Interactive comment on “Direct contribution of ammonia to CCN-size alpha-pinene secondary organic aerosol formation” by Liqing Hao et al.

Anonymous Referee #2
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This manuscript reports chamber results of alpha-pinene SOA formation in the presence of ammonia, and suggested that organic acids have a central role in the formation of particle phase ammonium through neutralization. Major arguments supporting this conclusion come from the stoichiometric neutralization analysis, and correlations between gas-phase organic acids and particle-phase NH4+. The experiment looks carefully conducted and calibrated, with interesting results. However, the interpretation of the results is ambiguous and confusing. Although I trust in the data, I’m not convinced by the logic that draws to the conclusions. The following concerns should be addressed before this work can be considered for publication for ACP.

Major concerns
1. How did the authors estimate the NH3 concentration in the chamber “by assuming that the particulate ammonium salt (NH4+) was converted from the gas-phase NH3 (Fig. 1)”? Did the authors assume a 1:1 conversion ratio, namely all gas-phase NH3 are converted to NH4+? If so, this assumption is too arbitrary as NH3 is highly volatile. If the authors are introducing the NH3 from the “background gas”, I’d suggest at least an estimate of the typical NH3 level (e.g., through a supplementary measurement of typical background NH3 concentration in your lab, or provide the measured NH3 level in the vicinity environments), and do a calculation to check whether all the NH3 would have been depleted by the organic acids at your measured concentration level.

2. What on earth is the definition of CCN size in this paper?

3. The results seem to suggest that the measured NH4 concentration is more than enough to neutralize the inorganic acids (SO4, NO3 and Cl) (Eq. 1), while not enough to neutralize all organic acids (CO2+), as “the amount of CO2+ required for neutralizing ammonium accounted for the 27.0 ± 3.1 % of total CO2+ mass in the nucleated SOA experiments, and 18.7 ± 6.0 % in the seeded SOA experiments”. If so, then ammonium would be the limiting species, the concentration of which should be determined by available total ammonia concentrations. In this case, it is confusing why the NH4+ would correlated to gas-phase organic acids. If the authors don’t really think the CO2+ all comes from organic acids, it should be clarified in the manuscript.

4. Following above question, the good correlations among gas-phase organic acids and NH4+ should be expected if the gas-phase organic acids are the limiting species, with total ammonium more than enough to influence the partitioning equilibriums. The results seem self-conflicting.

5. The authors argue that “The reaction of HNO3 and NH3 takes precedence over the reaction between organic acids and NH3” (Line 186). Fig. 1 also shows formation of sulfate and nitrate from photooxidation of SO2 and NOx. If so, the influences from inorganics should be first excluded in analyzing the correlations of NH4+ and gas-phase organic acids (Fig. 5). Instead of AMS NH4+, the difference between measured
NH4+ and that predicted by stoichiometric neutralization analysis of inorganic acid (Eq. 1) should be used (i.e., Free NH4+ = NH4+,meas - NH4+,pre). This is similar concept with the correction of (NH4)2SO4 for the seeded experiment in Fig. 5. How would the correlation look like after this kind of correction?

Minor concerns

1. There're some typos in the manuscript. For example, Line 59, “updake” should be “uptake”. Line 131, “aftert” should be “after”. Line 135, there's duplicate periods. The manuscript should be read through more carefully.

2. I'd suggest name the predicted NH4+ differently for that based on Eq. 1 and Eq. 2, i.e. that predicted with / without consideration of CO2+.