Responses to Reviewer #1 Comments

General comments:

This manuscript systematically evaluates the feasibility and applicability of the YZ method proposed by Yuan and Zhao (2023) for retrieving the aerosol mixing state index (χ) from H-TDMA data, using PartMC-MOSAIC simulations. The results show that the YZ method is a promising approach for investigating aerosol mixing states in urban environments. Importantly, this work offers a valuable insight into the assessment of χ without direct reliance on expensive and limited single-particle mass spectrometry or electron microscopy observations and will be useful for improving our understanding of aerosol mixing states and their evolution, as well as for discussing their implications for radiative forcing and aerosol activation. In this respect, the work is both valuable and encouraging. Overall, this manuscript is generally well written and logically structured. I recommend publication after the minor revisions, as outlined below:

> We appreciate the reviewer for dedicating their time to review our paper and for the constructive and encouraging feedback. We have revised the manuscript to clarify the validation library, justify the discussion of sea-salt-containing aerosols, and expand the discussion of the atmospheric implications of χ. The line numbers referenced below correspond to the revised clean manuscript.

Specific Comments:

1. The scenario library employed for validation in the PartMC-MOSAIC simulation appears to be largely representative of urban environments and does not include primary sea-salt emissions. Likewise, the YZ method was originally developed on the basis of urban H-TDMA observations to infer aerosol hygroscopic mixing state. In this context, the close agreement between the χ values derived from the simulations and those retrieved by the YZ method already lends support to the validity of the underlying binary assumption. In Sect. 5.3, the authors further discuss the limitations of the YZ method in the presence of sea-salt aerosols, mainly through conceptual and theoretical arguments. I am not fully convinced that sea-salt-containing aerosols constitute the most appropriate example for illustrating the limitations of the YZ method. Sea-salt are typically coarse particles and relatively short-lived, and they may therefore mix less extensively with other components than fine-mode urban aerosols. As a result, χ may not be the most informative metric for characterizing their mixing state, even at coastal sites. This interpretation also appears broadly consistent with the authors' own χ calculations for the four regions using the YZ method. Since χ was developed primarily to describe the hygroscopicity of multi-modal continental aerosols, I suggest that the discussion in Sect. 5.3 be reframed more carefully, with a clearer justification for if sea salt should be added here to discuss the limitation of the YZ method.

> We appreciate this comment and agree that the scope of the validation library and the motivation for discussing sea salt needed to be clarified. The PartMC-MOSAIC validation library used here is primarily representative of continental anthropogenic aerosol populations and does not include primary sea-salt emissions. Therefore, the model-based evaluation should not be interpreted as a validation of the YZ method for marine-aerosol-dominated populations.
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> We have revised Section 5.3 to make clear that sea salt is not presented as the central application case for the YZ method, nor as a component represented in the validation library. Instead, it is used as an example of a broader end-member limitation of the binary hygroscopicity framework. For continental aerosol, the prescribed more-hygroscopic end member represents typical inorganic salts such as sulfate, nitrate, and ammonium. In marine-influenced environments, however, submicron sea-salt particles may be present and may have hygroscopicities that extend beyond this prescribed continental end-member range. In that case, the binary mapping from κ to LH–MH composition becomes less unique, and the retrieved χ^{YZ} may become sensitive to the assumed end-member values.
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> To avoid overinterpreting the sea-salt example, we reframed the discussion in Section 5.3 as an end-member sensitivity/limitation rather than as a realistic marine aerosol simulation. We now explicitly state that the $\kappa_{\mathrm{MH}} = 1.28$ test is a diagnostic sensitivity test performed on the same PartMC aerosol populations, not a validation of the method for sea-salt-containing aerosol. We also state that applications at

coastal sites should be interpreted cautiously when marine aerosol influence is substantial and independent measurements are not available to constrain the abundance and particle-level mixing state of sea salt.

2. One key element of the YZ method is the specification of the κ values assigned to the two components in the binary system. In the present manuscript, the authors extend the original parameter settings from κ_{LH} =0.01 and κ_{MH} =0.60 to κ_{LH} =0 and κ_{MH} =0.65. In the real atmosphere, however, the κ value of the more hygroscopic component may vary over a certain range, depending in particular on the abundance of each inorganic salt. Could the authors include a sensitivity analysis to quantify how uncertainties in κ MH propagate into the retrieved χ? For example, if κ_{MH} varies from 0.60 to 0.65, or even to 0.70, how large would the resulting deviation in χ be? Would such changes increase or decrease the discrepancy between the YZ retrieval and the PartMC-MOSAIC results? Addressing this point would provide a useful practical reference for other researchers when selecting appropriate parameter bounds for the analysis of actual H-TDMA observations.

