have to consult the Pryor et al. (2011) paper for basic measurement details. The experimental section makes no mention of an OH measurement, but an observed value is given on Pg. 11994 ln. 13. No mention of the NH_3 measurement in the methods section…etc.
- Pg. 11993, ln. 5: "underestimation of nucleation intensity" - Tables 1 and 5 do not seem to agree: e.g., for Day 17, Table 1 gives an observed nucleation intensity of 1.06×10^5 cm^{-3}, and the simulated nucleation intensity using the measured PSD is 3.82×10^4 cm^{-3} – so the simulated-to-observed ratio is 0.36. In Table 5, it lists the simulated/observed ratio using the PSD initialization as 1.94? Overall, Table 5 suggests the simulation initialized using the measured PSD overestimates the nucleation intensity in 5 of 6 Class A events. This should be clarified.
- Figures 9, 10, and 12 need significant improvement: consider making stacked plots, with 'a' on top of 'b', and each figure having at least double the horizontal width of the current figures.

- From what I can tell, all of the data presented in the figures and all of the discussion centers on the MMSF site – why then does Figure 5 include data from all three sites? Data from the other two sites should be excluded.
- Why was the analysis presented in this study limited to only two weeks of the measurements at one of three sites? Why not at least use data for the entire study period?

Technical Corrections:

- Figure 6 panels are mislabeled (compared to Fig. caption)
- Pg. 11981, ln. 16-17: no need to quote directly from the Wehner et al. (2010) study – paraphrase their results and include citation.
- Pg. 11995, ln. 12 – delete: 'mostly European' - the Pierce et al. (2012) study is from North America.
- On four occasions in the paper, the Pierce et al. study is wrongly cited as 2011 – should be 2012

References

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