REVIEWER #1

We thank the referee #1 for the positive assessment and helpful comments. Following are the comments raised (in ***red** and italics*), and our responses in plain text.

This manuscript describes the phase state of ambient organic dominated particles in the Southeast US during the SOAS campaign. The phase state is using the aerosol bounce technique. The phase state was measured concurrently with hygroscopicity measurements using a HTDMA and chemical composition using an AMS. The ambient particles are further aged using an oxidation flow reactor and the phase state observed. The manuscript is well-written and I support publication in ACP after the following comments are addressed.

Comments:
1) The authors mention that the most interesting effects on phase state occur between 30 -70% RH (for isoprene). However, 75% of the time the ambient RH was above 70% RH. Therefore, the authors are not measuring the ambient phase state of the particles. How can the authors state that at ambient RH the particles will be liquid, if these measurements were never made?

Typical BF curve of amorphous SOA particles with RH is a decreasing sigmoidal curve (see e.g. Saukko et al. 2012 Fig. 5., Pajunoja et al. 2015 Fig. 2., Bateman et al. 2016 Fig 2.), and this information can be used when extrapolating the bounce measurement results to cover the whole RH range. In this study we wanted to study the effect of O:C and hygroscopicity on BF. During the first days of the campaign we performed some measurements at RH > 75% and as the BF values were very low in general, the differences in BF were small. As we focused on studying the effect of O:C and hytgroscopicity on BF, we lowered the measured RH range to 30-70% to see clearer variations in bounce. However, the reviewer is correct, we do not have bounce measurements reported in the range of RH>70% in the manuscript. Hence, we have reworded the manuscript and we now conclude that (Page 1 line 24, Page 9 line 2, and Page 11 line 31) “our results indicate that organic dominated particles stay mostly liquid in the atmospheric conditions in the Southeast US”.

2) The data in Figure 3 do not reach zero. Again, how can the authors state that the ambient particles are liquid, if they are bouncing during impaction?

As mentioned above, the typical bounce curve for SOA particles is previously studied and based on the studies we can extend our results to higher RH range. Based on e.g. Bateman et al. (2015), the viscocity of SOA particles having approx. BF < 0.4 is already low enough to assume liquid like behavior of particles when atmospheric processes are considered.

3) How can the authors rue out the possibility of two particle types, one solid phase (bouncing) and one liquid phase (sticking) vs. one particle type of intermediate phase (50% bouncing)?

Externally mixed particle population containing both liquid and semisolid particles with different chemical compositions should have different hygroscopicity resulting multimodal GF-PDFs. Time periods when GF-PDFs were multimodal were excluded from the analysis and only time periods representing the internally mixed particles (single mode GF-PDFs) were included in the analysis (mentioned in Page 5 line 28-).

4) Page 4 line 25-27: This range and phase transition is true only for a homogeneous distribution. How do the authors know this is the case in the ambient? Bateman et al., 2015 published a relationship between viscosity of sucrose particles and bounce fraction.

Externally mixed particles were filtered with the method explained above. Based on the dataset used in this study, we cannot distinguish liquid-semisolid phase separation from fully mixed semisolid particles which have equal BF. To avoid the dominating effect of inorganics on total BF we narrowed our analysis to include only periods where the organic mass fraction (derived from AMS analysis) was greater than 0.6. As shown in Saukko et al. (2015), within this range the effect of ammonium sulfate deliquescence is diminished and the total BF is dominated by organic fraction of the particles.

5) Page 11 line 10-12: Any correlation with the AMS collection efficiency? Was the collection efficiency on the same order predicted by the bounce fraction? Why or Why not?

The correlation between AMS collection efficiency and bounced fraction is very interesting topic and under investigation in near future, which will be addressed in an another technique paper on AMS quantification due to its complexity. It is good to keep in mind that when the particles are dried before sampling inlet with Nafion dryer to achieve RH<30% for quantification purpose and impacted to the heated AMS vaporizer. In the vacuum system of AMS, the particles are in low pressure and thus should be further dried. As the viscosity is a function of RH and T, the actual viscosity in the AMS vaporizer may be very different compared to viscosities in the impactor conditions. Thermal decomposition of particle on the AMS vaporizer may also influence the CE, which will not be seen in the impactor condition.

Minor Comments:

1) Page 2 Line24 -26: Perhaps Shrivasta et al. 2013 and Zaveri et al. 2014 or Scott et al 2015 have used this in their modeling. If not, they might have 2015 or 2016 papers in which these effects are included.

We thank reviewer for the good references. Those are now included to the text as follow (Page 2 line 19-21): "Zaveri et al. (2014) took the lower diffusion rates into account in modeling kinetic partitioning and size distribution kinetics. Recently, the kinetic approach where organic material condenses according to the surface area rather than organic mass of the particles has been tested in the aerosol microphysical models (Shrivastava et al. 2013; Scott et al. 2015). We have also modified the text (Page 2 line 28).

