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 #4

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This is a very nice paper that combines measurements with modelling. It is based in a technically challenging frontier of aerosol science: determining and verifying the composition of the smallest nucleating particles. It is generally well-defined and clearly written. The authors acknowledge the limitations of their methods and describe the approximations and assumptions they have relied on.

My major concern in this paper relates to the way in which the findings are communicated to the reader. Rather than having a huge data dump, it would be better to both group and separate the scenarios/simulations more clearly. Some methods are suggested below:

In Tables 4-6, for each group of three simulations, the first and last identifiers are constant and the middle one changes. Either change the order of the identifiers or the order in which the simulations are presented. I think it would be clearer to have all the MAL simulations first, divided into thirds by 1ox/10ox/100ox, each of which is further divided based on DMA_L and TMA_L. After that, a similar breakdown of MAL_LoVP, then OX, then OX_LoVP. This would give a logical progression, while still leaving the reference scenario in second place on the list. However, the authors may prefer to use another system based on what they feel is the most important characteristic to group simulations for important comparisons. Based on my reading, it seems that the characteristics change from case to case, and so the ordering may be less important from that perspective.

We have changed Tables 4-6 to Figures 3, 5, and 7. Figures 3, 5, and 7 display both the mole fractions and mass fractions of each species' contributions to the particle for each case. This more clearly communicates how the relative amounts of each species may or may not change across different assumptions. We have further grouped all malonic cases together and all oxalic cases together, instead of grouping MAL/OX then MAL_LoVP/OX_LoVP cases together.

Add vertical lines between the case identifiers and growth rates, and between growth rates and mole fractions.

ACP provides strict formatting guidelines for tables; unfortunately, “Vertical lines must be avoided”. However, we have translated these tables into Figures 3, 5, and 7, thus removing the tables entirely.

Please colour code the simulations which differ significantly from the base case, or which provide the best reproduction of observations; at the very least, those which are discussed in-depth in the text. Refer to the cases by colour in the figure caption, for ease of understanding.

Deviations should hopefully be more apparent with the new figures (Figures 3, 5, and 7).

Add explanatory text to the captions of Tables 4-6. As a general rule, a caption should provide enough information that the item can be at least minimally understood without any other context.

Tables 4-6 have been replaced with Figures 3, 5, and 7. We believe that the captions provided for each of these figure gives a clear explanation of each case and case identifier.

Using T to represent Total amines measured at SGP is quite confusing in the text, because it usually means Temperature (as it does in Table 1, for example). Maybe use TAm?

We agree and have changed all instances of T and L to Tam and Lam in the text and figures.

Use the format A.B × 10C in e.g. Table 1. ($\times$ in LaTeX, Insert →Symbol →× in MS Word)

Your suggested format is correct under ACP guidelines. The numbers have been reformatted in both the tables and text.

In Figure 1, please add units to the colour bar for panels (a)-(c) (and it would be better to label the colour bar as “log N”, or to only label whole powers of ten).

Done.

In panel (f) of Figures 2-4, ELVOCs and organic acids seem to be the same colour. Is this intentional? The mole fractions are shown separately in Tables 4-6, so I assume they can be distinguished at all.

The ELVOCs are a dark green color, and the organic acids are a bright lime green color. There is so little organic acid in the particle phase throughout the shown MABNAG simulations that it cannot be seen for Figure 2 or 6; the predicted organic acid mole fraction is slightly distinguishable in Figure 4.

I would like to see an explicit equation for "the same calculations as used for April 19". I'm more of a physicist than a chemist, and while I tinkered around with the numbers in some of the tables, I couldn't reproduce 12.5% by mole. Of course, there were quite a lot of scenarios in the tables, so it was hard to be sure exactly which numbers I was meant to be using…

