

**Reviewer1:** *This paper investigated dry and wet deposition effects on the generated and impacted aerosol particles' mixing states that display core-shell morphology. In general, the techniques are well-used and the manuscript is well written. However, some suggestions are worth noting.*

**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.

**Reviewer1:1** *The abstract section is too long. Only key information is needed.*

**Authors:** Thanks for the comment; we revised the abstract to make it shorter. It is now 306 words in length. New abstract reads as follows,

“Understanding the impact of sea spray aerosols (SSA) on the climate and atmosphere requires quantitative knowledge of their chemical composition and mixing states. Furthermore, single particle measurements are needed to accurately represent large particle-to-particle variability. To quantify the mixing state, organic volume fraction (OVF), defined as the relative organic volume with respect to the total particle volume, is measured after generating and collecting aerosol particles, often using deposition impactors. In this process, the aerosol streams are either dried or kept wet prior to impacting on solid substrates. However, the atmospheric community has yet to establish how dry versus wet aerosol deposition influences the impacted particle morphologies and mixing states. Here, we apply complementary offline single particle atomic force microscopy (AFM) and bulk ensemble high performance liquid chromatography (HPLC) techniques to assess the effects of dry and wet deposition modes on the substrate-deposited aerosol particles' mixing states. Glucose and NaCl binary mixtures that form core-shell particle morphologies were studied as model systems, and the mixing states were quantified by measuring the OVF of individual particles using AFM and compared to the ensemble measured by HPLC. Dry deposited single particle OVF data positively deviated from the bulk HPLC data by up to 60%, which was attributed to significant spreading of the NaCl core upon impaction with the solid substrate. This led to underestimation of the core volume. This problem was circumvented by a) performing wet deposition and thus bypassing the effects of the solid core spreading upon impaction and b) performing a hydration-dehydration cycle on dry deposited particles to restructure the deformed NaCl core. Both approaches produced single particle OVF values that converge well with the bulk and expected OVF values, validating the methodology. These findings illustrate the importance of awareness in how conventional particle deposition methods may significantly alter the impacted particle morphologies and their mixing states.”

**Reviewer1:2** *As we know, levoglucosan dominates the sugar particulates, while little glucose exists in the atmosphere. Meanwhile, more details about glucose (or particle phase sugar) should be added in the Introduction section.*

**Authors:** Levoglucosan is a major component of biomass burning aerosol, however it is has not been detected in nascent sea spray aerosol, which is the chemical system that is modeled in this set of experiments. In regard to sea spray aerosol, glucose has been identified as the most abundant carbohydrate, contributing 5.2% and 14.4% of PM<sub>2.5</sub> and PM<sub>10-2.5</sub> (Jayarathne et al., 2016).

This is now reflected in the text, at the last paragraph of the 1 Introduction. Following sentences were included,

“Both chemical systems are highly relevant to the SSA, with glucose up to 5.2% and 14.4% of the total organic mass of PM<sub>2.5</sub> and PM<sub>10-2.5</sub> SSA, respectively. (Jayarathne et al., 2016) Further, both glucose and NaCl are surface inactive species, thus OVF is not expected to be size-dependent. (Cochran et al., 2017)”

**Reviewer1:3** *Section 3.1: Authors employed the RH of 25% to measure OVF. However, RH in the real atmosphere is usually higher than 30%. Additionally, whether the authors considered the particle hygroscopicity?*

**Authors:** It is true that the RH in the atmosphere fluctuates. However, controlling of RH in our experiments, to quantify the organic volume fraction, is solely to minimize the amount of water within the particles to improve the accuracy of this methodology. As outlined in Section 3.1 Morphology of phase-separated binary component particles, we previously stressed that,

