

Interactive comment on “Relating CCN activity, volatility, and droplet growth kinetics of β -caryophyllene secondary organic aerosol” by A. Asa-Awuku et al.

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Received and published: 31 May 2008

“Relating CCN activity, volatility, and droplet growth kinetics of beta-caryophyllene secondary organic aerosol” by Asa-Awuku et al. (AA2008) is a very interesting study, and I would like to compliment the authors on their achievements.

The paper presents a wealth of information that can, should, and will certainly be used as a reference and input for further investigations of cloud droplet formation and growth in the atmosphere (lab, field, and model studies).

In order to facilitate further exchange and comparison, and to enhance the usefulness of the presented results, I would like to contribute the following comments and sugges-

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tions:

1) Water vapor supersaturation is the primary variable inducing and determining the CCN activation of aerosol particles in the atmosphere as well as in laboratory experiments. Only recently, Rose et al. (2008) have demonstrated that:

(a) The water vapor supersaturation values inferred by Köhler model calculations depend strongly on the type of Köhler model and on the water activity parameterisation and other parameters applied in these calculations. Specifically, the critical water vapor supersaturation values calculated for ammonium sulfate particles, which are usually used for reference and calibration, can deviate by up to 20% or more from the most accurate models available.

(b) The applicability of the flow model of Lance et al. (2006) for calculating the water vapor supersaturation in the CFSTGC depends on the experimental calibration of the instrument (non-ideal temperature offset, etc.), and that the model results can deviate substantially from measurement results, especially at low supersaturation.

Therefore, many of the quantitative results and parameters presented in AA2008 depend strongly on the experimental procedures and Köhler models applied for calibration. For example, the data points and lines in Figs. 5 and 8 (and probably also in Figs. 9, 10, and 11) could shift by up to 20% or more.

In the manuscript, however, I found little information about the calibration procedures and reference Köhler models. Moreover, the indication of uncertainty for the water vapor supersaturation in the static diffusion (SD) CCN counter appears very optimistic: plus/minus 0.02% (accuracy or precision?, full range or standard deviation?).

Thus, I would like to ask the authors to include more information about the determination and uncertainty of water vapor supersaturation in their study, and to refer to related studies addressing these issues such as Rose et al. (2008).

2) The mechanistic understanding, kinetic description, and predictability of aerosol and

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cloud properties, interactions, transformations, and effects are limited not only because of the limited availability of measurement data, but also because of the limited applicability and compatibility of model formalisms used for the analysis, interpretation, and description of heterogeneous and multiphase processes.

In fact, scientific progress in this field appears to be inhibited by a “Babylonian confusion” of terms and parameters such as “uptake coefficients” (usually symbolized by “gammas”) and “accommodation coefficients” (usually symbolized by “alphas”; Pöschl et al., 2007; and references therein).

As pointed out by Pöschl et al. (2007; Sect. 4.6, p. 6011), long-standing scientific disputes and apparent discrepancies between different studies and values reported for the “mass accommodation coefficient” of water vapor on liquid water are likely due to a confusion of different types of accommodation coefficients (“alphas”).

In AA2008, an “effective water vapor accommodation coefficient (alpha)” is introduced on p. 10110, but then the term “uptake coefficient” is used in conjunction with the symbol “alpha” throughout the remainder of the text and figures. I would like to ask the authors to consider revising the terms and symbols used in the manuscript for consistency with the existing body of literature on related topics. Specifically, I would suggest to:

(a) reserve the term “uptake coefficient” with the symbol “gamma” for normalized fluxes of net uptake;

(b) use the term “accommodation coefficient” with the symbol “alpha” only after explicitly specifying for which molecular processes it accounts (surface/bulk transport, net/gross flux, etc.) and mentioning that it should not be confused with other related parameters (see Pöschl et al., 2007);

(c) replace the potentially misleading term “accommodation coefficient” by a more straightforward term like “condensation coefficient” when water vapor condensation

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and droplet growth are described without actually resolving molecular processes at the gas-particle interface.

References:

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