

Interactive comment on “Influence of particle size and chemistry on the cloud nucleating properties of aerosols” by P. K. Quinn et al.

Anonymous Referee #2

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Reviewer comments:

This manuscript describes CCN measurements in a polluted coastal area. The CCN concentrations are combined with size distribution measurements to derive cut-off diameters for activation. These are related to measurements of chemical composition (in particular the HOA fraction). A sensitivity study compares actual CCN concentrations to CCN concentrations predicted for pure ammonium sulfate aerosols with the same size distributions.

The manuscript is based on a very interesting data set that covers a wide range of aerosol types (marine to industrial) within a relatively small geographical area. This leads to a wide variation in aerosol chemical composition and CCN cut-off diameters.

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The manuscript is very well written and all steps of the data analysis are clearly described. It contains many interesting and valuable points (e.g., comparison of chemical composition in PM₁ and CCN size range; relation of D_c and HOA fraction). It definitely has the potential to make important contributions to the CCN literature, especially with further sensitivity studies (as suggested below).

However, I have some comments that should be addressed before publication. The most important ones revolve around the shortcomings of the current and the benefit of further sensitivity studies. In the former point there is some overlap with reviewers 1 and 2.

1) Instrument calibration:

Please specify which theoretical model for calculating the critical supersaturations (S_c) of ammonium sulfate was used in the calibration. In the literature many different ways are used to derive S_c (Rose et al., 2007 and references therein). It is therefore important to be specific to ensure comparability to other papers.

Was the CCN counter also calibrated with respect to number concentration?

2) Derivation of D_c :

The critical diameter in this work is not directly measured but derived from number concentration measurements. Therefore the uncertainty of D_c should be estimated based on uncertainties in size distributions and CCN concentrations, at least for a few representative examples.

3) Correlation of D_c and HOA

This derived relationship between chemical composition and HOA fraction is an interesting result and could be very useful for parameterizations. However, HOA fractions are not very commonly measured. Therefore, it is important to present a similar correlation using the entire organic fraction. Maybe this would even give a better correlation. Or has this been tried and it did not work?

4) Sensitivity study

First part:

The first part of the sensitivity study assumes an idealized aerosol consisting of a single lognormal mode and a composition of ammonium sulfate and insoluble material. CCN concentrations are calculated as a function of mean diameter (20-140nm) and soluble volume fraction (0-1), which covers a wide range of conditions. The problem is relating this idealized study to actual data, i.e. where to place the boxes on the graph. This is made obvious by the comparison with the data of Dusek et al.: The presented simplified sensitivity study suggests roughly equal changes in CCN concentration with variation composition and size distribution for the Dusek et al. data set. However, the actual data in Dusek et al., (full size distributions, size resolved spectra) show that CCN concentrations vary much more with changes in the size distribution than with changes in chemical composition. The assertion of the authors that this sensitivity study overestimates the influence of the size distribution does therefore seem questionable. I think there are many reasons why it is problematic to superimpose actual data on this plot.

As reviewer 2 pointed out, the author's own results show that soluble volume fractions cannot simply be equated with HOA fraction. Soluble volume fractions can also not be equated with the total organic fraction as is done in the comparison with Dusek et al.. And especially to equate it with HOA fraction for one study and with OC fraction for another study will lead to inconsistencies, as also noted by reviewer 1. One of these inconsistencies is that the cut-off diameters at $S=0.44\%$ for this experiment lie most frequently between 60 and 100nm (Figure 3b grey area). This overlaps well with cut-off diameters at 0.4% in Dusek et al., so it is hard to understand why the respective boxes are not overlapping in Figure 6.

In summary, why use epsilon at all and not just D_{gn} on one axis and D_c on the other axis (now epsilon)? Then the comparison to the data would be much more straightforward.

A last problem is that a single lognormal mode is not a good representation of the actual

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size distribution. In the case of a bimodal size distribution, a lot of the size distribution variability can lie in the relative fraction of Aitken and Accumulation mode particles. In this case a shift in D_g is not necessarily representative of the variability of the whole size distribution. This might contribute to the observed underestimate of the role of the size in Dusek et al. by the current study. If the first part of the sensitivity analysis is retained (with D_c instead of epsilon), this potential shortcoming should be discussed in the text.

Second part:

In a second part of the sensitivity study, the 'error' of using ammonium sulfate to predict CCN concentrations is assessed. In my opinion this should only be a first exploratory step. It is obvious from Figure 6 that the 'errors' are always positive, which indicates that the model is biased. Therefore this sensitivity study says mainly that pure ammonium sulfate is not a good model of the aerosol in this region. This is a valid result, but definitely not the most interesting that could be obtained with this data set.

The beauty of these data is that they cover a very complex region, where marine air and extreme pollution can be found in close proximity. From the data of Dusek et al., it seems that if marine air is advected over Germany for one day the cut-off diameters are already more 'continental' than 'marine'. This is not the case here. A very important question is how much detail in chemistry is needed to predict CCN concentrations in such an environment. I therefore suggest to repeat this sensitivity study using the mean cut-off diameter for the whole region as a reference (instead of ammonium sulfate). Further sensitivity studies could use mean cut-off diameters of the four sub-regions and the different air mass cases. The results could simply be summarized in a table, because the comparison with Dusek et al., and Hudson et al., would of course not be meaningful. (It was already not very convincing to present the errors these authors would have made, had they chosen to model their aerosol with ammonium sulfate, which they obviously never did).

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In any case, I encourage the authors to use D_c instead of epsilon to avoid the problems raised by me and the other reviewers. If they choose not to do this, I second the view of reviewer 2 that some entirely different sensitivity study should be done. In my opinion the data and analysis are also publishable without any sensitivity study at all, but I would find it a pity because of the great potential of this data set.

Specific comments:

p14184, line 1-5: It is important not to mix CCN activation in an instrument where the supersaturation is fixed and droplet activation in an actual cloud. In the present study the supersaturation is fixed in the instrument as well as in the calculation of CCN. If these CCN concentrations are modeled using a size distribution with constant sigma they will be less variable than otherwise. Therefore holding sigma constant will underestimate the impact of particle size on CCN at a certain S , as used in this study.

p14186, line 24: Is this loading significant? What would be a possible reason that D_{gn} could be related to D_c ?

p14189, line 20-25: It is not correct to superimpose the D_{gn} determined at 60% on this plot, because as far as I can tell the modeled size distributions were assumed to be dry. Instead of correcting the data of Dusek et al, and Hudson et al., D_{gn} of this study should be shifted to lower diameters.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 14171, 2007.

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