Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-454-RC3, 2017

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Hygroscopic behavior and chemical composition evolution of internally mixed aerosols composed of oxalic acid and ammonium sulfate" *by* Xiaowei Wang et al.

Anonymous Referee #3

Received and published: 26 June 2017

The authors presented a laboratory work on the hygroscopic growth and phase transitions of oxalic acid (OA), ammonium sulfate (AS), and their mixed particles. The growth factor and the phase transition of deliquescence and efflorescence were determined using the spectra collected by confocal Raman spectroscopy at room temperature. It is showing that the particles with different mixing ratios showed different hygroscopicity during the hydration and dehydration cycles. At higher OA/AS ratio, the dehydration process produced less hygroscopic organic salt, such as NH4HC2O2, from in particle phase reaction within the aqueous droplet as it loses water. In addition, the manuscript shows the possible effects on the growth factor by the different drying rates. The manuscript also provides explanation for the discrepancy on the ERH of

OA compared to the previous studies. This study provides a set of valuable data for the hygroscopicity of model particles generated in the lab. This work demonstrates the effects of aqueous phase reaction on particle hygroscopicity during dehydration which was overlooked in the past. There is a quite important implication to atmospheric chemistry. It is recommended for publication after a minor revision. Please see the following comments which the authors may want to consider in the revision.

Minor comments:

1. P1, L17, L18, “aerosol” refers to the mixture of particle and gases. It is suggested to change the “aerosol” to “particle”.

2. P1, L28, how do you define “the partial deliquescence relative humidity”?

3. P3, L23, this statement is not clear, in the previous sentences the authors showed that there are several studies on the OA/AS system. Please provide additional information or references to support this statement.

4. P4, L19, it is not clear how the authors would be able to prepare the 30-40 um aqueous particles with a syringe.

5. P4, L25, If the temperature accuracy is 0.7 K, the uncertainty of RH at 297 K and 95% should be 4%. How the sample temperature is controlled during the experiments?

6. P5, L25, I also suggested to move the Raman spectra to the main text.

7. P6, L11-12, it is not clear to me that how oxalic acid dihydrate can be converted to anhydrous form at these experimental conditions? How long it will take for such process and is it atmospheric relevant?

8. P7, L10, it is the reaction, not an equation.

9. P9, L6-11, the explanation for the discrepancy on the ERH of OA compared to the previous studies should be carefully addressed.

10. P11, L10-12, as suggested by the previous reviewers, it may be more straightforward if the authors can provide optical images to show the phase transitions. For this possible evidence on the coating of less hygroscopic materials, it may be easy to just provide Raman spectral at different location of particles or compositional mapping with the imaging mode.

11. P11, L28-29, it is not clear how the RH is controlled during the 10-12h experimental period, stepwise or continuously? What is the variation of sample temperature during this period?

12. Figure 4 and 5, It is suggested to compare the experimental results with model estimation, such as E-AIM, ZSR, or AIOMFAC. For example, the E-AIM model (http://www.aim.env.uea.ac.uk/aim/aim.php) includes the dissociation equilibrium for some organic/inorganic systems. The oxalic acid is included in current E-AIM. What would E-AIM predict and how does that compare with your experimental data? This can not only serve as validation of the determined Raman growth factor but may also provide additional insides to the effects of reactions on particle's hygroscopicity.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-454, 2017.

Printer-friendly version

Discussion paper