Responses to Reviewer #2 Comments

The manuscript presents a multi-site analysis of aerosol mixing state inferred from size-resolved HTDMA hygroscopicity measurements. The compilation of observations from different environments is potentially valuable, and the attempt to provide observational constraints on aerosol mixing state is relevant to Atmospheric Chemistry and Physics. The manuscript is generally well organized and the analysis provides useful information on the size and seasonal dependence of hygroscopicity-derived mixing state. However, several methodological and interpretational issues currently limit the robustness and generality of the conclusions. I therefore recommend major revision.

> We thank the reviewer for the careful and constructive evaluation. The revised manuscript presents the study as a multi-site synthesis rather than a climatology. It also clarifies that χ is a hygroscopicity-derived metric calculated using prescribed binary end members. We expanded the descriptions of the sites and measurement datasets, added dataset-specific sensitivity analyses, and incorporated available bulk-composition measurements, particle number-size distributions, diurnal analyses, and back trajectories. We used this additional evidence to reassess the original process interpretations and limit the conclusions to those supported by the observations. Researchers who contributed the MHD and HACPL observations and their interpretation have joined the manuscript as co-authors. The page and line numbers referenced below correspond to the marked-up version of the revised manuscript.

Major comments:

1. The use of the term "climatology" appears stronger than warranted by the temporal coverage of the datasets. The eight sites were sampled during substantially different periods, ranging from several years at MHD and SGP to approximately one year at several locations and only November 2016 and May–June 2017 at BJIAP. CRG also lacks a complete annual cycle. Consequently, site differences, seasonal variability, interannual variability, and long-term changes in aerosol composition are intrinsically mixed together. The present dataset therefore represents a useful multi-site synthesis, but its ability to establish a coherent climatology is limited.

> We thank the reviewer for identifying this limitation. We agree that differences in record length, calendar year, and seasonal coverage prevent the compiled observations from constituting a coherent climatology or supporting an assessment of long-term change. We have therefore reframed the study throughout the manuscript as a multi-site synthesis and interpret the reported patterns within the available observations. We reserve 'year-to-year variability' for the multi-year SGP record and limit the CRG and BJIAP discussions to their available sampling periods. Following clarification from the MHD co-authors, we also replaced the previously used dataset with their full-year 2009 HTDMA record.

2. The retrieved χ is fundamentally conditional on the assumed binary LH–MH representation of aerosol composition. Real atmospheric particles contain complex mixtures of organics, sulfate, nitrate, ammonium, black carbon, sea salt, dust, and other components, and chemically distinct particles can exhibit similar hygroscopicities. Therefore, a κ distribution does not uniquely constrain chemical composition or chemical mixing state. Several parts of the manuscript appear to interpret the hygroscopicity-derived χ as though it were equivalent to a composition-resolved aerosol mixing-state metric, which requires greater caution.

> We thank the reviewer for highlighting this distinction. We agree that the retrieved χ describes mixing with respect to hygroscopicity under the prescribed binary representation and cannot establish how chemical species are distributed among particles. We have made this distinction explicit throughout the manuscript. We also removed interpretations that treated κ-PDF shape or high χ as evidence of chemical homogeneity or specific chemical components.

3. A major concern is the comparability of χ among different environments. Continental and coastal sites use κ_{MH} = 0.65, whereas ENA and MHD use κ_{MH}= 1.5. The manuscript explicitly states that values retrieved at ENA and MHD

are not directly comparable with those obtained using $\kappa_{MH} = 0.65$ because of the different end-member assumptions. Nevertheless, the Abstract and Conclusions compare marine, urban, and continental environments and conclude that marine aerosol tends to be more externally mixed and more variable. This creates an important inconsistency between the methodological limitations acknowledged by the authors and the cross-environment conclusions drawn from the results.

> We thank the reviewer for identifying this inconsistency. We removed quantitative marine–continental comparisons from the Abstract, Section 4.1, and the Conclusions. ENA and MHD are now interpreted only in terms of within-site seasonal and size-dependent variation. Sections 2 and Conclusion explicitly state that their absolute χ values are not compared quantitatively with values obtained using $\kappa_{MH} = 0.65$.

