

Interactive comment on “On the link between hygroscopicity, volatility, and oxidation state of ambient and water-soluble aerosol in the Southeastern United States” by K. M. Cerully et al.

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“The paper presents results from a field study in which the link between measured hygroscopicity, volatility and oxidation state. As a paper presenting analysis from a field campaign it is clear and concise. I do not have any major issues with the results presented but there are a few scientific debates that need to be included before publication.”

We thank the referee for this positive assessment and feedback. Following are the comments raised (in quotes and italics), and our responses in plain text.

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“There seems to be no mention of the potential effect of kinetic mass transfer limitations on ambient consequences. Whilst the CCNc would not, should not (?), experience effects due to this phenomena, it might have consequences for ambient water uptake below super saturated humid conditions. Or do the authors not agree?”

This is an interesting point. The current study does not focus on the issue of water uptake kinetics, as it would considerably extend an already long study. We defer the important topic of water uptake kinetics to a future study.

“Section 4.1 The result on the relatively low change in hygroscopicity of denuded aerosol is interesting. Is there any evidence that a significant increase in temperature might change the properties of organics in the complex matrix? Or, is there evidence to suggest this isn't the case?”

This is an excellent point, and was also raised by reviewer 1. A subsequent study in preparation focuses on applying PMF on the SOAS TD aerosol, and confirms that the mass spectra of the PMF factors remain largely unaltered by the volatilization. As a consequence, the hygroscopicities are not expected to be affected by the volatilization process either. We refer to this future work briefly in the text.

“There is no discussion of any potential artifacts from semi-volatile re-condensation after the thermodenuder. Could this effect the measured, and relatively insensitive, change in hygroscopicity?”

We thank the reviewer for raising this point. Recondensation of vapors is a concern for TD measurements in general. However, we assert that for the measurements described in this manuscript, it is not an issue (see manuscript lines 128-131). TD cooling sections have been demonstrated to potentially impact the aerosol evaporation, depending on several factors – importantly, mass concentration and residence time. The cooling section will have a minor impact at low mass concentrations and low residence times unless the aerosol is very volatile (i.e., if the activated carbon, through its intense absorption of organic vapors, can generate a concentration gradient in the vapors near

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the aerosol surface that is notably larger than without the activated carbon; Cappa, 2010 and Saleh et al., 2011). The size distribution of the volatilized aerosol is measured also at about the same distance upon the exit of the thermodenuder as the CCN instrument, so any recondensation or additional evaporation occurring downstream of the heating section should be reflected in the corresponding sizing, hygroscopicity and composition measurements. Most importantly, our measurements occurred under both low aerosol mass loadings and residence times in the TD, which ensures that the cooling section has a minor impact on our results.

Regarding the presence of soluble gases in the CCN instrument affecting the observed hygroscopicity: very high levels of vapors are required for that to occur, because soluble vapors tend to be absorbed by the wetted walls of the CCN instrument instead of the CCN (Romakkaniemi et al., 2014). It is however possible that there may be some additional volatilization in the CCN instrument, especially when sampling the non-denuded ambient/PILS aerosol.

“Have the authors any existing, or future plans, to study proxy mixtures in the lab using the same instruments? For example, for a mixture of inorganic:organic with known composition it might be quite telling to observe the same behavior for a system that should be well characterized in terms of pure component and mixture properties.”

Thank you for the suggestion. We agree that studying proxy mixtures in the lab with components of varying degrees of volatility and hygroscopicity will be valuable for evaluating the methodology and its associated uncertainties, and is in our plans for a future study.

References

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