Reviewer #1,

Comment: This manuscript by Yuan et al. reports a new algorithm to quantifying the heterogeneity in aerosol hygroscopicity, by using the data from H-TDMA. Average per-particle species diversity Dα, the bulk population species diversity Dγ, and their affine ratio χ was calculated as three indices to describe aerosol heterogeneity. These data are valuable and the figures presented in the paper are good description of the research. Therefore, I would suggest publication after the following comments have been addressed.

Reply**:** Thanks for the comments and suggestions. The point-by-point responses are listed below.

Comment: Specific comments:

(1) Line 28-29: "Considering that ambient aerosol particles in an aerosol population differ dramatically in chemical composition due to the complex sources and aging processes." The authors might find the following paper relevant findings from individual particle analysis by electron microscope and models. For example, recent studies focused on different kinds of mixing states such as Journal of Geophysical Research: Atmospheres 2022, 127, (5), e2021JD036055; Atmos. Chem. Phys.2021, 21, (23), 17727-17741; Environmental Science & Technology2021, 55, (24), 16339-16346.

Reply**:** Thanks for the comment. We have added these references. These literatures are crucial for perfecting this paper. They help to deepening and highlighting the significance of this study. (Lines 29-30, Page 2 in the revised manuscript)

(2) Line 43: may change from "black carbon (BC)-containing" to "black carbon-containing (BC-containing)"

Reply**:** Thanks for the comment. We have changed "black carbon (BC)-containing" to "black carbon-containing (BC-containing)". (Lines 43-44, Page 2 in the revised manuscript)

(3) Line 48: "most literature is" should use plural form

Reply**:** Thanks for the comment. We have replaced "most literature is" with "most studies are". (Line 48, Page 2 in the revised manuscript)

(4) Line 50: “we propose” -> “we proposed”

***Reply*:** Thanks for the comment. We have replaced “We propose” with “We proposed”. (Line 50, Page 2 in the revised manuscript)

(5) Line 59: “propose” should be “proposed”

***Reply*:** Thanks for the comment. This comment seems to be the same as comment 4. We have revised this sentence. (Line 50, Page 2 in the revised manuscript)

(6) Line 52: “will describe” should be described

***Reply*:** Thanks for the comment. We have replaced “will describe” with “described”. (Line 52, Page 2 in the revised manuscript)

(7) Line 53: “will be” should be “was”

***Reply*:** Thanks for the comment. We have replaced “will be” with “was”. (Line 53, Page 2 in the revised manuscript)

(8) Line 54: “are discussed” should be “were discussed”

***Reply*:** Thanks for the comment. We have replaced “are discussed” with “were discussed”. (Line 54, Page 2 in the revised manuscript)

(9) Line 55: “comes” should be “came”

***Reply*:** Thanks for the comment. We have changed “comes” to “came”. (Line 54, Page 2 in the revised manuscript)

(10) Line 81: why the font of “$TDMA_{fit}$” is different?

***Reply*:** Sorry for the mistake caused by the text editor. This is revised and the other font issues are also revised. (Line 81, Page 3 in the revised manuscript)

(11) Line 139: “ageing” is different with other “aging” in your article, here should be unified

***Reply*:** Thanks for the comment. We have changed “ageing” to “aging” in this line and we revised the whole text.

(12) Line 154: “ranges 1 to 2” should be added “from”

***Reply*:** Thanks for the comment. This sentence has been revised. (Line 160, Page 7 in the revised manuscript)

(13) Line 155-156: “it is 1 when … while 2 when …”, the grammar here is strange. May add “the” or “be” behind “while”

***Reply*:** Thanks for the comment. This sentence has been revised. (Line 162, Page 7 in the revised manuscript)

(14) Figure 2: Does blue represent the LH or grey represent the LH? Need explanations here.

***Reply*:** Thanks for the comment. The blue ball in Figure 2 represents the MH component and the grey ball represents the LH component. We have added the legend in Figure 2 to clearly show the LH and MH components. (Figure 2 in the revised manuscript)

(15) Line 186: why chose 110 nm aerosol? Need reasons here.

***Reply*:** Thanks for the comment. Here we want to focus primarily on the variation of the heterogeneity in aerosol hygroscopicity in the real atmosphere, especially for the condition that the total hygroscopicity of aerosol remained constant, to highlight the importance of the heterogeneity as an indicator for the evolution of aerosols.