4. The datasets are not fully harmonized in terms of instruments, data-processing procedures, particle sizes, and original products. Five datasets are derived from ARM processed HTDMA data, BJIAP growth factors are converted to κ using κ-Köhler theory, and MHD and HACPL κ-PDFs are reconstructed from previously published distributions. These methodological differences may influence the width and shape of κ-PDFs and consequently propagate into D_α, D_γ, and χ. This issue is particularly relevant because the major conclusions are based on relatively subtle differences among sites and particle sizes.

> We thank the reviewer for this comment. To maximize consistency, we applied the same daily retrieval procedure to all datasets after constructing the κ-PDF. Some upstream differences could not be removed retrospectively because the archived datasets were produced using different instrument configurations, inversion methods, particle-size selections, and levels of processing. We document these differences in Section 3.1 and the processing applied to each dataset in Table S1. We also evaluated sensitivity to relevant analysis choices, as described in Section 3.2. In interpreting the results, we emphasize seasonal and size-dependent patterns within each site and do not interpret small differences in absolute χ among sites as environmental contrasts.

5. The uncertainty of the retrieved χ values is insufficiently characterized in the present study. Although previous work is cited for sensitivity and uncertainty analyses, the uncertainties associated with measured growth factors, κ conversion, κ-PDF construction, end-member assumptions, and differences among datasets are not quantitatively represented in the cross-site comparison. This is especially important for marine aerosol, where the authors acknowledge that the binary framework becomes more approximate.

> We thank the reviewer for identifying this important limitation. Because the archived datasets differ in their original products and do not report measurement and inversion uncertainties consistently, a common uncertainty model cannot be applied retrospectively across all eight sites. We therefore added sensitivity analyses in Section 3.2 to assess how choices made during data aggregation and retrieval affect χ. We compared retrievals from pooled daily κ-PDFs with daily averages of scan-level χ (Figures S1–S2) and varied the prescribed upper end member, the RH used for growth-factor-to-κ conversion, and the κ-PDF bin width (Figures S3–S6).

6. Several mechanistic interpretations go beyond what can be directly established from the HTDMA observations. Seasonal and size-dependent variations are attributed to nitrate formation, biomass burning, long-range transport, cloud processing, monsoon circulation, fresh anthropogenic emissions, and sea-spray contributions. These mechanisms are physically plausible, but they are not independently constrained by chemical composition, source apportionment, trajectory analysis, or other supporting observations in the present study. The D_α–D_γ analysis itself is also derived mathematically from the same κ-PDF and therefore provides a useful decomposition of χ rather than an independent constraint on atmospheric processes.

> We thank the reviewer for raising this concern. We agree that the D_α–D_γ analysis is derived from the same κ-PDFs and does not independently identify atmospheric processes. We revised Sections 3.3 and

4.1 to distinguish measured hygroscopicity patterns from proposed explanations. In Section 4.2, we added bulk-composition measurements (Figures S11–S13) and back-trajectory analyses (Figures 5–6 and S14–S18). These observations support the site-specific interpretations in Section 4.3, where we discuss possible contributions from aerosol composition, transport, and processing without assigning unique sources or mechanisms.

7. The interpretation of increasing χ with particle size primarily as evidence of stronger atmospheric aging appears somewhat oversimplified. Aging is one plausible explanation, but size-dependent source distributions, condensational growth, coagulation, cloud processing, and selective removal can also modify the mixing state of different particle sizes. Moreover, fixed particle diameters do not necessarily correspond to the same aerosol mode across all environments.

We thank the reviewer for this clarification. We agree that particle diameter alone does not establish atmospheric age. We revised Section 4.1 to interpret size-dependent χ in terms of possible contributions from sources, condensation, coagulation, cloud processing, and selective removal, rather than attributing higher χ at larger diameters primarily to aging. The ENA discussion now relates its nonmonotonic size dependence to possible cloud processing, while the SGP and EPC discussion recognizes that larger particles can retain substantial hygroscopicity heterogeneity.

We also added seasonal particle number-size distributions (Figure S9) to assess where the selected HTDMA diameters fall relative to the aerosol modes at sites with available measurements. We distinguish differences between modes from variation within a mode and use the measured diameters, rather than fixed Aitken- and accumulation-mode labels, in Section 4.3 and the Figure 7 caption.

