

Reviewer2: The authors describe the effect of dry and wet deposition on the mixing states of organic/NaCl particles using atomic force microscopy and bulk ensemble HPLC. The result can provide additional insight for the mixing state of aerosol particles which is still unknown in the atmospheric community. However, some results are not convincing and need more explanation.

Authors: We thank the reviewer for their time and diligence. We carefully addressed all of the specific questions and concerns raised by the reviewer, which is shown below. This reviewer expresses genuine support of our work. The comments improved the quality of the manuscript.

Reviewer2: 1. Title should describe the topic and key words of the paper. 'Dry versus wet' is too simple. Also 'Implications' is not a main content of this paper. Please reconsider the title.

Authors: Thanks for the comment. The implications relate the dry or wet nature of the particle deposition to the measured organic volume fraction. In consideration of this, we changed the title to the following, "Effect of dry or wet substrate deposition on aerosol organic volume fraction"

Reviewer2: 2. The authors selected glucose-NaCl mixtures as a model system consisting of a coreshell morphology. I am confusing whether the mixture really showed a core-shell morphology. Previous studies have established that particles containing organic/inorganic salts underwent phase separation when the O:C of the organic material was smaller than 0.8 (Bertram et al., 2011; Krieger et al., 2012; Song et al., 2012; You et al., 2013; You et al., 2014). Since the O: C of glucose is 1.0, I expect the particle is present in one phase before NaCl effloresces. Once the particle reaches low relative humidity, it would show that nucleation of NaCl initiates efflorescence process over the particle. The authors should state the discrepancy in the previous work in detail.

Bertram, A. K., Martin, S. T., Hanna, S. J., Smith, M. L., Bodsworth, A., Chen, Q., Kuwata, M., Liu, A., You, Y., and Zorn, S. R.: Predicting the relative humidities of liquid-liquid phase separation, efflorescence, and deliquescence of mixed particles of ammonium sulfate, organic material, and water using the organic-to-sulfate mass ratio of the particle and the oxygen-to-carbon elemental ratio of the organic component, *Atmos. Chem. Phys.*, 11, 10995-11006, DOI 10.5194/acp-11-10995-2011, 2011.

Krieger, U. K., Marcolli, C., and Reid, J. P.: Exploring the complexity of aerosol particle properties and processes using single particle techniques, *Chem. Soc. Rev.*, 41, 6631- 6662, 10.1039/c2cs35082c, 2012.

Song, M., Marcolli, C., Krieger, U. K., Zuend, A., and Peter, T.: Liquid-liquid phase separation in aerosol particles: Dependence on O: C, organic functionalities, and compositional complexity, *Geophys. Res. Lett.*, 39, Art. L19801, Doi 10.1029/2012gl052807, 2012.

You, Y., Renbaum-Wolff, L., and Bertram, A. K.: Liquid-liquid phase separation in particles containing organics mixed with ammonium sulfate, ammonium bisulfate, ammonium nitrate or sodium chloride, *Atmos. Chem. Phys.*, 13, 11723-11734, 10.5194/acp- 13-11723-2013, 2013.

You, Y., Smith, M. L., Song, M. J., Martin, S. T., and Bertram, A. K.: Liquid-liquid phase separation in atmospherically relevant particles consisting of organic species and inorganic salts, *Int. Rev. Phys. Chem.*, 33, 43-77, 10.1080/0144235X.2014.890786, 2014.

Authors: This is an interesting comment. To begin, the phase separation/ core shell morphology we observe is solid-semisolid phase separation, not liquid-liquid phase separation. This is evident in Figure 1,

where phase images clearly identify the solid core surrounded within a shell. This solid NaCl is also evident in Figure 3. Moreover, we are not aware of AFM phase imaging capable of identifying liquid-liquid phase separation.

The reviewer's comment that the glucose + NaCl mixture, above the deliquescence point is likely not liquid-liquid phase separated, is reasonable. In this work, we do not claim that this methodology applies to the glucose + NaCl mixture beyond the deliquescence point at relative humidity greater than 75%. Below the efflorescence point, however, the NaCl would indeed become a solid, which is again, evident in Figures 1 and 3. Therefore, we do not see any discrepancy to note, as the cited works pertain to different type of phase separation.

That being said, publications cited by this reviewer are credible works that studied liquid-liquid phase separation, and any that were not previously cited, are now cited within this manuscript. To the last paragraph of the introduction, we added,

“Unlike liquid-liquid phase separation, to the best of our knowledge, parameterization to predict a solid-semisolid or solid-liquid phase separation does not yet exist.(Bertram et al., 2011;You et al., 2013;You et al., 2014;Krieger et al., 2012;Song et al., 2012)”

Reviewer2: 3. Section 2.3. Please add information of the capacity for the size resolution of the AFM.

Authors: Good point. We added in Section 2.3 AFM imaging,

“The microscope permits (sub)nanometer-level spatial resolution and 1 pN force resolution. (Binnig et al., 1986;Santos et al., 2011;Gan, 2009;Gerber, 2017)”

Reviewer2: 4. Experimental (Sect. 2.3, line 10) and results: Ten minutes would be too short to reach equilibrium with the surrounding air at the given RH of 25 – 30 %. Please check and compare with the equilibrium time from Grayson et al., 2017.

Grayson, J. W., Evoy, E., Song, M., Chu, Y. X., Maclean, A., Nguyen, A., Upshur, M. A., Ebrahimi, M., Chan, C. K., Geiger, F. M., Thomson, R. J., and Bertram, A. K.: The effect of hydroxyl functional groups and molar mass on the viscosity of noncrystalline organic and organic-water particles, *Atmos. Chem. Phys.*, 17, 8509-8524, 10.5194/acp-17-8509-2017, 2017

Authors: Thanks for the comment. Time required to equilibrate with respect to the relative humidity of the surrounding depends on the rate of diffusion. In Grayson et al., particles of size range of 55 – 80 um in diameter are probed at 19% relative humidity. This is shown to correspond to characteristic time of mass-transport and mixing of water to be between 365 to 771 minutes.

However, the largest particles we probed are at best, 0.9 um in volume equivalent diameter, as illustrated by Figures 2 and 4. Since the diffusion coefficient is inversely proportional to diameter, one can expect a proportional decrease in the equilibration time. For example, decrease in size from 80 to 0.8 um is approximately 2 orders of magnitude, so equilibration time of approximately 7.7 minutes is expected for 0.8 um sized particles.

This is further supported by previous works that studied the half time of particle radius as a function of RH, where a 1.6 um diameter sucrose at 20% RH showed half time of approximately 250 seconds. Even

the largest particle that we studied, would be a factor of nearly 2 lower in diameter, and thus we expect the half time to further significantly decrease from 250 seconds. (Lu et al., 2014)

This is now reflected in the manuscript, in Section 2.3 AFM imaging,

“This timeframe is reasonable given the diffusive nature of water transport within a particle, which is dependent on the particle size.(Grayson et al., 2017;Lu et al., 2014)”

Reviewer2: 5. Experimental and results: If the particles undergo RH decreasing gradually from high RH (i.e. 100%) to 25-30%, would you still expect core-shell morphology at the RH? It would be worth to try additional experiments.

Authors: For this mixture, we would indeed expect the core-shell morphology. In fact, in our hydration-dehydration cycles, the RH is raised up to 80%, which is beyond the deliquescence point. Further increasing the RH to nearly 100% and then lowering down to 25-30% would not affect our results, as the difference will lie in the amount of water that the particle uptakes at high RH. Since the core-shell morphology is also reproducible with multiple hydration-dehydration cycles leading to similar OVF, we feel there is no obvious need to do any additional experiments.