

***Interactive comment on “Alkene ozonolysis SOA:  
inferences of composition and droplet growth  
kinetics from Köhler theory analysis” by  
A. Asa-Awuku et al.***

**Anonymous Referee #2**

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This manuscript addresses the properties of secondary organic aerosol formed in the oxidation of three alkenes: terpinolene, 1-methyl cycloheptene and cycloheptene. The authors aim at determining ccn properties, surfactant characteristics and droplet growth kinetics. The authors should be complemented for presenting new ideas to elucidate the cloud droplet activation of SOA. In this work they suggest to add salt to extracts of filter samples of organic aerosols and determine critical supersaturations of particles generated from these mixtures. From the measured activation curves the authors infer surface tensions at the point of activation as well as organic molecular weights. The manuscript in its current state does however not provide enough consideration on the experiments and related uncertainties nor on the underlying assumptions and theory

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used to model the data and infer surface tensions and molecular weights. The magnitude of uncertainties introduced by various assumptions: densities, van't Hoff factors and mass fractions of organic and inorganic material in the particles should be addressed. Some of the following issues have also been addressed by referees #1 and #3.

#### Specific comments

##### Page 8986

There is too little information about the conditions in the smog chamber. What was the reactant concentrations?, what was the time scale? relative humidity and temperature? In the study by Gao et al. 2004a seed particles and an OH scavenger were used - were seed particles and OH scavenger also used in the present study?

##### Page 8987

Likewise, in relation to the CCN experiments more information on the experimental details would be useful. The authors should show examples of activation curves to give an impression of the quality of the data. How was doubly charged particles accounted for in the analysis? I have some questions regarding the calculated mass fraction of inorganic material: 1) The organic carbon-to-carbon ratio seems to be a quite uncertain parameter. It should be better explained what this ratio is and how it was obtained. How is a general factor of 2 justified from the rather limited information in Table 1? 2) Evaporation: Glutaric acid is listed as a major soluble organic compound - it has a high vapor pressure. What is the time that the particles spend between the atomizer and the ccn-counter? Could evaporation of the organic fraction of the atomized aerosol particles take place and thus change the composition of the particles studied compared to the composition of the mixture in the atomizer?

##### Page 8988

The authors mention critical micelles: this should be more thoroughly addressed -

can the relevant range of the critical micelle concentrations for the major products be estimated? When are micelles formed in the present study?

Page 8989

It says that “If the salt mass fraction exceeds 50% the majority of dissolved solute is usually from the inorganic salt”, I suggest to give some actual calculated values from the current study as example. Why was 33% (and not  $> 50\%$ ) used in many of the experiments?

Which van't Hoff factor was used for ammonium sulfate?

Page 8990

If the organic contribution to the Raoult effect is not negligible the authors say that it must be accounted for in equations 6-8. To help the reader it should be better explained how this is actually done.

The authors do not address partitioning of surfactants between the bulk and the surface as discussed by Sorjamaa et al. [1-3] and Li et al. [4]. Surfactant partitioning is not accounted for in the traditional derivation of the critical supersaturation (power dependence of  $3/2$ ) used by the authors - it is not clear how it looks if surfactant partitioning is included in the derivation of the critical supersaturation.

Page 8991

What were the estimated values of surface tension used to obtain the INCA parameter?

Method b1 in Padro et al. assumes that there are no strong surfactants present at the point of activation, yet the conclusion is that surface active compounds do exist in the mixture. This does not seem consistent?

Page 8992

Figures 1-3: it should be explained what the solid lines in the figures are. How can

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it be concluded that the SOA are soluble hydrophilic relatively low molecular weight compounds that are not surfactants based on the  $m^{-1/2}$  dependence? The  $m^{-1/2}$  dependence should be demonstrated more clearly, for example in a table.

Page 8993+8994

The density of 1.4 was adopted by Goa et al. from Kalberer et al. [5]. Also it is discussed by Gao et al. 2004 [6] that the density could very well be larger than 1.4 and that the density is a quite uncertain parameter. What are the consequences of a different density?

Table 5 and related text: The inferred surface tension shows roughly 15% depression from pure water, only in the case of cycloheptene and terpinolene SOA with 33% ammonium sulfate. It does not seem justifiable to make conclusions and comparison with HULIS based on this without consideration of the uncertainties (in particular for terpinolene), assumptions and the lack of information about terpinolene SOA (Table 1). How much should the van't Hoff factor used for the organics (cycloheptene as well as terpinolene) deviate from one, or how much should the experimental value be in error to get an inferred surface tension equal to that of water or on the other extreme HULIS (e.g. Kiss et al.)? I miss numerical values on critical supersaturations to compare different approaches. For example calculated values of critical supersaturations and inferred surface tensions using different values of the parameters (van't Hoff factor, density,  $m_i$ ) could be given in a table together with experimental values including uncertainty ranges.

The authors state that the agreement of the effective molecular weights with the Gao data [6] validates the use of inferred surface tension values in KTA - this seems like an overstatement since the inferred values on  $\sigma_{crit}$ ; relies on results from Gao et al [6]?

Technical corrections

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The notation is not consistent: Page 8988 minorganic, page 8990 mi Explain what the difference between &#61550;i and &#61550;o is. In the text the term &#61550;organic is used

Reference to Gao et al. 2004a: Volume number (108) is missing

1. Sorjamaa, R., et al., The role of surfactants in Kohler theory reconsidered. *Atmospheric Chemistry and Physics*, 2004. 4: p. 2107-2117.
2. Kokkola, H., et al., Cloud formation of particles containing humic-like substances. *Geophysical Research Letters*, 2006. 33(10).
3. Sorjamaa, R. and A. Laaksonen, The influence of surfactant properties on critical supersaturations of cloud condensation nuclei. *Journal of Aerosol Science*, 2006. 37(12): p. 1730-1736.
4. Li, Z.D., A.L. Williams, and M.J. Rood, Influence of soluble surfactant properties on the activation of aerosol particles containing inorganic solute. *Journal of the Atmospheric Sciences*, 1998. 55(10): p. 1859-1866.
5. Kalberer, M., et al., Aerosol formation in the cyclohexene-ozone system. *Environmental Science & Technology*, 2000. 34(23): p. 4894-4901.
6. Gao, S., et al., Low-molecular-weight and oligomeric components in secondary organic aerosol from the ozonolysis of cycloalkenes and alpha-pinene. *Journal of Physical Chemistry A*, 2004. 108(46): p. 10147-10164.

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