Response to community' comments on "Characterization of fog microphysics and their relationships with visibility at a mountain site in China"

General comment:

Thank you for your interesting research manuscript! We discussed your work within our research group since we are doing similar research and got interested in your findings. We have some remarks and questions concerning the experimental set-up. Many technical details (e.g. on the sampling efficiency of the GCVI inlet) are currently missing and should be added to allow a reliable assessment of the presented results. Moreover, the reasoning behind many of the key findings are often not clear to the reader and some more clarifications (incl. add the right references) would clearly help here.

[*Response*] Thanks you very much for your interests in our work and the positive comments and suggestions. We have revised the manuscript according to the comments point by point.

Below, we have listed a few questions and remarks.

1. Our most important comment is the lack of describing the sampling efficiency of the GCVI system. Has it been determined? How well do you sample larger droplets? Have zero-measurements been performed? This is an important task and will have a substantial impact on most of the results and interpretation presented here. (see e.g. Figure S4 and others in Karlsson et al, 2021). At the moment, it is not clear if any particle loss corrections (for the aerosol instrumentation behind the inlets and for the fog monitor) have been done.

[*Response*] Thanks for your suggestion. Before the observation, we operated the GCVI inlet system on a clear day for zero-measurements. The CPC installed downstream of the GCVI system and measured a concentration of 0 during this test. We admit the correction of sampling efficiency of GCVI system is quite important for the quantitation of cloud residual particles. This study mainly focused on the effects of aerosols on fog microphysical characteristics. For the measurements downstream of the GCVI inlet, only the activation ratio of cloud residual particles was shown in Fig. 7 and Fig. S10 to exhibit the scavenging of un-activated aerosol particle by large fog droplets. This activation ratio, defined as the CCN number concentration to the total particle concentration, is almost not influenced by the sampling efficiency in GCVI systems. As it reported in Karlsson et el. (2021), the correction of sampling efficiency plays an important role in comparing the GCVI measurements to other instrumentation methods, such as aerosol particle measurements from the whole-air inlet and fog droplets size distribution measured by fog monitor. Ongoing work will address these descriptions as well as a comparison of the physicochemical properties of cloud residual particles and cloud interstitial particles.

2. A schematic of the set-up which includes instrument names, inlets, piping, flow rates (or a reference to it) would be very useful to the reader.

[*Response*] Thanks for your suggestion. We have added a figure showing the instruments installation in the supplement as below:

