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Atmospheric
Chemistry
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Discussions

# *Interactive comment on* "Cloud droplet activation: solubility revisited" *by* L. T. Padró and A. Nenes

**Anonymous Referee #1**

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General comment:

The present study shows that the agreement between modelled and measured critical/activation diameter of aerosol particles is improved by considering an "equilibrium solubility" instead of the "bulk solubility". The authors shows that the "bulk solubility" is not sufficient to activate small aerosol particles made by sparingly soluble organic species (Table 2) at some super saturations. Then, they introduce a new term: "curved-enhanced solubility (CES)" into the Kohler equation to account for the equilibrium solid-liquid in the activated droplet and obtain better estimates for the critical diameters of aerosol particles for a few organic compounds. The approach is new and the paper deserves publication after clarification of the aspects mentioned here.

Specific comments:

- This approach treats consistently from the thermodynamic point of view the solid-

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liquid equilibrium in the wet aerosol particle but this equilibrium is then treated independently from the liquid-vapour equilibrium described by Kohler equation. At least, this is the impression of the reviewer looking at the Equations 1 and 2, which contain the surface tension of the pure water (not of organic solution) and the wet diameter of the aerosol particle (not the difference between the wet diameter of the aerosol particle and the diameter of the insoluble core). I suggest to the authors to devote some effort to clarify how they treat the solid-liquid and liquid-vapor equilibriums and on which basis they treat them independently, if this is the case.

- The Equation 2 derived by Shulmann et al. (1994) was also used by other authors (Laaksonen et al. 1998; Mircea et al., 2002; Lohman et al. 2004, etc.) for modelling the hygroscopicity and activation of slightly soluble compounds taking into account the presence of the insoluble core. If the insoluble core is not considered the number of moles of water is overestimated and the Raoult effect is underestimated in Equation 3. Given that C (bulk) (calculated as &#61550;ss /Dp3, see Eq. 4) is then used in the estimation of CES, the impact of this assumption on CES should be explained. This can be also an important source for the disagreement between the activation diameters modelled without CES and the measurements

- The Kohler theory (Equations 1-3) has two parameters, which take into account the non ideality of the droplet solution: &#61550;s and &#61550;ss, the "effective" van't Hoff factors of soluble and slightly soluble species. The values of these two parameters are still a matter of debate in literature, especially for organic species. Moreover, these parameters vary with the concentration of solute and, therefore, depend on the solubility. This study does not contain any indication on the approach used for these unknowns parameters. The authors need to address in more detail this topic since the effect of &#61550;s&&#61550;ss is in competition with the effect of CES.

- Page 2328, lines12-15 state "The curvature of the initial solute cluster however can raise the solute chemical potential in the solid phase; in other words, the solute in the curved particle can have a higher solubility than its "bulk" value." This phrase is mis-

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leading. The solubility of the specie does not change, but, in an explicit treatment of solid-liquid equilibrium taking into account the droplet curvature, it changes the number of moles of solute per number of moles of water. For very small spherical drops, the number of moles of water is mainly controlled by the Kelvin effect (the effect of curvature). On the other hand, in a spherical drop, less solute is in contact with water and consequently, less moles of solute will dissolve in water. Therefore, for the same amount of water and less moles of solute, the “solubility” (no. moles solute/ no. moles water) of the specie in the spherical drop is higher than the “bulk” solubility. Such explanation is not straightforward for the readers without thermodynamic and microphysics background. We suggest to the authors to explain this concept better .

- The paper uses both the terms “slightly” and “sparingly” soluble compounds. The use of only one term will be less confusing and more consistent as long as they express the same thing. - Surface tension measurements reported in Tab 3 : at which concentration have been the measurements performed? I presume at the saturation point but please specify this in the caption. These numbers are very low similar to pure water: by comparing these data with other published data in literature large differences are evident. Please comment.

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 2325, 2007.

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