Atmos. Chem. Phys. Discuss., 12, C8532–C8534, 2012
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "CCN activity and volatility of β-caryophyllene secondary organic aerosol" *by* M. Frosch et al.

Anonymous Referee #2

Received and published: 25 October 2012

Review of CCN activity and volatility of β-caryophyllene secondary organic aerosol by Frosch et al.

The paper discusses the trend in hygroscopicity of β-caryophyllene SOA under supersaturated conditions using κ_{CCN} as a primary diagnostic of CCN activity. It appears to be largely motivated by the Asa-Awuku et al. (2009) study and borrows strongly from the methodology therein. The key differences appear to be the range of experimental conditions (e.g. presence / absence of OH scavenger, addition of illumination or an additional OH source in HONO). There is broad agreement with previously published work, but some discrepancies. Whilst the study is predominantly observationally driven and remains largely inconclusive, the experiments seem to have been carefully conducted and are worthy additions to the literature. I recommend publication of the

manuscript once the authors have considered and responded to the following comments (in addition to those of the first referee):

On page 20758, the last paragraph seems to ask more questions than it answers and indicates apparent inconsistency between the TD measurements and the hypothesis of Asa-Awuku that was used to explain the SD-CCNc and CF-CCNc discrepancy in terms of volatility. The last full paragraph on p20762 seems to provide further evidence to counter the argument that volatility alone can explain the discrepancy. Whilst it is clearly important simply to report the observations, it would be useful for the authors to indicate the most likely of the alternative explanations suggested and possible experimental / interpretational strategies to investigate them.

In the paragraph starting on line 7 of page 20758, the authors stop short of stating that the Tang et al measurements were in error, but the tone clearly implies mistrust of them. Do they have any means of rationalising the discrepancy between their study and the Tang measurements?

p20754, lines 9-11, can the authors expand on the avoidance of problems associated with multiple charging? Since determination of $D_{dry,c}$ requires a fit to activated fraction at all diameters, surely all diameters with activated fractions > 0 need to be above the modal diameter to avoid multiple charges being a problem at some point in the fit. Does this not place a very low limit on maximum supersaturations that can be used and still avoid multiple charge problems? The modal size does not appear to be small enough to avoid this problem. Or do these lines only apply to the "fixed D_{dry}" method and only for the SD-CCNc as described in line 20 p 20753? If this is the case, the authors need to explain clearly how they ensure that multi-charge correction is carried out accurately when using the "fixed S" method with varying D, particularly for the CF-CCNc (e.g. how are the diameters matched between doubly and triply charged particles and their corresponding singly charged bins - is it an interpolation between bins or an exact match of diameters at high resolution?). There can be very great sensitivity of critical supersaturation and $D_{dry,c}$ to the accuracy of the correction depending on the distribution shape.

Can the authors comment on whether the magnitude in the difference of supersaturation dependent κ shown in Figure 4b from the "fixed S" method using the monodisperse CF-CCNc can be influenced by incorrect multiple-charge correction because of the difference in how close to the modal diameter the $D_{dry,c}$ is found? Is propagation of the maximum error from multi-charge correction able to explain the discrepancy in predicted CCN activity between "fixed D_{dry}" SD-CCNc and "fixed S" CF-CCNc analyses?

Though it is noted in the text, the authors offer no suggestion as to why the $D_{dry,c}$ of particles at supersaturation 1.51$\%$ lies above those at 1.02 and 0.6$\%$ made using the CF-CCNc in ozonolysis experiments with no butanol reported in Figure 3. Can they speculate as to a cause? Is it some obvious error, or expected variability?

The figure resolution should be improved to enable them to be read at reasonable magnification.

Minor comments: p20747 line 25: The first and second parts of the sentence are not linked appropriately such that it is a non-sequitur; it should be rewritten. Also, Hamilton et al., 2011 and Jenkin et al., 2012 should be cited in the first part of the sentence.

p20748, line 19 - in this sentence, the main point leading to β-caryophyllene SOA having a higher Mw and lower O:C is that the earlier generations of oxidation product are lower volatility than monoterpene parent VOCs, so the products can condense with a lower degree of oxygenation. This should be mentioned prior to this sentence.

It would have been useful for the κ_{GF} to have been reported from measurements of subsaturated water uptake using one of the HTDMAs available at PSI for comparison with the Alfarra study. Were there no HTDMA measurements?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 20745, 2012.