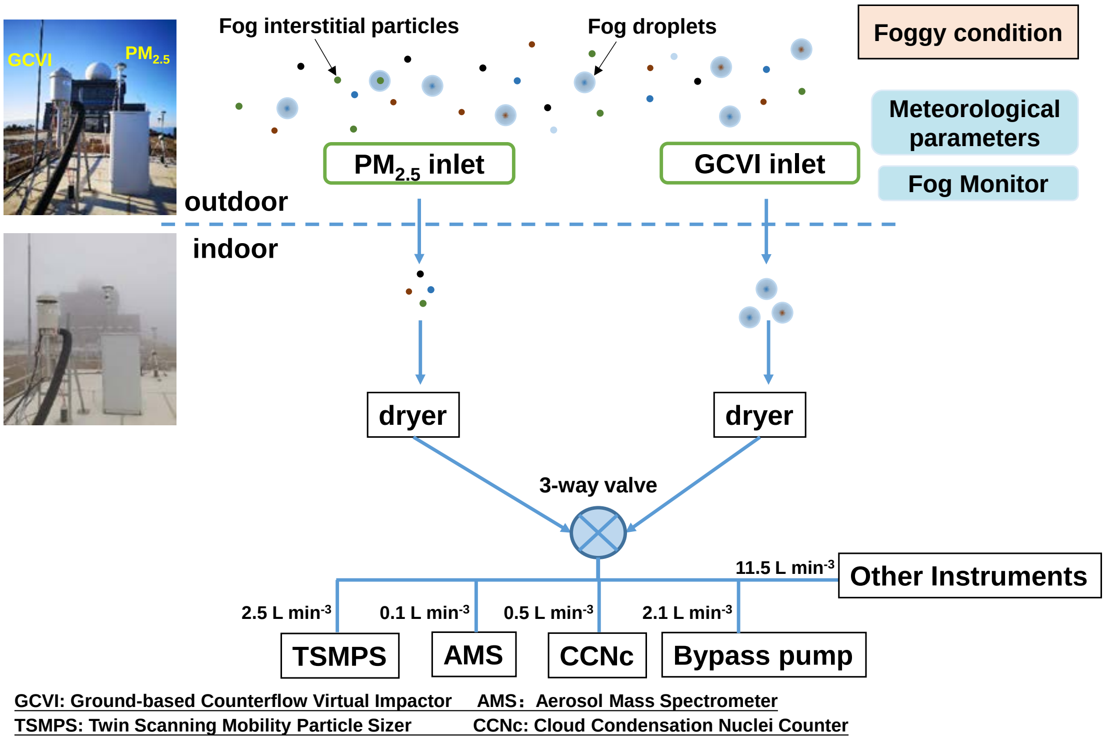

Fig. S2. Schematic of the experimental setup at the Daming Mountain site. An automatic three-way switching system was placed between the sample inlets and instruments. Meteorological parameters and fog droplets were simultaneously measured on the roof of the observation container. The bypass pump only operated when the three-way valve connected to the $PM_{2.5}$ inlet. Its flow rate was controlled at 4.5 L min^{-1} via a mass flow controller, ensuring the total sample flow reached the 16.7 L min^{-1} required by the $PM_{2.5}$ cyclone inlet.

3. Line 23 and 240: How do you know that it was indeed collision-coalescence? Just because another peak in the size distribution appeared? Could it be that it is just condensational growth? Please elaborate.

[*Response*] Thanks for pointing this out. In the mature stage, N_d experienced a significant decrease due to a substantial reduction in small droplets, meanwhile, D_{eff} notably increased with an additional peak of the droplets size distribution appearing at 23 μm. Besides that, the activation ratio of fog residual particles significantly reduced during this stage, implying certain un-activated aerosol particles were scavenged by the uptake of larger fog droplets. Based on the evidences described above, we infer that the collision-coalescence process occurred at this stage.

4. One of the key findings is that secondary activation was observed after additional cooling. However, this is not really clear from the figures and key parameters like wind-direction and speed are not shown.

[*Response*] Thanks for your suggestion. The wind direction and speed have been added in Fig. 5 in the revised manuscript. We also added the descriptions on the secondary activation process as below:

"However, after reaching and maintaining a quasi-equilibrium supersaturation state (SS_{Q1}) in the early mature stage, a notable decrease in temperature occurred (Fig. 5a) without obvious changes in wind direction and speed (Fig. 5b). This decrease caused an increase in both excess water vapor pressure and supersaturation, as the temperature-dependent equilibrium vapor pressure dropped faster than the ambient partial vapor pressure. Consequently, a new quasi-equilibrium supersaturation state (SS_{Q2}) was established, exhibiting distinct fog microphysical characteristics (Fig. 6b). Compared to SS_{Q1}, the N_d substantially increased in the SS_{Q2} stage, while the *LWC* and D_{eff} notably decreased (Fig. 5b). The enhanced *SS* facilitated the further activation of smaller particles that were un-activated during the SS_{Q1} stage, resulting in a secondary activation-dominated process during the E3 (Fig. 5c and Fig. 6b)."

5. Introduction: Please refer to e.g. Elias et al. (2009) and Hammer et al. (2014) who also discussed the contribution of hydrated aerosol to light extinction.

[*Response*] Thanks for your suggestion. We add the relevant references in the revised manuscript. Please see Line 73.

6. Line 68: You mention that particle number size distributions were measured but the actual findings/curves (mean distributions and timelines) are never shown. However, it would be useful to add these graphs to the manuscript or SI to better interpret the findings.

[*Response*] Thanks for pointing this out. This study primarily focuses on the evolutionary characteristics of fog microphysical processes. The number size distributions of pre-fog aerosols, fog interstitial particles, and fog residual particles during this campaign are present in another study (Shen et al., 2024), which is also in preprint of ACP.

Shen, X., Liu, Q., Sun, J., Kong, W., Ma, Q., Qi, B., Han, L., Zhang, Y., Liang, L., Liu, L., Liu, S., Hu, X., Lu, J., Yu, A., Che, H., and Zhang, X.: Measurement report: The influence of particle number size distribution and hygroscopicity on the microphysical properties of cloud droplets at a mountain site, EGUsphere, 2024, 1-24, 10.5194/egusphere-2024-2850, 2024.

7. Line 83: Do you have supporting data showing that the site is usually near the top of the PBL?

[*Response*] Thanks for pointing this out. Due to the lack of measurement of the PBL height on this site, we have removed the relevant description from the revised manuscript.

