

Interactive comment on “Joint effect of organic acids and inorganic salts on cloud droplet activation” by M. Frosch et al.

Anonymous Referee #1

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The authors present laboratory results of on the CCN ability of mixed organic/inorganic particles. The presented results fit well in the discussion of previous studies that are, however, not properly discussed in the current study.

This paper is one among many others on the same topic (CCN ability of organics, mixed particles). It is clearly unfeasible to explore all possible combinations of atmospheric compositions, species and mixing ratios in particles but the current study shows once more that relatively simple assumptions lead to reasonable predictions of CCN activity. Instead of adding more complexity to the descriptions of aerosol effects on clouds, a more general description is desired and it should be clearly pointed out under what circumstances deviations from common approaches (e.g., Köhler equation) can be expected.

C6214

In addition of a discussion of the broader impacts of the results to the aerosol indirect effect, there are several unclear sections in the paper that need to be addressed before it can be considered for publication.

General comments

1) The authors show that for most systems simple assumptions based on Köhler theory lead to reasonable good predictions of supersaturation. This result should be highlighted much stronger throughout the paper (and in the abstract). Regarding the high complexity of particle compositions, it is necessary to find simplified descriptions for their representation in models. How common are the systems in the atmosphere where common assumptions fail (e.g., high fractions of NRFA, mixtures with high mass fractions of oxalic acid, ...)?

The authors use terms like ‘considerable’ and ‘significantly’ to describe deviations from Köhler equation for several systems. Can those be better quantified? In particular, the authors should mention the implications to ‘real clouds’ where it has been shown that in dynamic systems (as opposed to equilibrium situations as in CCN counters) composition effects are even further reduced (e.g., [Ervens et al., 2010; Feingold, 2003])

2) The authors explain in great detail at several places in the manuscript that a low molecular weight, high density and high degree of dissociation decrease the water activity and thus make ‘good CCN’. This concept is not new as it follows directly from Köhler equation. The effect of these parameters on water activity has been parameterized by the hygroscopicity parameter (‘kappa’) [Petters and Kreidenweis, 2007] that should be discussed here. Most of the results of the present study can be explained by the additive character of this parameter. Since kappa has been used so widely for similar systems I suggest to add a column to Figure 5 (or replace column ‘Entities per volume dry aerosol’ as it is directly proportional to kappa and is just an unnecessary additional parameter) with the corresponding kappa values.

3) The results of the current study are expressed as a function of a growth factor GF.

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The usefulness of this parameter is not really obvious to me. Here, GF is used to represent a certain solute concentration (i.e. water/solute ratio). Usually GF for different compounds are compared at the same relative humidity and thus, give a measure for the amount of water uptake under the same conditions. How meaningful is the range of GF shown in Figs 3-6 for the water uptake at activation? For each particle, a critical radius can be calculated that represents the particle size at the point of activation (cf. [Pruppacher and Klett, 2003]). What GF corresponds to the critical size of the particles?

Specific comments

p. 17982, l. 13: reword 'the more significant term in the Köhler equation'. I assume that you want to say that there were only differences in the Raoult term for the different mixtures but not in the Kelvin term.

- p. 17982, l. 19: reword 'higher effect on water activity': The relative contributions of water activity vs Kelvin effect are not explored here. Is it meant here that inorganics have a smaller Raoult term (higher kappa).

p. 17983, l. 1-3: In the abstract the error should be quantified that is introduced if no surface partitioning for these systems is assumed.

p. 17983, l. 17/18: It is true that not for all species (mixtures) CCN activity has been explored in detail. However, it should be noted here that it might not be necessary as sufficient frameworks are available (e.g., kappa) that allow a reasonable estimate.

p. 17985, l. 24 (and other places, too): Reword 'produce more species', e.g. replace by 'dissociate into more ions'.

p. 17987, l. 11: 'activates and takes up water'. A growth factor greater than 1 means that the particles take up water but do not necessarily activate yet. Activation requires that the wet particle sizes exceed the critical radius as can be derived to describe the maximum of the Köhler curve.

C6216

17990, l. 19 ff: How do the parameterizations of surface tension for other pure compounds (e.g., [Ervens et al., 2004; Tuckermann and Cammenga, 2005; Tuckermann, 2007] compare to those you show in Figure 1 and Table 3? Can they be applied in order to reduce complexity in CCN representations (i.e. avoiding additional equations)? How significant is surface tension suppression at the point of activation (i.e. where would critical radii fall on the scale of GF in Figure 3)? Is the fact that the surface tension suppression in organic/inorganic mixtures is less than for pure organics simply a concentration effect? I.e. inorganics usually contribute more water to the particle and thus, the organics can dissolve into a larger volume which results in a lower concentration.

p. 17991, l. 25 ff: is the surface tension reduction by NRFA comparable to other similar molecules (e.g., humic acid, or levoglucosan) [Tuckermann and Cammenga, 2005; Tuckermann, 2007]?

p. 17992, l. 11/12: 'water activity will be the main factor determining supersaturation'. Do you mean that the surface tension suppression is minor as compared to the effect on water activity and thus different compositions affect water activity but not surface tension?

p. 17993, l. 26 ff: this discussion is trivial and can be removed or shortened as it directly follows from Equation 1.

p. 18001, l. 2/3: AIM has been applied to many atmospheric applications. So, the statement that it was found that it indeed works sounds somewhat presumptuous.

p. 18001, l. 14 ff: Could the 'unusual behavior' of the NaCl/oxalate mixture be a result of salt formation? The hygroscopic properties of Na-oxalate might be different than for NaCl. Though I understand that the performed experiments were not designed to explore such effects, it might be worth to raise this possibility.

Technical comments

p. 17982, l. 12: negligible

C6217

References

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