2) Page 3 line 5: I believe Bateman et al. 2016 used terms such as liquid particles dominate, but they are not "always" liquid.

We have modified the text accordingly.

3) Page 4 line 11: Is eight seconds enough time for kinetically limited particle to uptake water? Berkemier has some calculations and timescales for this.

This is a relevant question! For condensation/absorption of big organic molecules the timescale might be too short. Based on Pajunoja et al. (2015), water uptake of 100-120nm semisolid SOA-particles did not increase when the humidification time in HTDMA was increased. Thus, 8 seconds should be long enough for small water molecules to diffuse. It should be also noted that Stokes-Einstein relation is not valid for water molecules diffusing inside the organic matrix and the diffusion coefficients are clearly higher than predicted using Stokes-Einstein (e.g. Marshall et al., 2016).

4) Page 4 lines 19 – 21: I have no idea what this means? What do the RH sensors and the CPC offsets have to do with each other? Please clarify this sentence.

We agree with the reviewer; the sentence is unclear. Checking the RH sensors has nothing to do with the CPCs. The text is now modified:
"Outputs of RH sensors (Vaisala, HMP-110) used in the ABI were compared to the theoretical deliquescence RH of pure ammonium sulfate by measuring its humidogram with the ABI prior and after the campaign. The offset of the two CPCs (i.e. $BF_{reference}$) instead was measured every second day, at the very least."

5) Page 5 line 29-20: how any points were lost from this analysis? How was the external mixture determined?

Data coverage of multimodal GF-PDFs (i.e. externally mixed cases) was less than 10%. All the HTDMA scans were analyzed and both unimodal and multimodal distributions were fitted to each scan. In most of the cases the unimodal distribution was clearly the best fit for the GF data. This was confirmed by comparing the goodness of fitting of unimodal and multimodal fits and also confirmed by eye.

6) Page 6 Equations 2 – 4: How much will uncertainties in the calculation of these quantities effect the conclusion the authors draw? The authors should comment on this in the manuscript.

It is true that the mixing rule calculations are sensitive to assumptions made in defining the inorganic fraction. The approach how the inorganic fractions are calculated is based on thermodynamic equilibrium model ISORROPIA (Nenes et al. 1998). If we trust AMS analysis, the composition of inorganics should be reliable. Anyhow, we have limited our analysis to periods where the mass fraction of inorganics has been lower than 40%. In the worst scenario all the inorganics would be consisted of either pure sulfuric acid or pure ammonium sulfate which is not the case since the particles have shown to be acid during the SOAS campaign. Nevertheless, we tested this scenario and it generates less than 0.04 spread (the difference between the scenarios and the reported κ_{OA} being somewhere in between) in κ_{OA} over 95% of the dataset. The uncertainties do not affect the trends in κ_{OA} but the absolute values may be slightly different. We have now added a phrase about the uncertainties in mixing rule calculations as follow (Page 10 line 5-7):

"The sensitivity of the method to calculate κ_{OA} was also tested by varying f_{AS}, f_{ABS} and f_{SA}. In more than 95% of the cases the spread in κ_{OA} is less than 0.04 and it does not change the trends in Fig. 4d."

Technical Correction

1) Page 3 line 16: "physical phase state" remove either physical or phase

Word "phase" now removed.

2) Page 11 line 11: replace bounciness with a term that describes physical state i.e. viscosity, semi-solidness

Word "bounciness" is replaced with "semi-solidness".

> *1 Bateman, A. P., Bertram, A. K. & Martin, S. T. Hygroscopic influence on the semisolid-to-liquid transition of secondary organic materials. J. Phys. Chem. A 119, 4386-4395 (2015).*
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> *2 Scott, C. E. et al. Impact of gas-to-particle partitioning approaches on the simulated radiative effects of biogenic secondary organic aerosol. Atmos. Chem. Phys. 15, 12989-13001 (2015).*
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> *3 Shrivastava, M. et al. Implications of low volatility soa and gas-phase fragmentation reactions on soa loadings and their spatial and temporal evolution in the atmosphere. J. Geophys. Res. Atmos. 118, 3328-3342 (2013).*
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> *4 Zaveri, R. A., Easter, R. C., Shilling, J. E. & Seinfeld, J. H. Modeling kinetic partitioning of secondary organic aerosol and size distribution dynamics: Representing effects of volatility, phase state, and particle-phase reaction. Atmos. Chem. Phys. 14, 5153-5181 (2014).*

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Saukko, E., Kuuluvainen, H., and Virtanen, A.: A method to resolve phase state of aerosol particles. Atmos. Meas. Tech., 1, 259-265, 2012.