8. Line 132: The CCN counters are usually kept longer at one fixed temperature in

order to achieve a stable supersaturation. Have you checked that 1 min is a long enough period? Especially, when switching from 0.7% down to 0.1% we doubt that this will be sufficient.

[*Response*] We agree your opinion. We had checked the CCN data. After altering the *SS* in the CCNc column, the CCN concentration can reach a stable state within 1 min. In our study, the four *SS* setpoints were sequentially scanned from low to high and then back from high to low to avoid large change of SS in the CCNc column. The relevant descriptions had been added in the revised manuscript. Please see Lines 139-140.

9. Line 149: Consider including the interstitial aerosol number concentration in Tab. 1. Have you performed a closure study to see if N_d and the number of fog residuals agree?

[*Response*] Thanks for your suggestion. In table 1, we give out the microphysical parameters of 8 fog events during the campaign. This manuscript focuses on the analysis of fog monitor data with some support from CCNc and TSMPS data. Ongoing work will address the comparison of fog monitor data and fog residuals data, such as measured N_d and estimated N_d, following the method of Karlsson et al. (2021), and will also derive N_d from fog interstitial particles or fog residual particles.

10. Line 176: What is the p-value if you talk about significance but only have a few data points? Have you also looked at the size distribution? Has that also changed in the different pre-fog N_{total} conditions? If you have so much more particles than droplets, why would you expect the N_{total} to be correlated with N_d and not just the N_{100} or even higher? How does the size distribution behind the CVI look like?

[*Response*] Thanks for pointing this out. Indeed, the size distribution of pre-fog aerosols varied in different fog events. We have performed a t-test for the correlation between pre-fog aerosols and the peak N_d. The *p*-values for both pre-fog N_{a_total} and N_{a_100} were less than 0.05, indicating a significant level of correlation for them. We have added the *p*-values in Fig. 3 and Fig. S5. As shown, the concentrations of particle diameter larger than 70 nm (N_{a_70}) or 100 nm (N_{a_100}) had a much stronger correlation with the peak N_d than that of total pre-fog N_a. The particles number size distribution behind the CVI during this campaign can be found in Shen et al. (2024).

11. Line 180 and Fig. 3: It is not really clear why certain points were excluded. Please explain and reason why the data points after rain events should be excluded. Do you then sample artifacts? In addition, in Fig. 3, please state which kind of linear regression has been applied. Since both x- and y-values are prone to errors, you should use an orthogonal regression, which does not seem to be used here.

[*Response*] Thanks for pointing this out. For the fog events occurred without precipitation, the N_d dramatically increased due to the activation of aerosol particles. This is also the main reason for the good positive correlation between the pre-fog N_a and the peak N_d. However, raindrops can significantly influence the N_d through collision-coalescence, resulting in no clear correlation between pre-fog N_a and peak N_d. When precipitation was detected by a rain/snow sensor, the GCVI inlet system automatically shut down, and the sampling flow from $PM_{2.5}$ pathway. Additionally, the linear regression used in Fig. 3 is based on the least squares fit, the p-values have been added in this figure.

12. Line 184: What does it indicate that your slope values are higher than those measured by Duplessis et al. (2021)? What are the consequences? It would help to elaborate more here.

[Response] Thanks for pointing this out. We have added the description to illustrate its implication as below:

"The slope value of 0.09 in this study is significantly higher than the 0.014 observed by Duplessis et al. (2021) on the eastern coast of Canada, indicating stronger bulk activity observed at this mountain site."

13. Line 214: Please also include the *SS* values from those four publications you are referring to.

[Response] We have added these *SS* values in the revised manuscript. Please see Lines 265-268.

14. Line 225: To us, the classification into fog stages seems to only have worked semi-well, especially in E3. How exactly did you divide the fog events and why did you choose this definition?

[Response] Thanks for pointing this out. The changes in visibility during fog events are closely related to the evolution of fog microphysical characteristics. The classification of fog stages can be based on changes in visibility (Mazoyer et al., 2022) or the ratio of *LWC* to N_d (Li et al., 2020). The classification depending on visibility was adopted not only in reference to previous studies (Mazoyer et al., 2022; Niu et al., 2010b; Pilie et al., 1975), but also to align with the subsequent discussions in this paper, regarding the relationship between fog microphysical parameters and visibility.

15. Line 228: What concentrations do you consider to be "high" or "low"? It would improve the interpretation to explicitly state the measured concentrations here.

