Responses to Reviewer #3 Comments

This manuscript presents a multi-site analysis of aerosol mixing state inferred from size-resolved HTDMA measurements. The authors combine observations from eight sites representing urban, continental, coastal, marine, and mountainous environments and examine variations with particle size and season. Long-term observations of aerosol mixing state are limited, and I see value in bringing these datasets together within a common framework. The size-resolved comparison is interesting, and the manuscript is generally well organized. I have some minor comments to help improve the manuscript.

> We thank the reviewer for the positive assessment and helpful comments. We have carefully considered each point and revised the manuscript accordingly. 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.

General comments:

1. Because the main objective of this paper is to compare mixing state across different sites, a more comprehensive summary table for each site will be beneficial. The current Table 1 provides the site type, general measurement period, data source, and site type, but this is not enough to evaluate whether the datasets are directly comparable. I suggest expanding Table 1, or adding a new table, that summarizes for each site: coordinates and elevation, exact measurement dates, seasons represented, instrument or original data product, average measurement RH, particle sizes, κ-PDF or inversion method, κ_{LH} and κ_{MH} and the number of valid observations or sampling days. It would also be helpful to include a short summary of the main result for each site, such as the overall median or range of χ and whether χ generally increases or decreases with particle size. More detailed seasonal statistics could be placed in a supplementary table.

> We thank the reviewer for this helpful suggestion. We added Table S1 to document measurement conditions, original data products, and dataset-specific processing. Tables S2–S3 report the numbers of valid daily observations and the seasonal medians and interquartile ranges of daily χ. The measured particle diameters were already listed in Table 1, and the size-dependent patterns are discussed in Section 4.1.

2. Major quality-control procedures and pollution filtering procedures are not very clear. For example, it is not clear to me how to perform quality control in line 108 or what a 6-minute pollution flag is.

> We thank the reviewer for pointing this out. We expanded Section 3.2 to describe how the ENA local-pollution flag was defined, aggregated from 1-minute to 6-minute intervals, and matched to HTDMA observations. We also specified the validity criteria applied before constructing daily κ-PDFs.
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> We made the following change:
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> - P6L148–P7L164: "For ENA, we applied an additional quality-control procedure to remove HTDMA observations affected by local anthropogenic pollution near the measurement facility. We used the 1-minute local-pollution flag developed by Zhou et al. (2025), which identifies periods with elevated and strongly fluctuating number concentrations of particles larger than 10 nm while retaining contributions from sea spray generated by shoreline wave breaking. To align the flags with HTDMA observation timestamps, we grouped consecutive 1-minute flags into 6-minute intervals and classified an interval as polluted if any of its six minutes was flagged. Each HTDMA observation was matched in UTC to the nearest 6-minute flag interval and retained only if the time difference was no more than 3 minutes and the interval was unflagged. The quality-control procedure removed approximately 35% of the ENA HTDMA observations, with the highest rejection rates occurring under winds from the island interior and during periods of greater daytime local activity."

3. It might be good to add more details in the data processing section. For example, in lines 101-103, it would be good to provide the conversion, at least in SI units, although it is a well-known method. It is not clear to me why kappa is constructed between 0 and 0.65 in line 104.

We thank the reviewer for this suggestion. We expanded Section 2 to explain why the prescribed end members are 0 and 0.65 for the six non-marine sites, and why a higher upper end member is used for ENA and MHD. These choices account for the measured κ-PDFs and reported hygroscopicities of common aerosol components. We also added the growth-factor-to-κ conversion equation in Section 3.2 and defined its inputs.

We made the following changes:

- P4L88–102: "For the continental, urban, coastal-urban, and mountainous sites, we follow Liu et al. (2026) and prescribe $\kappa_{\mathrm{LH}} = 0$ as the non-hygroscopic limit and $\kappa_{\mathrm{MH}} = 0.65$ as representative of highly hygroscopic inorganic salts, including sulfate- and nitrate-containing salts. For the marine-influenced sites ENA (Eastern North Atlantic) and MHD (Mace Head, Ireland), we use $\kappa_{\mathrm{MH}} = 1.5$. The baseline end members were selected to encompass the resolved structures of the measured κ-PDFs without truncating their upper modes, while remaining consistent with reported hygroscopicity ranges for common aerosol components (Petters and Kreidenweis, 2007; Carrico et al., 2008; Zieger et al., 2017; Lewis, 2019; Chen et al., 2022). They are effective retrieval bounds rather than species-specific κ values. The broader interval used at ENA and MHD makes the binary representation more approximate because common inorganic particles can be represented as LH–MH mixtures rather than pure MH material. We therefore use the ENA and MHD retrievals to examine temporal and size-dependent variations at these sites and do not compare their χ values quantitatively with values retrieved using $\kappa_{\mathrm{MH}} = 0.65$."
- P6L143–147: "For a dry diameter D_{d} and measured growth factor GF, we calculated

$$\kappa = (GF^3 - 1)\left[\frac{\exp\left(\frac{4\sigma_{\mathrm{w}} M_{\mathrm{w}}}{RT\rho_{\mathrm{w}} D_{\mathrm{d}} GF}\right)}{RH} - 1\right].$$