“Here, RH was maintained at a constant range of 25 – 35% throughout the imaging experiment, to minimize “phase bleeding” and water uptake (Fig. S1). In this work, phase bleeding describes an instance in which the viscosity of the two phase-separated materials is too similar at a given RH, and therefore the phase contrast between organic and inorganic components is relatively weak in the AFM images. A high phase contrast image aids in correctly differentiating the core and shell phase boundaries. For this particular system, a dramatic increase in phase bleeding is evident below 25% RH, due to a significant increase in the glucose viscosity as it becomes more solid-like, which closely resembles the NaCl viscosity at this RH value (Fig. S1). (Song et al., 2016) Since phase imaging inherently relies on measuring differences in tip-sample interactions originating from different viscoelastic properties, lowering the RH will further converge the two different viscosities of organic and inorganic components together, lessening the accuracy of the core and shell phase boundary determination. Although higher RH values would produce less “phase bleeding”, significant particle water uptake will also introduce erroneous OVF values, due to an increase in particle volume from water uptake.”

The authors did indeed consider the hygroscopicity of the chemical systems. In fact, the smooth and slow uptake of water, unique for organic species such as saccharides, contributes to access to solid, semisolid, and liquid phase states for glucose, which was mentioned in the introduction. Under Section 3.1 Morphology of phase-separated binary component particles, the following sentence was revised to address the influence of the change in organic shell volume due to hygroscopic growth (only 3% here for glucose):

“The value of 25% RH was thus used for the measurements of OVF, because growth in the organic shell size due to water uptake was measured to be less than 1.03, or merely 3% growth while showing excellent phase contrast. (Lee et al., 2017a)”

**Reviewer1:4** *For the equation S1, which kind of density is used here? Please explain in detail.*

**Authors:** Thanks for the comment. The solute densities of pure glucose and NaCl were used. The Ensemble average OVF comparison between AFM and bulk in the supporting information now reads,

“In this case, the concentrations of glucose, sodium and chloride were determined in aqueous extracts of filter samples and were field blank subtracted. Masses of these species were converted to volumes, and used to calculate  $OVF_{\text{bulk}}$  following Equation S1,

$$OVF_{bulk} = \frac{\frac{m_{org}}{\rho_{org}}}{\frac{m_{org}}{\rho_{org}} + \frac{m_{inorg}}{\rho_{inorg}}} \quad \text{Eq. S1}$$

where  $OVF_{bulk}$  is organic volume fraction of bulk,  $m_{org}$  &  $m_{inorg}$  are masses, and  $\rho_{org}$  &  $\rho_{inorg}$  are pure solute densities of organic and inorganic components, respectively.

**Reviewer1:5** *The authors used one paragraph to describe the t-test and null hypothesis. But t-test is a simple statistical tool and only the significant result should be put in manuscript.*

**Authors:** Thanks for the comment. We moved the explanation of the t-test and null hypothesis in Sections 3.2 to the supporting information. In addition, replacements in sections 3.2 and 3.4 now read,

“OVF measured from dry deposited particles and wet-deposited particles were compared with a two-tailed t-test, which demonstrated that these two datasets are significantly different ( $p < 0.0001$ , see SI).”

“To confirm this statistically, same student’s t test methodology was employed, which demonstrated that these two datasets are significantly different ( $p < 0.0001$ , see SI).”

**Reviewer1:6** *Conclusion section. The atmospheric implications of the experimental results should be discussed.*

**Authors:** To begin, this work heavily implicates that the particle generation method may affect measuring of organic volume fraction using offline, single particle techniques. This may lead to erroneous representation of the mixing state, which may affect the accuracy of climate models that require this knowledge to understand aerosol-cloud interactions.(Ault and Axson, 2017) This reference was added into the introduction. On the first paragraph of the 1 Introduction, we added,

“Uncertainty in the mixing state, even with the knowledge of the chemical composition, may produce erroneous predictions of cloud activation from individual particles. (Ault and Axson, 2017)”

Moreover, the following was added to the conclusion,

“Overall, our findings provide implications of aerosol generation on accurately identifying the mixing state of individual, phase separated particles, important for the atmospheric community and climate predicting models that heavily rely on not only the knowledge of the chemical composition, but also the resultant mixing states of atmospherically-relevant aerosol particles.”