From the time series of the heterogeneity for aerosols of the five measured sizes (40 nm,80 nm, 110 nm, 150 nm, and 200 nm) during the selected episodes (Figure R1), we find the similar variation trends for aerosols larger than 100 nm. Difference is showed for the aerosol of 40 nm and 80 nm mainly due to their different formation mechanism, which will be further investigated in our following studies.

110 nm aerosol is chosen here by considering that 1) aerosol around 100 nm generally has the largest number concentration. Among the five measured sizes, the diameter of 110 nm aerosol is the closest to 100 nm. The heterogeneity in aerosol hygroscopicity of this diameter may have great

impact on the direct radiative forcing, especially for the condition with high relative humidity. 2) aerosol smaller than 100 mainly shows the gas to particle process in the atmosphere, while aerosol larger than 100 mainly reflects atmospheric aging process. The 110 nm aerosol lies in the transition zone between these two processes, the variation of the heterogeneity in 110 nm aerosol can better reflect the evolution of aerosol particles in the real atmosphere. Thus, we choose 110 nm aerosol as the example to show the variation characteristic.

We have added the reason in the revised manuscript. (Lines 187-195, Page 8 in the revised manuscript)

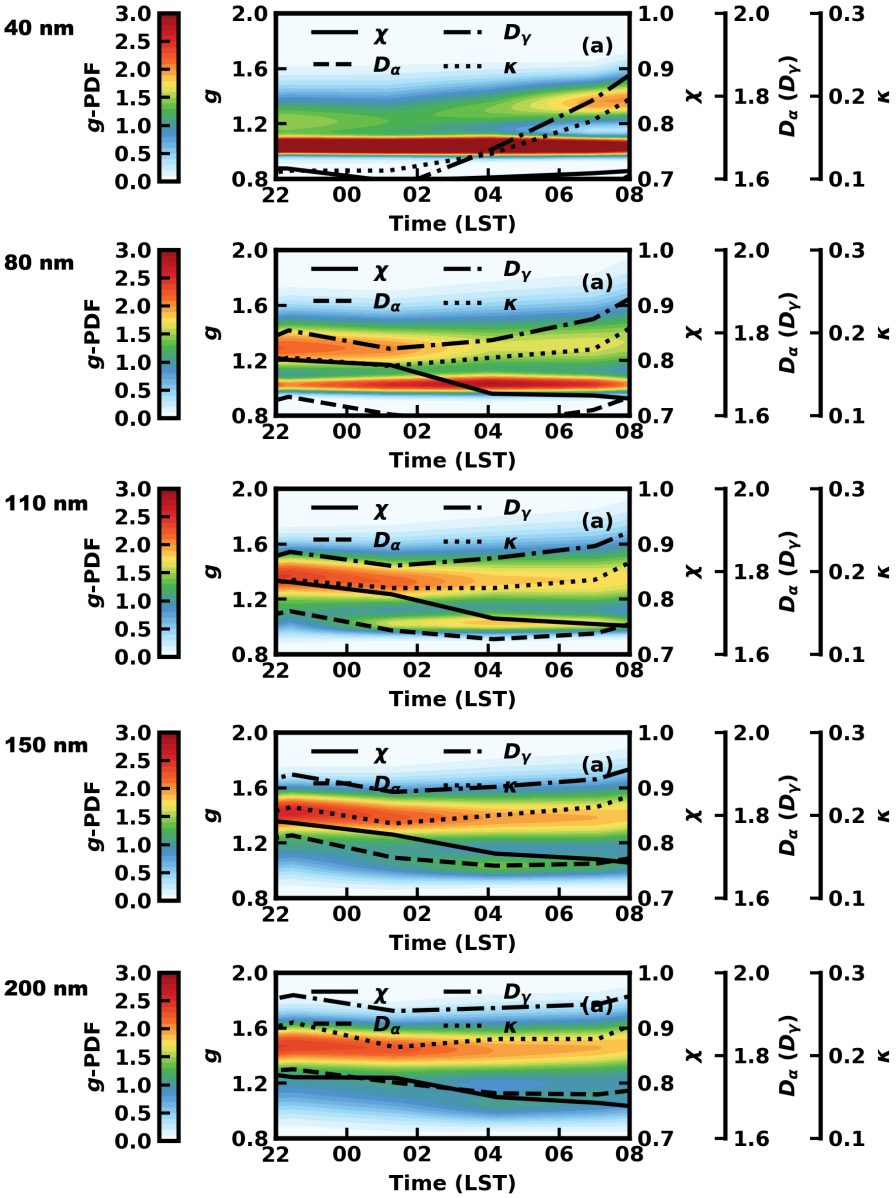

Figure R1. The variation of κ-PDF, D_α, D_γ, and χ during two typical evolution processes for aerosols of five measured diameters in the winter observation period.

