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

## Reviewer 1

### General comment:

Eight fog events are observed and analyzed in this manuscript, with a focus on the characterization of fog microphysics and their relationships with visibility. This is a meaningful study that will likely attract the attention of ACP readers. However, I struggled with the manuscript for the following reasons:

[*Response*] We thank the reviewers for their thoughtful and constructive comments that help us improve the manuscript substantially. We have revised the manuscript accordingly. Listed below is our point-to-point response in blue to each comment that was offered by the reviewers. We hope that our revised manuscript will now be suitable for publication in ACP.

### Major comments

1. Analysis of Pre-Fog Aerosols

In Section 3.2, the authors explore the relationship between pre-fog aerosols and fog droplets. Under stable conditions, this relationship is logically sound due to weak wind speed. However, the article reports that wind speed during observation is relatively high (4 to 8 m/s), which suggests that advection plays a significant role in these fog events. The authors also state that "the pre-fog aerosols measured at the observation site may not fully represent the particles that actually activated into fog droplets." This raises the question: Can pre-fog aerosols be reliably replaced by aerosols observed during fog? The rationale behind this needs further explanation. Additionally, how does Section 3.2 lay the foundation for the subsequent content? The logic in Section 3.2 should be clarified.

In Section 3.3, pre-fog aerosols are used in the estimation by the $\kappa$-Köhler equation. How can the authors be certain that the pre-fog aerosols and those that activated into fog droplets share similar physical and chemical properties? For instance, fog event E3 had a long lifetime. Are the changes in aerosol physicochemical properties negligible? Observing supersaturation in fog is challenging, and bias is inevitable. The authors should discuss the sources of errors in this algorithm and provide references to support this approach. Wang et al. (2021) can be referenced.

[*Response*] Thanks for pointing this out. Although there is a temporal difference between the observation of pre-fog aerosols and the subsequent fog process at a fixed site, the measured pre-fog aerosol particles may not fully represent the particles that actually activated into fog droplets. However, due to the high altitude of this mountain site, it is located above the top of the boundary layer for most of the day (Sun et al., 2018). At this height, the aerosol concentration and properties are relatively homogeneous within a large spatial range. Although the observed fog droplets were partly formed elsewhere and advected to the site, especially in high wind speed conditions, the aerosol particles at the site are regionally representative, resulting in a good correlation between the pre-fog aerosol and the peak $N_d$ discussed in Section 3.2. Conversely, the good correlation between them also indicated the observations at this site were representative of a relatively large spatial scale. This provides a rational basis for estimating water vapor supersaturation by using the pre-fog aerosol size distribution in Section 3.3. We add these descriptions in the revised manuscript. Please see Lines 231-238. Additionally, we also consider add a sample inlet of total suspended particles in future experiments, which can obtain the information of both aerosol particles and fog droplets. This can help us gain a more comprehensive understanding of the properties of fog residual particles and fog interstitial particles.

As pointed out by the referee, the *SS* estimation algorithm in Section 3.3 considered only adiabatic processes such as activation and condensation, and ignores non-adiabatic processes such as collision-coalescence (Wang et al., 2021). If the reduction of $N_d$ caused by the collision-coalescence process is considered, the actual effective *SS* should be greater than the calculated value. We have added the sources of errors in this algorithm and provide relevant references. Please see Lines 256-259.

2. Mechanism in Fog Event E3

The authors note that "the main wind speeds ranged from 4 to 8 m/s" in lines 157-158, indicating that advection influences the observations. In lines 256-258, they state, "The enhanced supersaturation facilitated the further activation of smaller particles that were un-activated during the $SS_{Q1}$ stage, resulting in a secondary activation-dominated process during E3." Does this imply that un-activated aerosols from the $SS_{Q1}$ stage remained stationary without being affected by advection? This statement is confusing and potentially misleading.

