1 Response to Reviewer #2

We greatly appreciate the reviewer's comments. Our responses are as follows:

(2.1) The mixing state framework appears to be primarily built around diversity measures. This is intuitive and the tables and figures support the use of diversity measures in quantifying mixing state beyond simply the extremes of internal and external mixing. However, I stumbled to see how the use of entropy helped to advance this understanding? It was not clear in Section 2 whether entropy was a completely parallel concept with diversity, or whether part of the diversity argument was mathematically based entropy. Further clarification of unique information that is gained from entropy measures should be included to justify its use in the title/abstract. I think this would help alleviate some of the confusion that it brings forth as many readers will be struggling to relate the thermodynamic interpretation of entropy to its use in this analysis.

> *We agree with the reviewer that it is possible to define the mixing state index χ only in terms of diversities, and to thereby avoid introducing the concept of entropy at all in this paper, as entropy and diversity are completely parallel. We also agree that there is a potential for confusion if a reader tries to use the thermodynamic definition of entropy while reading this paper, rather than the information-theoretic or statistical-mechanical definition. Nonetheless, we feel that it is crucial to discuss both entropy and diversity for two reasons:*
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> 1. *The entropy definitions and concepts are necessary to understand the history and related literature of mixing state measures in many fields of science and mathematics.*
> 2. *The key properties of the mixing state index χ, which make it a powerful concept, arise directly from the underlying entropic definitions. While we could formally write the proofs only in terms of diversities, this would obscure the reasons that they are true in terms of the conventional entropy properties.*
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> *To attempt to minimize the possibility for confusion, we have:*
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> 1. *Clarified in the introduction that we are working with information-theoretic entropy (not thermodynamic entropy):*
>   - *On page 3, line 24 of the updated manuscript, we now write: "In this paper we present the first quantitative measure of aerosol mixing state, the mixing state index χ, based on diversity measures derived from* **the information-theoretic** *entropy of the chemical species distribution among particles."*
>   - *On page 3, line 26, the sentence now reads: "The measurement of species diversity and distribution* **using information-theoretic entropy measures** *has a long history in many scientific fields."*
> 2. *Added a remark (page 5, lines 21–23): "Note that entropy and diversity are equivalent concepts, and that either could be taken as fundamental. We retain both in this paper to enable connections with the historical and current literature."*

(**2.2**) The authors are commended for tackling this challenging question. It would be incredibly useful to the reader to have an assessment of how the properties that depend on mixing state (e.g., optical, water uptake, trace gas reactivity) would be incorporated into such a model. There are few laboratory experiments that are run on complex, known mixtures spanning ranges in mixing state. Is this a call for more laboratory studies on these types of system? Even if this information were available, how challenging would this be to implement (e.g., each particle in the population having a different value for hygroscopicity)?

> *The question of how the dependence of optical properties, CCN activities, etc., on mixing state can be explored within this framework is extremely interesting and indeed challenging. Now that we have this framework in place, we have started tackling this question, but the results are still*

too premature to be included in this paper, and also beyond its scope, so we need to refer to future publications that will elucidate this very question.

Controlled laboratory studies on aerosol populations with evolving mixing state would indeed be very useful, and following the reviewer's suggestion we included a comment on page 16, lines 16–18, to point this out: "In this context, it would be very useful to obtain quantitative information on per-particle composition from field observations and laboratory experiments, together with measurements of application-relevant bulk properties."

To answer the reviewer's last question ("how challenging would this be to implemement?"): Fortunately this would be straightforward. With PartMC-MOSAIC we do track the per-particle composition explicitly, and based on this information we can calculate values of hygroscopicity, scattering cross section, absorption cross section, etc. for each particle. We have done this in several studies already. We included a sentence to clarify this on page 8, lines 4–7: "Note that from the information on per-particle composition it is straightforward to calculate per-particle properties, such as hygroscopicity [Riemer et al., 2010, Zaveri et al., 2010, Ching et al., 2012, Tian et al., 2013], optical properties [Zaveri et al., 2010], or particle reactivity [Kaiser et al., 2011]."

References

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