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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of multiple new-particle growth pathways observed at the US DOE Southern Great Plains field site" *by* Anna L. Hodshire et al.

Anonymous Referee #1

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This article presents an important study of growth pathways of nano-particles, comparing field measurements with results from process modeling. It is well writing and falls within the scope of the journal. While both modeling uncertainties and lack of some key measurements do not allow for closure, uncertainties and limitations are fully acknowledged and discussed. This discussion of limitations is an important aspect of the article, identifying key unknowns holding back out current understanding of nano-particle growth and signaling ways forward for future work. I recommend this article for publication in ACP with minor revisions.

Scientific Comments

1. GR methods

Page 12 lines 1-5: There are many methods that can be used to calculate GR from measured size distributions. Why is this method used as opposed to e.g. continuous dDp/dt using the mode diameter or using the leading edge of the banana instead of the mode diameter? An evaluation of how the assumptions in this method chosen compare with the way MABNAG calculates GR would show whether any of the systematic discrepancy between measured and modeled GRS is due to method differences instead of missing species as implicitly assumed in later sections. Part of the consistent under prediction of GR from MABNAG compared to these measurements could be due to systematic differences from the method of GR calculation. MABNAG measures wet diam. Temperature of saturators in CPCs of the SMPSs could mean measuring dry diam. For the compositions observed do you have an estimate of how much this could be affecting the particle diameters and thus GRs?

2. GR uncertainties

Page 12 paragraph 2: This range of GRs given by the 3 diameter ranges is not a measure of uncertainty, but is presented almost like an uncertainty in the results and evaluation section. Table 3 would be more useful if it showed which GR comes from which diameter range. Is there any trend in how GR varies over the diameter range? A more continuous dDp/dt plotted as a function of Dp would give a clearer view of this and could be usefully compared with how the MABNAG GRs vary with size and an experimental view on whether the reduction of the Kelvin effect with size (line 15) is a significant contributor or not.

Page 15 line 5: 50% MABNAG GR uncertainty from ELVOC concentration uncertainty would lead to a maximum GR of 2.1nmph, which is still lower than the measured 3nm GR. It needs to be stated clearly that this uncertainty in the ELVOC concentration cannot alone account for the model-measurement discrepancy in this case.

Page 15 line 9 and lines 16-19 : If LVOC and SVOC contribute more to growth as the particle size increases we would expect the modeled GR to deviate more from

the measured result at larger sizes, especially for the case where organics dominate growth. Is this apparent in the data?

Page 15 line 26: What is the uncertainty on the cluster CIMS SA measurement and also for the ammonia and amine concentrations on page 16 line 35?

Page 18 lines 27-36: RH uncertainty on MABNAG GR would be more usefully included quantitatively in the discussion of GRs for the separate case. This and the ELVOC concentration uncertainty (put at 50% earlier in the manuscript), the oxalic acid factor 100 uncertainty and perhaps other sources of uncertainty could allow a fuller basis for comparison between model and measurements if they were included quantitatively in the results.

Page 12 lines 5-10: Hypothesis that double nucleation is mixing of nucleation event that occurred higher up + later event lower down seems plausible for May 9th, but May 11th both events show particles growing from the smallest sizes. The plots in fig1. are composite plots from multiple SMPSs – could the larger concentrations at larger sizes on May 11th 1st nucleation event be a compatibility issue between SMPSs (i.e. could the SMPS measuring at larger sizes be measuring with a higher efficiency – perhaps because of unaccounted for diffusion losses in the sampling lines?)

May 11th 2nd nucleation event: condensation/coagulation sink from 1st nucleation event will likely be affecting the GR making it appear smaller than it would be for a single event – some estimation of the size of this effect would be useful as it would further increase the difference between the measured GR and the MABNAG modeled GR

3. Other

Page 7 paragraph 4: Some discussion on the accuracy of MEGAN2.1 estimations of monoterpine emissions and concentrations and applicability for this site/study is necessary to give confidence in their use.

Page 9 paragraph 1: What was the basis for choosing 20 molecules as the initial particle size? How does this choice affect the modeled results?

Page 12 line 5: On what is the assertion that nucleation potentially occurred aloft based?

Page 16 line 18: GR for ammonium sulfate case shows better model-measurement agreement than for the growth by organics case. SA/amine/organic growth on p17 also shows better agreement. Comment on why this might be?

Page 17 line 16: The base simulation predicts GR that are way too low compared to measured GR. Therefore the relevance of the composition from this model seems tenuous. In general, different MABNAG simulations have better/worse agreement with the measured GR and also predict different compositions – more could be made of which compositions are more likely to be accurate based on this.

Page 17 line 18: This sentence should include the fact that there are significant additional unknown growth pathways (N, LVOC, SVOC) as well

Technical Comments

Page 9 line 21: SVOC doesn't seem to be defined here or earlier and should be

Page 7 lines 10-11: "estimated uncertainty in oxalic acid . . . is approximately a factor 100 lower" is unclear. Need a better way of saying the oxalic acid concentrations could be up to 100x larger than measured as done later on page 11

Page 13 line 16: Reference needed for sulfuric acid concentration of 2e-6 leading to 0.2nmph growth rate

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