(16) Line 190: why is χ high at night? Here needs more explanations.

***Reply*:** Thanks for the comment. Here we wanted to focus on the variation of the heterogeneity in aerosol hygroscopicity in the real atmosphere, especially for the condition that the total hygroscopicity of aerosol remained constant. Thus, just two episodes with tiny changes of κ_{mean} during the observation periods are chosen. These two episodes were both from the night to the morning on the next day and both χ showed high values at night with decreasing trend from the night to the morning.

To clearly explain this phenomenon, the diurnal variation of χ and aerosol particle number size distribution (PNSD) during the winter observation period is shown in Figure R2a. It can be found that χ had a relatively lower value than that in the afternoon at the traffic rush hour around 17:00-20:00 LST, which made the heterogeneous distribution of the LH and MH components within and between aerosol particles. Then χ gradually increased after the rush hour and had a relative higher value at night due to intensive mixing in the various atmospheric aging processes with less emissions, which led to the homogenous distribution of the LH and MH components in aerosols. This process was confirmed by the PNSD (Figure R1b) that the total number concentration decreased while the median diameter increased after the rush hour. The details in the variation characteristics will be further discussed in our following studies.

We revised Lines 196-200 to make the expression clearer. (Lines 196-200, Page 8 in the revised manuscript)

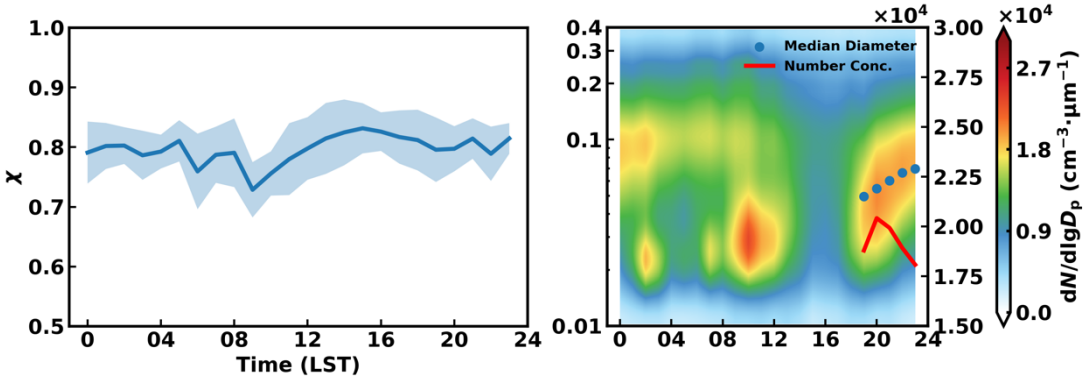

Figure R2. Diurnal variation of χ and PNSD during the winter observation period.

(17) Line 217: remove “will”

***Reply*:** Thanks for the comment. We have changed “will discuss” to “discussed”. (Line 230, Page 9 in the revised manuscript)

(18) Figure 6 (a) and (d) need a legend

***Reply*:** Thanks for the comment. The solid lines (blue) in (a) and (d) are the measured PNSD that can be fitted by a four-mode (a nucleation mode (NM), an Aitken mode (AM), an accumulation mode (AcM) and a coarse mode (CM)) lognormal distribution, which are represented by the thin solid line (orange), the dashed line (green), the dotted line (red), and the dash-dotted line (purple), respectively. We have added the legend in Figure 6 to clearly show the PNSD. (Figure 6 in the revised manuscript)

(19) Line 230: “Aitken mode” Why capitalize here?

***Reply*:** Thanks for the comment. “Aitken mode” is named after John Aitken, who is one of the founders of cloud physics and aerosol science. Thus, we capitalize “Aitken”.