We made the following changes:

- P11L295–P12L301: "Within the ENA record, particles in the 100–200 nm range have higher median χ and narrower interquartile ranges (IQRs) than both 50- and 250-nm particles. This pattern may partly reflect cloud processing, as repeated cloud activation and aqueous-phase growth can reduce particle-to-particle differences in hygroscopicity (Gong et al., 2023; Wang et al., 2021). The bimodal ENA particle number-size distributions measured during 2024–2026 (Figure S9) are consistent with cloud processing at the site, but do not overlap the HTDMA observation period. The lower χ at 50 and 250 nm indicates greater particle-to-particle variability in hygroscopicity at those diameters."
- P12L318–326: "Figure S9 places the selected HTDMA diameters within the seasonal particle number-size distributions. Across the sites with available data, 50 nm generally falls within the Aitken or transition size range, diameters of 150 nm and above generally fall within the accumulation-mode range, and diameters near 75–110 nm often lie between these ranges. The observed size dependence of χ may therefore include differences between Aitken- and accumulation-size particle populations as well as changes within a mode. The higher χ observed toward the intermediate or accumulation-mode sizes at ENA and MHD is consistent with condensation, coagulation, or cloud processing reducing hygroscopicity differences among particles. Conversely, the decrease in χ with diameter at SGP and EPC indicates that accumulation-size particles can retain substantial hygroscopicity heterogeneity, potentially because of multiple source contributions, atmospheric processing, or selective removal."
- Figure 7 caption: "[...] Panels (a)–(c) show results for the smaller particles (50 nm) and panels (d)–(f) show results for the larger particles (250 nm at SGP and EPC and 260 nm at HACPL). [...]"
- P23L485–498: "At the six non-marine sites, most χ values fall between 50% and 100%, with size-dependent and seasonal patterns differing among sites. These patterns likely reflect the combined influence of size-dependent sources and atmospheric processing."

8. The implications for CCN and aerosol optical properties are stronger than directly demonstrated by the analysis. No CCN activation calculation is performed using the observed distributions, and the effect of mixing state on CCN depends on supersaturation, number-size distribution, and the full hygroscopicity distribution. Similarly, a high hygroscopicity-derived χ for accumulation-mode particles does not necessarily imply that internal-mixing assumptions introduce only small errors in aerosol optical calculations, particularly for absorbing aerosols such as black carbon, where coating state, refractive index, and particle morphology can strongly affect absorption.

We thank the reviewer for identifying this overinterpretation. We revised Section 4.1 to discuss how hygroscopicity differences among particles may affect CCN activation without implying a quantified effect on CCN concentrations. We also removed the statement concerning errors in aerosol optical calculations from the Conclusions.

We have made the following change:

- P12L327–330: "The observed size dependence of χ may be relevant to CCN activation because particle-to-particle differences in hygroscopicity produce differences in critical supersaturation. Aerosol mixing state can therefore influence the fraction of particles that activate at a given size and supersaturation. This influence is most relevant near the activation threshold, where modest differences in hygroscopicity can determine whether individual particles activate (Gasparini et al., 2006; Patel and Jiang, 2021)."

9. The manuscript presents the derived χ distributions as potential benchmarks for aerosol models, but the observational and model quantities are not necessarily equivalent. Composition-resolved models generally calculate mixing state from species mass fractions, whereas χ in this study is inferred from hygroscopicity under a binary LH–MH assumption. Therefore, the extent to which the reported χ can serve as a quantitative benchmark for model-derived composition-based mixing-state metrics requires careful interpretation.

We thank the reviewer for the suggestion. Our retrieval estimates χ for two effective less- and more-hygroscopic (LH–MH) components inferred from the measured κ-PDF. A model can calculate a corresponding hygroscopicity-based index directly from its composition by grouping species on the same LH–MH basis and applying consistent component fractions (Zheng et al., 2021). This applies to particle-resolved, modal, or sectional models where the represented particle populations retain sufficient composition differences. A model χ based on all chemical species separately measures a different aspect of mixing state and is not directly comparable with the values reported here. We have therefore clarified that these observations provide constraints for comparisons using a consistent hygroscopicity-based definition, rather than direct benchmarks for composition-based mixing-state indices.

