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Interactive comment on “Trends in particle phase liquid water during the Southern Oxidant and Aerosol Study” by T. K. V. Nguyen et al.

Anonymous Referee #1

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This paper is a single study to present a methodology to try and resolve the potential effects of semi-volatile loss/gain in deriving instrument based measured hygroscopicity. As the authors are aware, this area is fraught with difficulties, not least forced by the measurement techniques used and the complexity of ambient aerosol and single particle processes. Much work is needed in the area before the effects are truly resolved. The authors provide a valid point in that actual concentrations of aerosol water are not routinely measured. This study presents a methodology to rectify this. Whilst I feel the paper is appropriate for ACP, there are a number of general points that need to be addressed before publication that will affect the tone of the delivered conclusions. Presently, the results are interesting as a proof of concept study, but obviously require caution in further extrapolation. In addition, there are a number of highly qualitative dis-

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cussions that would benefit from some clarity, including an appropriate testing ground for the proposed methodology.

General comments:

General introduction. There is obviously still debate as to the role of water in SOA partitioning. I would suggest providing a few more succinct upto dates more wider references to discussions had on this in your introduction: the potential impact of LLE, the mass and molar based absorptive partitioning frameworks.

Page 7476: Line 4: 'Presumably some semi-volatile substances were irreversibly removed inside the chilled copper tube. We expect that warming of the sample flow after the copper tube would result in re-equilibration of the semi-volatile compounds, similarly to what is observed with water. Thus this configuration was intended to measure the particle size distribution with minimal perturbation to sample temperature and RH after removal of some fraction of semi-volatiles.' - This is very qualitative. I appreciate the difficulties in quantifying this effect but it would help the reader to know how these qualitative hypotheses might be untangled. If the authors feel that discussion exists, then please better reference different parts of the manuscript. See my comment below re testing using proxy systems.

Page 7479: Equation 6. So the authors are presuming no semi-volatiles re-equilibrate on re-humidifying as might be expected if a gaseous component were to re-equilibrate according to Raoult's law? This is confusing in relation to the statement made : 'We expect that warming of the sample flow after the copper tube would result in re-equilibration of the semi-volatile compounds, similarly to what is observed with water.'

Page 7479: Equation 6, according to the other equations this should be V_w, l_v and not total water shouldn't it? So equation 6 just gives you water associated with your definition of low volatility compounds?

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Page 7480: ‘Non-hygroscopic materials lost during drying’: So these compounds evaporate due to the temperature increase? What volatility range of compounds would you expect to lose from this effect based on a simple C^* calculation with a fixed typical enthalpy of vaporisation? What about timescales used in the instrument, do these reflect typical conditions used in other studies of hygroscopicity?

Section 2.4 How much is ‘slightly lower’?

Isn't there any supporting aerosol composition information? It is very hard to try joining all pieces of information without this data. There is a brief discussion as to the potential effects in section 4 which is interesting. Similarly, isn't one crucial piece of information at least some idea as to the ‘expected’ volatility of the aerosol you are sampling. Is it heavily aged SOA for which the proposed effect would be minimal anyway? One interesting hypothesis comes to mind is the following. The partitioning of compounds, if we are to believe absorptive partitioning playing a dominant role, is dependent on the available absorptive mass. That is, in the atmosphere, the re-partitioning of available semi-volatile material is sensitive to particle size distributions and number concentrations. I presume the authors do not feel the instrument samples gaseous components in the inlet that would prevent the evaporation of compounds from mono-disperse particles (very small mass relative to available mass in the gas phase perhaps)?

On this note, as the paper is presenting a new technique, I'm surprised to not find any tests on proxy systems for which you could confidently extract the myriad of processes effecting these measurements: Individual component volatility, non-ideal effects, phase separation effects, diffusion effects. Would the authors not feel it appropriate to study at least a few, say, ternary or quaternary proxy mixtures before moving to complex ambient aerosol?

Do you think the conditions and timescales from which you are extracting impacts are appropriate for atmospheric timescales and conditions. If, for example, the aerosol was in a viscous state with equilibrium timescales approaching 10's of minutes, then what

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difficulties arise in using these measurement techniques to prescribe wider impacts?

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