

Joseph Ching
ppching@gmail.com

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Holger Tost
Editor, Atmospheric Chemistry and Physics

Re: Response to Reviews

Manuscript Number: acp-2016-1044

Manuscript Title: Metrics to quantify the importance of mixing state for CCN activity

Manuscript Authors: J. Ching, J. Fast, M. West, and N. Riemer

1 Response to Reviewer #1's comments

We greatly appreciate the reviewer's comments. We revised the manuscript accordingly with changes marked in blue. Our responses are as follows:

(1.1) Figure 1 and Table 1 and 2 are 100% identical to figures and tables in Riemer et. al. 2013. Even with the authors and the Journal being the same I would consider this a questionable practice. The least the authors can do is to add a note to the captions saying "Taken from Riemer et. al. 2013".

We updated the captions to Figure 1 and Tables 1 and 2 accordingly.

We also added text to emphasize that we summarize the important key points from Riemer and West (2013) for the convenience of the reader:

- page 3, line 4: "The salient points are summarized as follows."
- page 3, lines 7–8: "From this quantity, all other mass-related quantities can be defined, as detailed in Riemer and West (2013) and here listed in Table 1, and the diversity metrics can be constructed as shown in Table 2."

(1.2) Most presented findings are statistical observations. I am convinced it would add great value to the manuscript if the authors discussed potential physical origins of their findings. E.g. why do different populations experience different amounts of error cancellations (page 12, line 9)? What physical characteristic might explain the dependence of the relative error on supersaturation threshold (page 12&13)?

We have made a number of changes and clarifications to the paper in an attempt to explain the physical origins of our findings. We renamed Section 5 to "Relationship of error in CCN concentration and mixing state index χ " to highlight that this is where we will discuss this relationship. In this section we discuss Figure 5, which clarifies that "Error is caused only by the *difference* in the number of particles that activate in the reference case, but do not activate after composition averaging [...], and the cases that activate after composition averaging, but did not activate in the reference case."

In this section we also explain the physical basis of zero error for internally mixed populations as the fact that "For populations with $\chi = 100\%$, the particles' composition is the same across the entire population, and hence [...] the number concentrations in quadrants B and D are zero." Correspondingly, we explain the physical basis of the error for partially-internally mixed populations as depending on "both the extent of distortion of the critical supersaturation distribution and the supersaturation threshold."

To explain the dependence of the relative error on the supersaturation threshold, we clarified that "for a given supersaturation threshold and a given χ value, different populations experience different amounts of the error cancellations" described above. A detailed investigation of this is provided by Figure 8, which together with Figure 5 provides physical understanding of the cancellation phenomenon. We explained in the paper that "the change in particle number concentration per supersaturation interval is smaller for s_c of 0.1% compared to $0.3 < s_c < 1.5\%$ " due to the shape of the population \mathbb{P} in Figure 5.

(1.3) Figure 7: the x-axis looks like a ratio rather than %. Also, the caption mentions insets which do not seem to be there.

Thanks for pointing this out. We fixed both of these issues.

References

N. Riemer and M. West. Quantifying aerosol mixing state with entropy and diversity measures. *Atmos. Chem. Phys.*, 13:11423–11439, 2013.