We made the following changes:

- P3L69–71: "The resulting χ distributions and diversity diagnostics provide observational constraints for evaluating model representations of hygroscopicity-based mixing state."
- P24L520–531: "The results provide observational constraints on hygroscopicity-derived mixing state across diverse environments and particle sizes. They can support quantitative model evaluation when modeled species are grouped on the same LH–MH hygroscopicity basis and χ is calculated for comparable particle-size populations."

Minor comments:

1. The Abstract contains an incomplete phrase: "with attention to retrieval limitations and site-dependent."

We rewrote the Abstract and removed the incomplete phrase.

2. The statement that HTDMA instruments "operate continuously" appears too general considering that several datasets used here are campaign-based or contain incomplete seasonal coverage.

We revised the sentence to distinguish long-term monitoring from shorter campaigns:

- P3L55–58: "HTDMA instruments can be deployed for long-term monitoring or shorter field campaigns, providing size-resolved hygroscopicity observations across diverse environments."

3. The description of $\kappa_{LH} = 0$ as a nearly non-hygroscopic "carbonaceous" end-member is somewhat simplified because carbonaceous aerosol can span a substantial range of hygroscopicity depending on composition and atmospheric aging.

We removed the identification of $\kappa_{LH} = 0$ as a carbonaceous end member. Section 2 now describes it as the non-hygroscopic limit and clarifies that the prescribed end members are effective retrieval bounds.

- P4L88–91: "For the continental, urban, coastal-urban, and mountainous sites, we follow Liu et al. (2026) and prescribe $\kappa_{LH} = 0$ as the non-hygroscopic limit and $\kappa_{MH} = 0.65$ as representative of highly hygroscopic inorganic salts, including sulfate- and nitrate-containing salts."
- P4L96–97: "They are effective retrieval bounds rather than species-specific κ values."

4. The term "spatial variability" may overstate the spatial representativeness of only eight geographically sparse sites. "Cross-site variability" may better reflect the observational evidence.

We use "cross-site variability" rather than "spatial variability" for comparisons among the eight sites:

- P3L63–67: "We apply the method of Yuan and Zhao (2023) to HTDMA datasets from eight sites spanning urban, continental, coastal, marine, and mountainous environments and examine cross-site, size-dependent, and seasonal variability."
- Section 3.3 title: "Aerosol hygroscopicity across sites and seasons"

5. The manuscript occasionally uses "interannual variability" when discussing sites with approximately one year of observations. Such terminology is not appropriate where multiple years are unavailable.

We reserve "year-to-year" for records spanning multiple years and describe shorter records within their available periods:

- P11L270–272: "At the rural CRG site, median χ is high across the measured diameters during the March–July 2025 record. The decrease in median χ with diameter is more pronounced in spring than in summer, consistent with the greater separation among size-resolved κ-PDFs in spring (Figure S8)."
- P20L429–432, P21L446–448: "At the continental SGP site, seasonal values for 50 nm particles remain between the $\chi = 75\%$ and 100% isolines across the measurement years, despite substantial variability in D_α and D_γ (Figure 7a). [...] The dispersion among same-season values shows additional year-to-year variability, particularly for 50 nm particles."

6. Several statements use words such as "significantly" or "significant" even though no formal statistical significance test is presented.

We removed wording that implies a formal significance test and describe the observed differences directly:

- P8L201–205: "In winter and spring, the 50-nm distributions peak at lower κ than the distributions for larger particles, whose peaks shift toward $\kappa \approx 0.3$–0.4."
- The "less significant" optical-property claim was removed.

7. Narrow κ-PDFs are occasionally interpreted as evidence of chemically homogeneous aerosol populations. Chemically different particles may have similar κ values, so hygroscopic homogeneity does not necessarily imply chemical homogeneity.

We now interpret narrow κ-PDFs in terms of hygroscopicity, without inferring chemical homogeneity:

- P3L78–82: "We infer χ from the HTDMA-measured κ-PDF using the framework of Yuan and Zhao (2023), in which each particle is represented as a binary mixture of effective less-hygroscopic (LH) and more-hygroscopic (MH) components. The retrieved χ therefore quantifies particle-to-particle mixing with respect to hygroscopicity. It does not uniquely determine chemical mixing state because particles with different compositions can have similar κ values."