Here, GF is the ratio of humidified to dry particle diameter, and RH is relative humidity expressed as a fraction. D_{d} [m] is the dry particle diameter, σ_{w} [N m^{-1}] is the surface tension of water, and M_{w} [kg mol^{-1}] is the molar mass of water. R [J mol^{-1} K^{-1}] is the universal gas constant, T [K] is absolute temperature, and ρ_{w} [kg m^{-3}] is the density of water."

4. Many of the atmospheric explanations offered in Sections 3.3 and 4 are physically plausible, but they are not directly established by the measurements presented in this paper. Examples include nitrate formation and biomass burning at SGP, fresh emissions and rapid processing at the urban sites, continental transport from Los Angeles at EPC, cloud processing at ENA, sea-spray contributions at MHD, and monsoon-driven transport at HACPL. A κ-PDF alone cannot uniquely identify these sources or processes. The D_α-D_γ analysis does not provide an independent source constraint because it is calculated from the same κ-PDF. Where aerosol composition, trace-gas measurements, wind direction, cloud occurrence, or trajectory information are available, these data could be used to strengthen the interpretations. This seems particularly feasible for the ARM sites. If comparable supporting information is not available for all sites, the discussion should acknowledge this limitation rather than presenting the proposed mechanisms as established conclusions.

We thank the reviewer for this comment. In Sections 3.3 and 4.1, we distinguish the measured hygroscopicity patterns from proposed explanations, including fresh emissions, cloud processing, and sea-salt contributions. In Section 4.2, we add available bulk-composition measurements and 72 h back trajectories to provide independent context for seasonal changes in aerosol composition and transport. We use these observations to inform the D_α–D_γ discussion in Section 4.3. We also acknowledge that bulk composition is not particle resolved and that trajectories cannot uniquely identify sources or processing mechanisms.

Minor comments:

1. Describing $\kappa_{LH} = 0$ as a non-hygroscopic "carbonaceous" end member is an oversimplification. Carbonaceous aerosol can span a wide range of hygroscopicities depending on its composition and atmospheric processing.

We removed the identification of $\kappa_{LH} = 0$ as a carbonaceous end member and clarified that the prescribed values 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."

2. Please use either fractional χ from 0 to 1 or percentage χ from 0% to 100% consistently in the text and figures.

We now consistently report χ as a percentage in the text, figures, and Equation 3:

- P3L76–78: "The aerosol mixing state index χ ranges from 0% for the external-mixing limit to 100% for the internal-mixing limit (Riemer and West, 2013)."
- Figure 7 caption: "Dashed lines represent mixing state index values of χ = 25%, 50%, 75%, and 100%."

3. Words such as "significantly" should be avoided unless a statistical significance test has been performed. If the authors want to make general statements about size or seasonal differences, some statistical evaluation would be helpful.

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.

4. "Spatial variability" may overstate the spatial representativeness of eight geographically sparse sites. "Cross-site variability" would be more accurate.

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 caption of Figure 4 states "Panels (a)–(c) shows"; this should be "show." The caption should also clarify whether the points are means or medians.

We corrected the verb and specified that the points are medians in Figure 7 caption:

- "Per-particle diversity (D_α) and bulk population diversity (D_γ) for Southern Great Plains, OK (SGP, continental), La Jolla, CA (EPC, coastal urban), and High Altitude Cloud Physics Laboratory, India (HACPL, mountainous). Each point represents the median for a given season and year and is included only when at least 10 daily observations are available. Horizontal and vertical bars denote one-half of the interquartile range in each direction. Dashed lines represent mixing state index values of χ = 25%, 50%, 75%, and 100%. 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). Colors denote seasons as indicated in the legend."

6. The bibliography appears to contain duplicate entries for Parworth et al. (2015), Niedek et al. (2025), and Zhao et al. (2021).

We thank the reviewer for catching this oversight. We have removed the duplicate entries from the bibliography.

7. The manuscript contains a number of typographical and grammatical errors, including "all particle," "observational constratins," "drwan from the literature," "variablity," "compare to," "aeorsol," and "particles populations." Careful proofreading is needed.

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

- P2L26–27: "At one extreme, a fully internally mixed population has all particles sharing the same composition as the bulk [...]"
- 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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