Thank you for the suggestion. We have added a sensitivity analysis in which the YZ retrieval is repeated using κ_{MH} = 0.60, 0.65, and 0.70, while keeping all other aspects of the retrieval unchanged. The resulting comparison is shown in Figure S1.

The results show that the retrieved χ^{YZ} is sensitive to the assumed more-hygroscopic end member in the expected direction. Decreasing κ_{MH} from 0.65 to 0.60 tends to shift particles with high hygroscopicity closer to the pure-MH end member and therefore lowers the retrieved χ^{YZ}, leading to an underestimation relative to χ^{PMC}. Increasing κ_{MH} to 0.70 has the opposite effect: particles with a given κ are interpreted as containing a smaller MH fraction and a larger LH fraction, which increases the inferred per-particle diversity and tends to overestimate χ^{YZ}. Across the tested values, the correlations remain high, but the mean absolute error increases when κ_{MH} differs from the baseline value of 0.65. Specifically, the MAE increases from 1.6%–2.4% for the baseline κ_{MH} = 0.65 case to 7.2%–11.8% for κ_{MH} = 0.60 and 10.5%–17.3% for κ_{MH} = 0.70, depending on particle diameter. This confirms that the choice of κ_{MH} is important for minimizing systematic bias, while also showing that the retrieval remains qualitatively robust for plausible continental inorganic-salt hygroscopicities.

We made the following changes:

- L175–185: The retrieval framework assumes a representative hygroscopicity for the inorganic-salt component. To assess the sensitivity of the retrieval to this assumption, we repeated the analysis using κ_{MH} values of 0.60, 0.65, and 0.70 (Figure S1). The sensitivity arises because the inferred MH volume fraction in each κ bin depends directly on the assumed end-member values through Eq. (1). For a given particle hygroscopicity, decreasing κ_{MH} increases the inferred P_{MH} and shifts particles with hygroscopicities near the MH end member closer to pure-MH composition, leading to a systematic underestimation of χ. In contrast, increasing κ_{MH} decreases the inferred P_{MH}, causing particles that would otherwise be assigned closer to the pure-MH end member to be interpreted as mixtures of LH and MH components, resulting in a systematic overestimation of χ. Across particle diameters, the MAE increases from 1.6%–2.4% for the baseline κ_{MH} = 0.65 case to 7.2%–11.8% for κ_{MH} = 0.60 and 10.5%–17.3% for κ_{MH} = 0.70 (Figure S1). Nevertheless, strong correlations are maintained across all tested values, indicating that the retrieval is robust to reasonable uncertainty in the representative hygroscopicity assigned to inorganic salts, although accurate specification of κ_{MH} is important for minimizing systematic bias.

3. Sect. 4.4 and Fig. 4 clearly demonstrate that aerosol populations may share a similar bulk mean hygroscopicity while exhibiting substantially different mixing states. This is important and the manuscript would benefit from a somewhat fuller discussion about the impacts of χ on aerosol radiation forcing and cloud droplet activation.

Thank you for the suggestion. We have expanded the discussion of the atmospheric implications of hygroscopicity-based mixing state. Specifically, we clarify that aerosol populations with similar bulk hygroscopicity but different χ values may exhibit different cloud activation behavior because hygroscopic

material is distributed differently among particles. This can affect CCN number concentrations and, consequently, aerosol indirect radiative effects. We focus this added discussion on cloud droplet activation, which is the implication most directly connected to the κ-based mixing-state comparison in Section 4.4.

We made the following changes:

- L226–234: "The distinction between aerosol populations with identical κ_{mean} but different mixing states may have important implications for aerosol-cloud interactions. Activation of a particle depends on whether its hygroscopicity is sufficient to lower its critical supersaturation below the ambient supersaturation (Petters and Kreidenweis, 2007). In an externally mixed population (lower χ), a substantial fraction of particles are dominated by less hygroscopic species, such as black carbon or primary organic aerosol, yielding high critical supersaturations that may exceed the maximum supersaturation reached in clouds. These particles are therefore unlikely to activate as cloud condensation nuclei (CCN). In contrast, a more internally mixed population (higher χ) with the same κ_{mean} distributes hygroscopic material more uniformly across all particles, lowering the critical supersaturation for a larger fraction of the population and increasing the number of CCN-active particles. As a result, aerosol populations with similar κ_{mean} but different χ values may exert different indirect radiative effects."