[Response] Thanks for pointing this out. We have added the descriptions of aerosol backgrounds as below:

"Although there were few anthropogenic sources near the site, the observed aerosol concentrations varied dramatically. As shown in Fig. 1e, the N_a ranged from 230 to 15620 cm^{-3}, with a median of 2750 cm^{-3}. Episodes with N_a exceeding 8000 cm^{-3} were typically associated with a pronounced increase in aerosol number concentration within the size range of 10-100 nm (Fig. 1e), which were likely driven by new particle formation (Shen et al., 2022). In the subsequent discussion, the pre-fog aerosol concentration below and above this median were defined as low and high number concentrations of aerosol backgrounds, respectively."

16. Line 242ff: Why should the activation ratio of the residuals show that there was scavenging? Please explain in more detail as a correlation doesn't mean causality. What is the reasoning to define the AR via the CCNC measurement and not via the CPC/SMPS measurements?

[Response] The estimating water vapor supersaturation (*SS*) in fogs during this

campaign were generally lower than 0.2%. If fog residual particles enter droplet though an activation process, these particles should also be activated in the CCN counter (CCNc) column, where can set different *SS* conditions. The activation ratio (AR) was defined as the CCN concentration measured by CCNc to the aerosol concentration measured by SMPS. In this case, the concentrations measured by CCNc and TSMPS after GCVI inlet should be consistent, i.e., the AR should be ~1, especially for high *SS* setpoints. As shown in Fig. 7, the AR measured downstream of the GCVI airflow were closed to 1 when the D_{eff} of fog droplets smaller than 12 μm. However, when the D_{eff} exceeding 12 μm, the AR of fog residual particles notably decreased. The reduced AR of cloud residual particles was caused by the uptake of particles less prone to activation into droplets, implying the fog scavenging efficiency for these particles significantly enhanced in this stage. We add these descriptions and figures of the AR variations under different *SS* conditions in the revised manuscript. Please see Lines 303-308 and Fig. S10.

17. Line 259ff: The statement about the evaporation of large droplets due to the formation of smaller droplets is not really clear. Could you elaborate more here and also provide some more references for this effect?

[Response] Thanks for pointing this out. We are sorry that we did not express clearly in the original manuscript. Because both large and small droplets are affected by evaporation, but small droplets are more susceptible to dry air because of a larger surface area concentration. The description has been revised as below:

"During this secondary activation process, a greater number of small droplets formed and competed for the limited water vapor, which led to a decrease in the D_{eff} (Fig. 6b)."

18. Line 270: Why are you using 880nm? Is it because of the visibility sensor that comes with the GCVI? In that case, it should probably also be 3 and not 3.912 in eq. 5 because of how the visibility sensor is calibrated (see manual of the Belfort visibility sensor).

[Response] Thanks for pointing this out. The data from Belfort visibility sensor that comes with the GCVI were lacking in several time periods due to GCVI instrument failure. The visibility data used in this study was from a simultaneously measurement of the forward scattering visibility meter (Model DNQ1, Huayun Inc., China) at 880 nm. The numerator in Eq. 5 should be 3, which is in accordance with the method of visibility meter. We apologize for the incorrect value given in Eq.5 and have corrected it in the revised manuscript.

19. I would suggest moving the first part of chapter 3.5.1 to the methods section. This is not really results.

[Response] Thans for your suggestion. We have moved this part to the Method section.

20. Line 289: Were all data points included when performing the linear regression? It would be helpful to add the result to the figure. Is the slope similar if you only include values below e.g. 1km?

[Response] Yes, the linear regression in Fig. 8a included all data points. We add the fitting lines to the figure. When we select the data of $VIS_{DSD} \leqslant 1000$ m to perform linear regression, only the slope for VIS_{GN} is similar with that of all data points.

21. Line 290ff: Adding a new parameter (here N_d) gives more information and therefore improves the parametrization. Please clarify the last two sentences of this paragraph.

[Response] The visibility degradation contributed by fog droplets is determined by fog droplets size distribution. Meanwhile, the fog microphysical parameters of N_d, LWC, and D_{eff} are derived from the measurement of fog droplets size distribution (Equation 1-3). When both LWC and N_d values are given, the information of D_{eff} can also be determined to a large extent. Comparing to the LWC-only parameterization, the $LWC{\cdot}N_d$ parameterization can better represent the fog droplets size distribution, and therefore is

expected to be more accurate in fog visibility forecasts. We clarify it in revised manuscript. Please see Lines 338-343.