We have determined that our calculations for the formation of organic salts are tenuous, given the many uncertainties associated with the organic acids as model inputs (e.g. concentration and chemical properties uncertainties). It is clear that (excluding the few unrealistic cases in which organic acid dominated the particle growth and particle growth rates exceeded 40-50 nm hr^{-1}) organic acids tend to contribute very little to the particle on both a molar and mass basis (see Figures 3, 5, and 7). Thus, we have removed the detailed discussions upon the possible numerical upper bounds of organic salt contribution to particle growth for each day and instead have made note of the small contributions of organic acids to particle growth and thus small contributions of organic salts to particle growth for each case day. For April 19, we state, "The majority of our simulations predict that less than 1% of the particle is organic acid by mole; thus, the contribution to particle growth from organic salt formation would be negligible, even when including the contribution from associated bases. Thus, we expect the majority of growth from organics to be coming from non-reactive organics (ELVOCs in our simulations) for this day." For May 9, we state, "However, given that most cases predict negligible (<3% by mole) of the particle to be composed of organic acid, the contribution to particle growth from organic salt formation is still predicted to be minor for this day." And for May 11, we state, "The majority of our simulations predict <5% by mole of the particle to be organic acid, thus again leading to only minor contributions from organic salt formation to particle growth."

The growth rates listed in Table 3 show a single number, whereas the text references three different numbers for each day. It would be better to see those numbers explicitly rather than be given a range.

We have revised our growth rate estimates for this work. We have decided to use three different methods of determining the growth rate: the leading edge method, the Dp mode method, and a visual method. We have inserted the following into the discussion on calculating observed growth rates:

"There is considerable noise in the SMPS data (Figure 1, a-c), especially for May 9 and May 11, due possibly to the hypothesized mixing down of particles and possible inhomogeneities in the air mass. For this reason, we have calculated the growth rate between 10-20 nm for each using three different methods. The first method, referred to here as the leading edge method, is adapted from Lehtipalo et al. (2014) and finds the time at which the binned aerosol distribution between 10-20 nm reaches one half of its maximum $dN/dlogD_p$ for each bin. A linear fit between the bin's median diameter and the associated time determines the growth rate. The second method, referred to here as the D_p-mode method, tracks the change in diameter of the maximum $dN/dlogD_p$ of the aerosol size distribution between 10-20 nm; a linear fit between the diameters and time determines the growth rate. When plotted against the size distribution (see supplement, Figures S1-S3), it is seen that the leading edge and D_p mode method both do not always track the growing size distribution well. For this reason, we have included a third method, which we call the visual method, in which we have made a linear growth rate between 10-20 nm for each day based upon visual inspection of the size distribution (see supplement, Figure S1-S3), using Eq. (3):

$$GR_{obs} = dDp/dt \sim = \Delta Dp/\Delta t \qquad (3)$$

These three methods provides a range of growth rates (Table 3) for the particles between 10-20 nm; the specific results for each day will be discussed in section 3. We do not attempt to provide uncertainty estimates for each method, due to the overall noise in the data. Instead, we present the ranges of calculated growth rates as a possible range of the actual growth rates. May 9 and May 11 tend to have higher growth rates: this could be from the influence of the continued mixing down from nucleation aloft and not actually representative of the growth rates of the particles forming near the surface."

We have also included in the supplement a figure for each day that shows the results and best-fit lines of these three methods, included below.

