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:

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.

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.

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.

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.

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

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.

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.

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.