22. Line 300: Mie theory should be a good prediction for observed visibility. Make your explanation more detailed.

[Response] We have added the explanation in Line 330-333:

"Compared to the parameterization schemes of fog visibility, Mie theory incorporates a specific extinction algorithm based on physical processes. Therefore, the fog visibility derived from fog DSD and Mie theory is expected to better reflect actual conditions, which can serve as a reference for fog visibility parameterization."

23. Line 254: Please also mark these quasi-equilibrium states in the temporal evolution shown in Fig. 5.

[Response] We have marked the quasi-equilibrium states in the revised manuscript.

Figures:

Fig. 1: We would recommend you to choose different colorbars which have a more intuitive and uniform distribution of colors, e.g. 'Blues'. Having white in the middle of the colorspectra is very misleading. https://journals.ametsoc.org/view/journals/bams/96/2/bams-d-13-00155.1.xml

[Response] Thans for your suggestion. We have changed the colorbar.

Fig. 2: As you calculate LWC by using D and Nd, isn't the outcome of this figure trivial? Maybe move it to the supplement?

[Response] Thans for your suggestion. These three parameters are derived from the observed droplets size distribution and Equations 1-3. We have moved it to the supplement.

Fig. 4: please plot dN/dlogD as commonly used. The x-axis should probably be 'nm' not 'um'. Please write somewhere that this is E3 as you later on talk a lot about this

specific event.

[Response] Thanks for pointing this out. This is a schematic of method for deriving water vapor supersaturation (*SS*) in fog. We have revised the figure and move it to the Method section. The 'E3' has been added in the figure caption to specify the event.

Fig. 5: For better comparability, we would suggest to use the same colorbar and axis limits for all events (also Fig. 1 and S5) and use the same axis for Dp and Deff in subplots (c).

[Response] Thans for your suggestion. We have revised the colorbars and axis limits in these figures. The different axis used for D_p and D_{eff} can help to clearly exhibit their variations.

Fig. 6: Please explain in the figure caption what SS_{Q1} and SS_{Q2} means. Have you considered plotting one subplot where the 4 average size distributions are plotted on top of each other so that one can more easily see the differences? Why did you choose a linear scale for the diameter (x-axis)? Typos: 'Development', 'Dissipation'

[Response] Thans for your suggestions. We have added the explanations for SS_{Q1} and SS_{Q2} in the figure caption. The averaged size distributions at the four stages have been plotted in one figure (Fig. S7). The x-axis is presented on a linear scale to clearly exhibit the variations in the large droplet size range.

Fig. 7: How come that you still measure so many fog residuals even though the effective diameter is smaller than the cut-off of the CVI?

[Response] Thanks for pointing this out. The datapoints in Fig. 7 are N_d and D_{eff} measured by the Fog Monitor and the color of these datapoints are activation ratio (AR). The AR, which was defined as CCN number concentration relative the total particle concentration, were measured downstream of the GCVI system. As shown in the droplets size distributions during the fog formation stage (Fig. 6), even when D_{eff} was smaller than the cut-size of CVI, there still exist some large droplets exceeding the cut-size, which can be sampled by GCVI system.

Fig. 8a and eq. 6 don't match. Which one is the correct value for a?

[Response] Revised. It should be 0.027.

Fig. 8a: isn't the interesting regime the small visibilities when LWC>0? Why did you choose a linear axis and not like in Fig. 8b a log-axis?

[Response] We have added the fitting lines to this figure. When we used a log-axis, the fitting lines overlapped and hard to be distinguished.

Fig. 8: I would recommend to stick to the notation that you introduced earlier: VIS_K, VIS_KN, VIS_G, VIS_GN

[Response] Revised.

Fig. S1: typo in the colorbar title: 'Terrain'.

[Response] Revised.

Fig. S6 isn't mentioned in the text.

[Response] Revised. We have added the figure number in Line 289.

Fig. S8: please use dN/dlogD and plot the xaxis on a log-scale. The labels of the 2 curves have been switched: black is dry, blue ambient

[Response] Revised.

Please be consistent with your statistic parameters and linear regressions in all plots (r/r^2, p)

[Response] Suggestion adopted.

Please also be consistent with the time on the x-axis in the different figures and do not shrink and stretch the time. It makes it hard to compare the different fog events.

[Response] Suggestion adopted.

References:

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