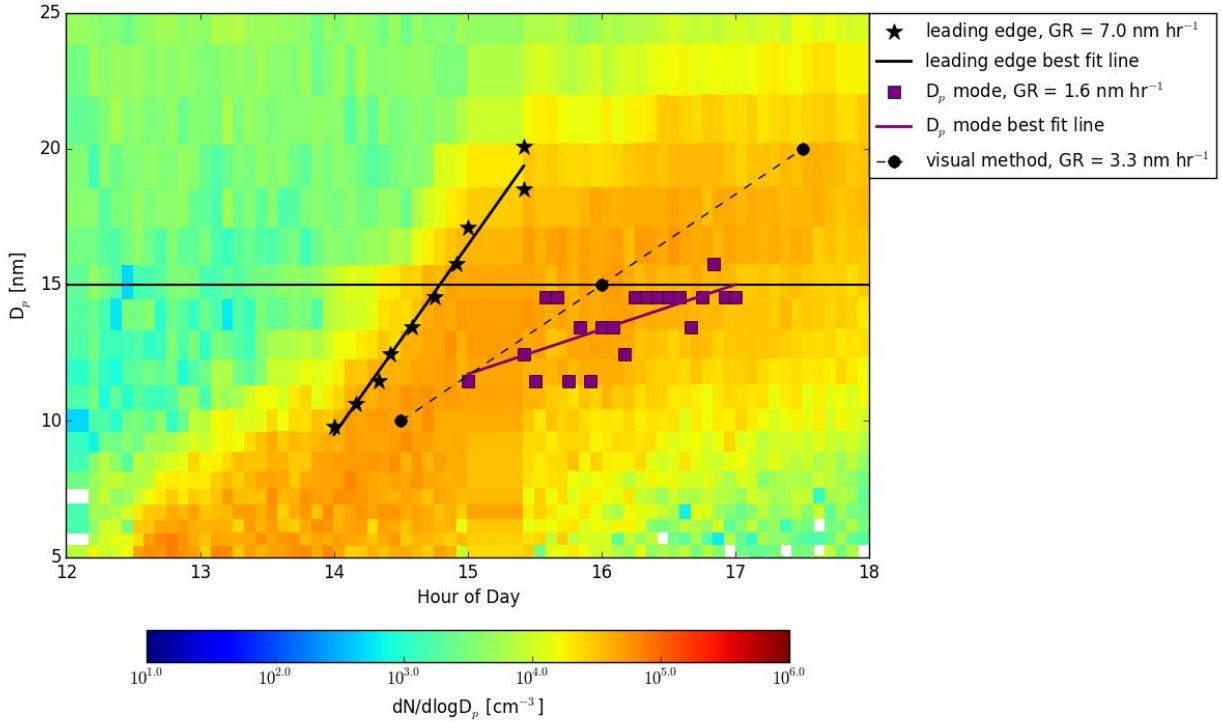

Figure S1. The results of the three growth rate calculations for April 19, 2013. The x-axis represents CDT time. The line at 15 nm D_p is to guide the eye.