Minor Comments:

1. Line 9: The term 'true mixing state' may be somewhat misleading. On the one hand, it may be interpreted as referring to the actual atmospheric mixing state; on the other hand, it may inadvertently imply that the YZ method-derived result is "untrue". I suggest replacing it with a more neutral and precise term.

Thank you for pointing this out. We have replaced this phrase with more precise wording, "particle-resolved reference mixing state index".

We made the following changes:

- L9–10: Replaced "true mixing state index" with "particle-resolved reference mixing state index" in the Abstract.
- L153/L155/L170: Replaced similar wording with "reference mixing state".

2. Line 150: It would be helpful if the authors could provide the κ-PDF diagnosed from the model output.

Thank you for the suggestion. We note that Figure 3a and 3b already provides examples of κ-PDFs generated from the PartMC validation library. To make this clearer, we have revised the figure caption to explicitly state that these panels show representative PartMC-derived κ-PDFs used to evaluate the retrieval.

- Figure 3 caption: "(a and b) Representative PartMC-derived κ-PDFs from the validation scenario library for two aerosol populations with high secondary organic aerosol (SOA) content at a dry diameter of 150 nm, corresponding to predominantly externally mixed (a) and internally mixed (b) particle populations."

3. Fig. 1: Please consider adding numerical values of the correlation coefficient, MAE, or RMSE, to the individual panels, so that the size dependence of the retrieval performance can be seen more directly.

Thank you for the suggestion. We have added quantitative performance metrics to Figure 1. Each panel now reports the correlation coefficient and the mean absolute error for that dry diameter.

4. Fig. 4: Based on the current description, I found the color distribution somewhat difficult to interpret. Why are there still green dots in the yellow region? If a higher dot density indicates a larger number of particles sharing the same P_{MH}, would the color not be expected to become more distinctly yellow? A brief clarification of the color mapping would be helpful.

Thank you for pointing this out. Each dot in Figure 4 represents a particle population from the PartMC simulation, and the color denotes its number concentration. The position of the dot suggests it's P_{MH} and χ^{PMC}, with a small random jitter added solely to improve visualization and reduce marker overlap. We have revised the caption to clarify the description.

- Figure 4 caption: "Strip plot of more-hygroscopic (MH) component volume fractions ($P_{\mathrm{MH},j}$) for particles at D_p = 150 nm. The seven aerosol populations shown have identical mean hygroscopicity (κ_{mean} = 0.31) but different mixing state indices (χ^{PMC}). Each dot represents a particle population from the PartMC simulation, and its color indicates number concentration. Dots are grouped by the χ^{PMC} value of each aerosol population, with a small horizontal jitter applied only for visualization."

5. Data Availability: If feasible, I encourage the authors to consider making the model simulation dataset publicly available, including the particle sizes and chemical compositions of the 4900 aerosol populations. Such a dataset could serve as a valuable benchmark for future algorithm development and intercomparison, much like H-TDMA observational datasets.

Thank you for the suggestion. The simulation dataset used in this study had already been deposited in the Illinois DataBank. In response to this comment, we have expanded the archived dataset to include the particle-resolved chemical composition of the 4900 aerosol populations, in addition to the particle size and the LH/MH spcecies volume fractions. The repository link is provided in the Data Availability section and will become publicly accessible upon publication.

We also note that the validation scenario library is designed to be extensible and can be regenerated or customized for different applications. The underlying tools are publicly available on Github, including the PartMC model (`https://github.com/compdyn/partmc`) and the scenario library generation framework (`https://github.com/zhonghua-zheng/code_scenario_generator`).

References

Petters, M. D. and Kreidenweis, S. M.: A Single Parameter Representation of Hygroscopic Growth and Cloud Condensation Nucleus Activity, Atmospheric Chemistry and Physics, 7, 1961–1971, https://doi.org/10.5194/acp-7-1961-2007, 2007.