REVIEWER #2

We thank the referee #2 for the positive assessment and helpful comments. Following are the comments raised (in ***red** and italics*), and our responses in plain text.

The paper by Pajunoja et al. investigates the phase state of ambient particles in the Southeastern US using an aerosol bounce instrument. The work shows that ambient particles in this region are mostly in the liquid state. In addition further analysis shows that the phase state is related to the hygroscopicity of the particles. The paper is very well written, the analysis is excellent, and the results are important for modelling aerosol formation and growth in the atmosphere. I highly recommend this paper for publication after the authors have had a chance to address the following comments.

Abstract, line 26-28. What sampling techniques are you referring to here? I don't think this was discussed anywhere in the main document. More specifics somewhere in the document would be useful to the reader.

By "sampling techniques" in the abstract we refer to all the aerosol sampling systems where the particles are dried to lower than ambient RH. Vast majority of the aerosol inlets used in the field and laboratory studies contains drying method, including chemical reactivity and volatility measurements. In worst case, the particle viscosity increases by three to four orders of magnitude due to drying, which may lead to dramatic changes in physical and chemical ability to interact with surrounding gas-phase. We have now added couple sentences to the main text about the issue as follow (Page 9 line 4-6):

"Such a clear difference in BF between dry and ambient RH indicates the possibility that the aerosol particles may undergo phase transition from liquid to semisolid when dried in any sampling system. This could cause measurement error when investigating for instance evaporation/condensation, chemical reactivity or volatility."

Page 2, line 31-32. The authors state: "Saukko et al. 2012, showed that the increasing O:C of SOA particles decreases the particle liquefying RH". After reading this sentence I went back and looked at the abstract for Saukko et al. 2012. In the abstract Saukko et al. state "in the majority of cases the bounce behavior of the various SOA *systems did not show correlation with the particle O:C." These two statements sound contradictory. Please clarify.*

The reviewer is correct, Saukko et al. (2012) is not the best reference to this specific questions since due to the methodological restrictions at that time the RH was limited to lower values where the differences in bounce where small regardless of varying O:C. Thus, we have now removed the sentence and used more recent reference (Pajunoja et al. 2015) showing clearly the effect of O:C on particle liquefying RH.

Please define the terms in equation 5. Also, why is equation 5 needed? Can't the OH exposure be calculated directly from the real-time decay of CO?

Very good point! The previous analysis rested on equation (5) due to poor data coverage in CO data. Recently the CO data has been re-analyzed and the data coverage was improved. Thus, the revised OH exposure is now calculated directly from the real-time decay of CO as the reviewer #2 suggested, and the equation (5) is removed from the manuscript. The methods are compared comprehensively in Hu et al. (2016) (see Supplementary Information Fig. S4 therein).
Due to the slight changes in OH exposure values, Figure 5a is reproduced with the new values. As can be seen, the overall trend in colors did not change even the absolute values of OH exposure changed slightly.

During the re-analysis of the OH data we noticed also that the Fig. 5c had been plotted with incomplete O:C data. Thus, the Fig. 5c is now replaced by the revised one. This did not affect the conclusions.

Figure 4c. I wonder if the occurrence of liquid-liquid phase separation (LLPS) in the particles is influencing the particle bounce. LLPS in particles containing organic and inorganic material is expected to occur at O:C values less than approximately 0.7 (very roughly). In Figure 4c, when the O:C is less than 0.7 significant bounce is observed even at high RH, which is when LLPS is expected. On the other hand, when the O:C is roughly 0.7 and greater, the bounce is significantly reduced, which is when LLPS is not expected.

This is very interesting question. Based on Saukko et al. (2015) it is not possible to distinguish mixed and phase separated cases by bounce measurement unless both, the humidification and drying cycles, are measured. If both cycles are measured it is possible to detect the hysteresis behavior in bounce curves, if the inorganic and organic phases are separated and if the organic fraction is not too high. Hence, based on the dataset shown in this study, we cannot distinguish the phase separation and fully mixed particles from the data and it is possible that the bounce is affected by the particle structure. Anyhow, to minimize the effect of inorganic fraction 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. We would like to also note, that the lab results (e.g. Pajunoja et al., 2015) indicates that at humidified conditions the bounce of SOA particles with no inorganic fraction clearly depends on O:C of organic material. Hence we believe that the behavior presented in Fig. 4. and Fig. 5. are dominated by the water uptake of particles and the possible structural effects play a minor role.

References

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