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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Surfactants in cloud droplet activation: mixed organic-inorganic particles" *by* N. L. Prisle et al.

Anonymous Referee #1

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general comments:

This manuscript describes measurements of the CCN activity of lab-generated particles composed of varying proportions of NaCl and one of four surface-active organic compounds. Although such CCN experiments have been fairly commonly conducted over the past decade, the influence of surface activity on CCN activity is still poorly constrained, and to my knowledge the specific compositions described here have not been previously reported. Therefore this manuscript could potentially be published in ACPD. However, in its current form I believe this manuscript is too long and repetitive, particularly in the Results/Discussion section. It might be that, once the repetition is eliminated, the manuscript will be too short to stand on its own. The authors might want to consider combining these results with the surface tension measurements in the

Prisle et al. manuscript in preparation and resubmitting. For example, the conclusion that the assumption of surface partitioning affecting both the Kelvin and Raoult terms fits the observations is made in lines 18-19, 85-86, 364-365, 377, 393-394, 556-558, and 681-683. Also, the conclusion that particles with $w_{SFT} < 0.5$ can be well represented by σ_W (i.e., neglecting surfactant properties) is made too many times, including in lines 19-22, 86-88, 396-398, 402-406, 582-590, 596-597, and 686-689. I suggest the authors rewrite section 5, making it both shorter and more clearly organized. First, present the results, then discuss the contributions of the individual Kelvin and Raoult terms.

Section 5.4 seemed unnecessary to me, as it did not contribute significantly to the conclusions already reached in earlier sections. Even the title of this section - "Activation properties of mixed surfactant-salt particles" - suggest that this is true. Is this not the subject of the entire manuscript? Furthermore, I suggest that section 5.5.1 ("Micelle Formation in Droplets") be removed. I am skeptical that any conclusions regarding micelle formation follow from CCN data due to the low bulk surfactant concentrations present at activation. The authors point out that the cmc is only exceeded when the "pure water" approximation is used - even this is surprising to me, but I could not verify for myself because cmcs for the relevant surfactants were not given. Even assuming this is true, because the pure water assumption involves no partitioning (and associated bulk depletion) at all, I would not expect concentrations equal to or greater than the cmc in any measurements presented here. So, while I agree that "micelle formation is not an issue for the experiments in this study," I think this is simply because this study presents CCN data, and cannot be considered a result or conclusion per se. Also, the comparison to Tabazadeh (2005) points out that anionic surfactants can lower surface tension to a greater extent than naturally occurring ("HULIS") OM does, which does not require micelle formation as an explanation.

specific comments

L52: Li et al. (1998) did not study the CCN activity of single-component particles;

rather, they presented results of calculations of the CCN activity of mixed NaCl-SDS particles.

L64: I don't think "to comply with" is the right phrase to use here - something more like "to distinguish from" or "as opposed to" makes more sense to me.

L67: I think a citation of Seidl and Hanel (1983) would be appropriate somewhere in this paragraph - they pointed out the importance of the high surface:volume ratio of activating cloud droplets with respect to surface-active compounds.

L84: I suggest that the main conclusions of the manuscript be removed from the introduction.

L115: I am a little troubled by the claim that the mass fractions in Table 1 are "exact", given the author's previous point that "(i)t is an underlying assumption that the relative mass fractions of organic-to-inorganic components in the dry particles reflect the solute composition in the atomizer solution." While I agree that this assumption is reasonable, I still suggest that the authors refer to the "exact mass fractions" of the solutes in the atomized solution, not the dried particles.

L228: It is somewhat misleading to refer to an ideal solution when accounting for the salting-out effect. I suggest the authors mention that salting-out behavior is explicitly included in some of the theoretical calculations.

L328: This paragraph seems like it belongs in the introduction.

L341: I think it would be better to say that enhanced surface partitioning may decrease surface tension, rather than "increase surfactant strength" which is more ambiguous. Alternatively, the authors could define "surfactant strength."

L358: It should be mentioned that the conclusions of this section are very similar to those of Li et al. (1998).

L386: I think it would be better to say something like "either the equilibrium Köhler or

the surface partitioning models."

L390-391: Why are the words "smaller" and "larger" in quotes?

L525: I think it would be better to say "due to the limited amount of SFT relative to the large surface area" than "due to surfactant partitioning" - it is confusing because it is, in fact, surfactant partitioning that causes sigma to be reduced in the first place.

L551: Capitalize "CCN".

L593-594: I am not sure what is meant by "NaCl by mass fraction" here.

I found Fig. 5 difficult to read – too many data points on top of one another. I would recommend splitting it into four panels, one for each SFT, as was done in Fig. 3. Also, both Figs. 3 and 5 might benefit from use of a variety of symbol shapes (squares, diamonds, etc.) - hard to say without seeing it done, but it seems like it would be worth a try to see if it improves clarity.

Also, regarding Fig. 5, there are some blue (σ, p) points depicting large errors ($\sim 0.6\%/\%$) between theoretical and experimental SS. I found this confusing, as the general conclusion of the paper is that the σ, p formulation matches the experimental data well. The authors should comment on these anomalous data. Also, I only see one point in Fig. 3 above 0.4 %/% for the σ, p formulation, which makes me wonder if one of the figures has incorrectly plotted data.

Technical comments:

L207: The variables for mass fraction should be italicized.

L318, and elsewhere: Correct "Fig.s" (either "Figs." or "Fig.")

L318: Variables should be italicized here and later in the manuscript.

L371: The word "respectively" is unnecessary.

Citations

Li, Z., Williams, A., and Rood, M.: Influence of soluble surfactant properties on the activation of aerosol particles containing inorganic solute, J. Atmos. Sci., 55, 1859–1866, 1998.

Seidl, W. and Hanel, G.: Surface-active substances on rainwater and atmospheric particles, Pure Appl. Geophys., 121, 1077–1093, 1983.

Tabazadeh, A.: Organic aggregate formation in aerosols and its impact on the physicochemical properties of atmospheric particles, Atmos. Environ., 39, 5472–5480, 2005.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 24669, 2009.