Comment on "Metrics to quantify the importance of mixing state for CCN activity" by Ching et al.

This manuscript presented particle-resolved model simulations to quantify error in CCN predictions when mixing state information is neglected. The authors classified the chemical model species according to hygroscopicity, defining two surrogate species, a low hygroscopicity group (black carbon and primary organic aerosol) and a high hygroscopicity group (inorganic and secondary organic aerosol species), and investigated the error for eight urban plume scenarios. The error was analyzed against the mixing state index (χ), a parameter proposed by Riemer and West (ACP, 13, 11423-11439, 2013) to describe the degree of external and internal mixing of aerosol populations. The results show that neglecting the mixing state information has less influence on the CCN prediction for more internally-mixed aerosol populations than more externally mixed aerosol particles. The relationship of χ and the error in CCN predictions is not unique and the reasons have been discussed. I would recommend publication if the authors could address my comments as listed below.

General comments

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?

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.

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 a standard deviation? The potential link to other mixing state parameter may help people to accept the new parameter.

4. Design of experiments and discussions

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.

Minor comments:

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.

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Can the authors specify which kappa values were used for the two surrogate groups and how to calculate kappa for internally mixed particle?

Ref:

Su, H., Rose, D., Cheng, Y., Gunthe, S., Massling, A., Stock, M., Wiedensohler, A., Andreae, M., and Poschl, U.: Hygroscopicity distribution concept for measurement data analysis and modeling of aerosol particle mixing state with regard to hygroscopic growth and CCN activation, Atmospheric Chemistry and Physics, 10, 7489-7503, 10.5194/acp-10-7489-2010, 2010.