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Holger Tost
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Re: Response to Reviews
Manuscript Number: acp-2016-1044
Manuscript Title: Metrics to quantify the importnce of mixing state for CCN activity
Manuscript Authors: J. Ching, J. Fast, M. West, and N. Riemer

1 Response to Reviewer #2's comments

We thank the reviewer for their comments and suggestions. We revised the manuscript accordingly with changes marked in blue. Our responses are as follows:

(2.1) Importance of the mixing state metrics: My major concern is how the new metrics (χ) will help to quantify the mixing state effect? In this study, the determination of mixing state effect was done by comparing CCN predictions of cases with and without composition averaging. If I understand correct, it means that the mixing state effect is determined without the metric χ. So, why would we need such a parameter if it is not even used?

> The reviewer is correct—since we have all the per-particle information we can determine the error, and if this was all we wanted to do then the metric χ (or any other mixing state metric) is not needed.
>
> However, our goal is to relate the error in CCN concentration due to the internal mixture assumption to a quantitative measure of mixing state. The paragraph in the introduction, p. 2, line 13 states this goal, and we added text to clarify this further, p. 2, lines 18–20: "The central question that we address is: For aerosol populations of a given mixing state, what magnitude of errors can we expect for estimating CCN concentrations when assuming that the population is internally mixed?"

(2.2) Performance of the mixing state metrics: One of my questions during my reading is that if a single χ corresponds to a unique error in CCN predictions and if it can be used in the CCN prediction or even better than existing parameters. The authors answered my first question, and showed that the relationship of χ and the error in CCN predictions is not unique. According to the size-resolved hygroscopicity distribution in Fig. 4, there are two kappa modes and my feeling is that the fraction of the low hygroscopic mode (F_{LH}) is a critical parameter for the errors when neglecting the mixing state information. Could you make similar plots as in Fig. 6 and Fig. 7 but using F_{LH} instead of χ? If the error shows more converged dependence on F_{LH}, χ may not be a better parameter for the CCN prediction. Besides, χ is hard to determine in practice by available measurement techniques.

> Thanks for this suggestion, which together with reviewer's point 2.3 inspired us to add another section to the paper. We think that including F_{LH} and the geometric standard deviation of the κ-distribution in the discussion will answer the questions that many readers might have.
>
> The new section (Section 6) is titled "Relationship of χ and CCN error to other metrics of hygroscopic mixing state", and we added two figures.
>
> Figure 10 shows the relationship of (a) size-restricted mixing state parameter χ_{res} and mixing state parameter, χ, (b) size-restricted mixing state parameter χ_{res} and number fraction of particles with low hygroscopicity, F_{LH}, and (c) size-restricted mixing state parameter χ_{res} and geometric standard deviation of the κ-distribution, σ_κ, for all 384 aerosol populations in $\mathbb{P}$.
>
> Figure 11 is analogous to Figure 6 and shows (a) Relative error $\epsilon(s_{\mathrm{env}}, \chi_{\mathrm{res}})$, (b) relative error $\epsilon(s_{\mathrm{env}}, F_{\mathrm{LH}})$, and (c) Relative error $\epsilon(s_{\mathrm{env}}, \sigma_\kappa)$ for each individual aerosol population in $\mathbb{P}$, evaluated for 20 supersaturation values between 0.05% and 1%.
>
> We also added text in the conclusions, p. 18, lines 19–23: "We also explored the relationship of CCN error other measures of mixing state, specifically a size-restricted χ, the fraction of particles with low hygroscopicity, and the geometric standard deviation of the κ-distribution. These other measures also capture aspects of the heterogeneity of the particle population and the dependence of CCN error on these quantities are qualitatively similar to the one when using χ. However, χ has advantages as a mixing state metric due to its defined range (0 to 100%) and well-defined extremes (0% is fully internally mixed and 100% is full externally mixed)."

(2.3) Comparison of χ to existing parameters: χ is a single parameter containing more intensive information. The authors have nicely presented its general concept by a nice illustration of Fig. 1. But it is still hard to fully understand it. Can you plot the series of χ and compared it to other well-established parameters, e.g., F_{LH}, or the (geometric) standard deviation of kappa distribution, etc.? Does a higher χ correspond to a larger F_{LH} or a smaller standard deviation? The potential link to other mixing state parameter may help people to accept the new parameter.

This was addressed in conjunction with the response to comment (2.2), and forms the content of the new Section 6.

(2.4) Design of experiments and discussion: In this study, the performance of χ is evaluated by comparing the error with kinds of averaged diversity value over the whole size range. I suggest the authors to reconsider this. The errors in CCN prediction are controlled by multiple parameters, i.e., the evaluated supersaturation, the size distribution and the kappa distribution. We know that the particle size has a dominant effect on the CCN activation. But if we want to quantify the effects of particle size on the CCN prediction, can we plot the error against the averaged particle size as what was done for χ? It is not clear what's the better solution but maybe if the authors could try to used size-resolved χ and check how to use it in CCN prediction or parameterization, e.g., maybe there is a compact empirical relation between χ and the averaged activation fraction at each size.

We did explore what the error distribution would look like if size-resolved composition information was retained. This is described on p. 14, lines 17–26. We did not include a figure for these results because they are qualitatively similar to Figure 6.

In the new Section 6, we now also added some material to show what happens if χ is calculated based on a size restricted population (Figure 10a and 11a, see response to comment (2.2) for details). Again, the error distribution looks qualitatively very similar to Figure 6.

(2.5) Abstract "However, it has been difficult to rigorously investigate this assumption because appropriate metrics for mixing state were lacking"

I think the kappa distribution and the corresponding parameters (mean kappa, mode kappa, and standard deviation) in Su et al. (2010) may be as good as χ in representing the CCN-relevant mixing state.

We agree with the reviewer and removed this sentence.

(2.6) Page 8 ln 10, Can the authors specify which kappa values were used for the two surrogate groups and how to calculate kappa for internally mixed particle?

We added the specifics about this on p. 8, lines 11/12– p. 9, lines 1–4:

"Since we track the composition evolution of each individual particle throughout the simulation, we can calculate the critical supersaturation s_{c} for each particle as described in Riemer et al. (2010), using the concept of the dimensionless hygroscopicity parameter κ (Petters and Kreidenweis, 2007). The overall κ for a particle is the volume-weighted average of the κ values of the constituent species. Based on Petters and Kreidenweis (2007) we assume $\kappa = 0.65$ for all salts formed from the $\mathrm{NH_4^+} - \mathrm{SO_4^{2-}} - \mathrm{NO_3^-}$ system. For all MOSAIC model species that represent SOA we assume $\kappa = 0.1$, and for POA and BC we assume $\kappa = 0.001$ and $\kappa = 0$, respectively."

Note that we do not assign kappa values for the surrogate species as such, but calculate the overall κ for a particle as the volume-weighted average of the κ values of the constituent species, and assign κ values of the constituent species as specified above.

References

M. D. Petters and S. M. Kreidenweis. A single parameter representation of hygroscopic growth and cloud condensation nucleus activity. *Atmos. Chem. Phys.*, 7:1961–1971, 2007.

N. Riemer, M. West, R. Zaveri, and R. Easter. Estimating black carbon aging time-scales with a particle-resolved aerosol model. *J. Aerosol Sci.*, 41:143, 2010.