- P10L233–237: "At the mountainous HACPL site, the κ-PDFs are narrow and predominantly unimodal, with peaks near $\kappa \approx 0.15$–0.20 across the measured sizes and seasons, indicating comparatively limited particle-to-particle variability in hygroscopicity."
- P11L280–281: "High χ indicates similarity in particle hygroscopicity, but particles with different chemical compositions can have overlapping κ values."

8. Similarly, multimodal κ-PDFs do not uniquely identify distinct source populations. Different modes are consistent with multiple particle types, but hygroscopicity alone does not uniquely identify their sources.

We describe the measured hygroscopicity modes without assigning them to unique sources:

- P10L231–242: "The seasonal κ-PDFs for CRG, EPC, HACPL, BJIAP, and MHD are shown in the Supplement (Figure S8). The rural CRG and coastal urban EPC sites show similar features to HOU, with a dominant low-κ contribution for small particles and broader distributions for larger particles across the available seasons. At the mountainous HACPL site, the κ-PDFs are narrow and predominantly unimodal, with peaks near $\kappa \approx 0.15$–0.20 across the measured sizes and seasons, indicating comparatively limited particle-to-particle variability in hygroscopicity. The urban BJIAP site shows broader, often multimodal distributions and stronger separation among sizes, and its winter distributions resemble the patterns observed at SGP. The coastal MHD site stands out for its broad, distinctly multimodal distributions, with peaks near $\kappa \approx 0.3$ and 0.9. The high-κ mode may include sea-salt-rich marine particles, which may contribute to the higher hygroscopicity reported for clean marine than polluted continental air masses at MHD (Xu et al., 2020, 2021b)."

9. The number of observations represented by each boxplot in Figure 3 is not provided. Given the strongly unequal sampling among sites and seasons, this information is important for interpreting the distributions.

We added Table S2 to report the number of daily observations in each seasonal boxplot:

- Figure 3 caption: "Seasonal variability of the hygroscopicity-derived aerosol mixing state index χ at eight sites. Columns denote winter (DJF), spring (MAM), summer (JJA), and autumn (SON), except at BJIAP, where winter and summer denote the original authors' campaign designations; rows denote sites, and boxplots within each site are ordered by dry particle diameter. Boxes show the interquartile range (25th–75th percentiles), central lines indicate medians, whiskers extend to 1.5 times the interquartile range, and points denote values beyond the whiskers. ENA includes only observations retained after application of the receptor-local contamination filter. The numbers of daily observations are reported in Table S2, and the corresponding medians and interquartile ranges are reported in Table S3."

10. The manuscript contains several typographical and grammatical issues, including "observational constratins," "drwan from the literature," "variablity," "compare to," "aeorsol," and "particles populations," and would benefit from careful language editing.

We corrected the listed errors and edited the affected sentences. Representative revised wording includes:

- P2L26–28: "At one extreme, a fully internally mixed population has all particles sharing the same composition as the bulk; at the other extreme, a fully externally mixed population consists of particles that each contain a single pure chemical species."
- P2L41–43: "Bridging the gap between model assumptions and atmospheric reality therefore requires cross-site, size-resolved observational constraints that characterize variability across environments and sampling periods."
- P4L112–P5L115: The sentence was rewritten as "[...] and three were obtained from published studies and their associated data products (Xu et al., 2020, 2021a,b; Ray et al., 2023; Fan et al., 2020)."

- P10L245: "Figure 3 shows the seasonal variability of estimated χ across the available sites and particle sizes."
- P11L283–286: "At the urban BJIAP site, median χ is lower in the winter campaign compared to the summer campaign. It decreases with particle diameter in winter, whereas in summer it increases to a maximum near 110 nm before decreasing at larger diameters."
- P12L298–300: The sentence was rewritten as "The bimodal ENA particle number-size distributions measured during 2024–2026 (Figure S9) are consistent with cloud processing at the site, but do not overlap the HTDMA observation period."
- P10L239–242: The sentence was rewritten as "The coastal MHD site stands out for its broad, distinctly multimodal distributions, with peaks near $\kappa \approx 0.3$ and 0.9. The high-κ mode may include sea-salt-rich marine particles, which may contribute to the higher hygroscopicity reported for clean marine than polluted continental air masses at MHD (Xu et al., 2020, 2021b)."

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