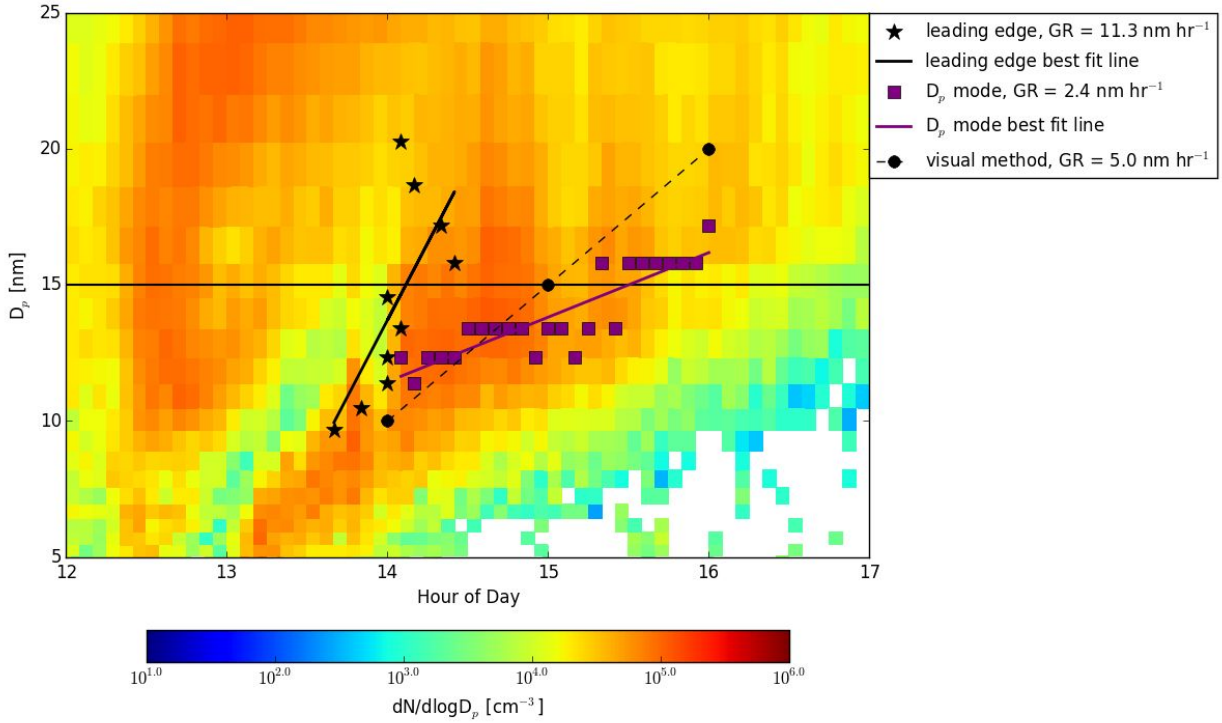

Figure S2. The results of the three growth rate calculations for May 9, 2013. The x-axis represents CDT time. The line at 15 nm D_p is to guide the eye.

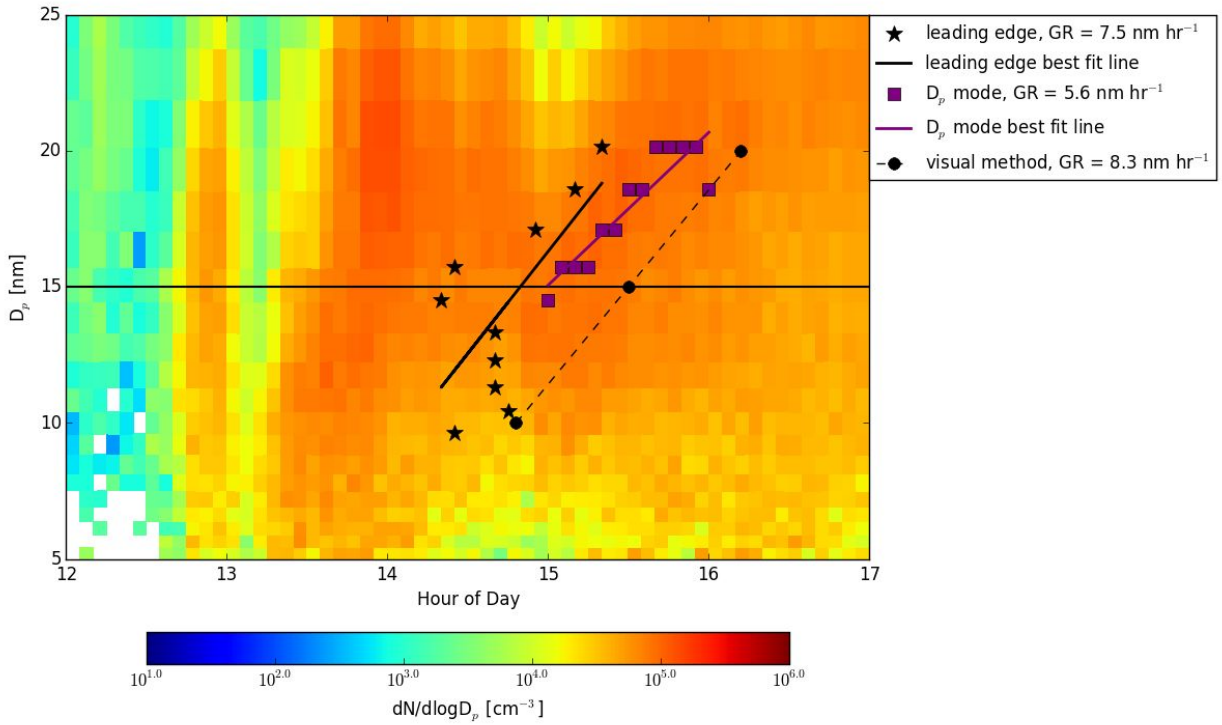

Figure S3. The results of the three growth rate calculations for May 11, 2013. The x-axis represents CDT time. The line at 15 nm D_p is to guide the eye.

Aside from these minor concerns, I found the paper interesting and feel that it makes an important contribution to its field. I would recommend that it be published subject to minor revisions.