The authors also mention "excess water vapor" in line 258. However, Figure 4 shows an increase in supersaturation from the $SS_{Q1}$ stage to the $SS_{Q2}$ stage during E3. Does lower supersaturation correspond to excess water vapor during the $SS_{Q1}$ stage? Please clarify this analysis.

[*Response*] Thanks for pointing this out. In-situ observations at a fixed site face significant challenges in continuously measuring the evolution of aerosols and fog droplets within a specific air mass. Here, we assume that at a certain height within the fog, the aerosols and fog droplets exhibit similar microphysical characteristics and undergo similar variations. Therefore, during a fog process, measurements at different time points at this site can, to some extent, reflect the evolution of the microphysical characteristics of aerosols and cloud droplets at that height. We add this assumption in the revised manuscript to clarify it. Please see Lines 281-285.

The excess water vapor mentioned in Line 258 is the difference between the partial pressure of vapor and the equilibrium value. When the production and depletion of excess water vapor in the early mature stage were in approximate balance, the first quasi-stationary supersaturation ($SS_{Q1}$) was reached. As the temperature decreased after the $SS_{Q1}$ state, the temperature-dependent equilibrium vapor pressure decreased faster than the partial pressure of vapor, leading to increases both in excess water vapor pressure and supersaturation during the $SS_{Q2}$ stage. We revised the description to further clarify that mechanism as follows:

Lines 296-298: "This indicated that the excess water vapor, defined as the difference of the ambient water vapor pressure and the equilibrium value, was produced and consumed in approximate balance, thus reaching a quasi-stationary supersaturation state."

Lines 317-322: "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). 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)"

3. In line 261, the authors discuss the "evaporation of liquid water from previously formed large fog droplets." 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 authors only mention large droplets in this context. Moreover, under the influence of advection, even if previous large droplets evaporate, they may not affect current observations. Is this correct? I suggest revising the analysis to clarify the mechanism.

[*Response*] As pointed out by the reviewer, both large and small droplets are affected by evaporation. The discussion here aims to explain the reduction in effective droplet radius. To avoid ambiguity, this is 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)."

**Minor Comments**

1. There is a formatting issue. When there is no space before a paragraph, a blank line should be inserted between consecutive paragraphs (e.g., a blank line is needed between lines 42 and 43). Alternatively, please refer to the formatting style of articles already published in ACP.

[*Response*] Thanks for the reviewer's suggestion. We have formatted the revised manuscript according to published articles in ACP.

2. In line 37, the article focuses on mountain fog; there is no need to mention maritime fog in the introduction.

[*Response*] Thanks for the reviewer's suggestion. We have removed the information of maritime fog from the Introduction in the revised manuscript.

3. Distinction Between Clean and Polluted Backgrounds.

In lines 159-163, the authors differentiate between clean and polluted backgrounds based on fog microphysical properties. However, the distinction between clean and polluted backgrounds should be based on aerosol concentration, as fog microphysics are also influenced by meteorological conditions. The concentration of cloud condensation nuclei (CCN) at the same supersaturation level would be more appropriate for this distinction. Numerous studies, such as Figure 2 in Wang et al. (2024), provide CCN concentration data under different background conditions.

[*Response*] Thanks for the reviewer's suggestion. In the revised manuscript, the aerosol concentrations have been used to differentiate between low and high number concentrations of aerosol backgrounds. Relevant information has been added in Lines 219-224 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 100-100 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."

4. In Section 2.1, the authors mention that the observation site is far from Hangzhou but claim that the site is generally near the top of the planetary boundary layer (PBL) around midday based on the PBL height of Hangzhou. This is unreliable because the boundary layer height varies by location.

[*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.

5. The installation of instruments is important for observation results. Could you provide photos of the observation setup in the supplement? This would help readers better understand the instrument installation.

[*Response*] As the reviewer suggested, we have combined the photos and the schematic of instrument setup together as Fig. S2 in the supplement, also shown as below:

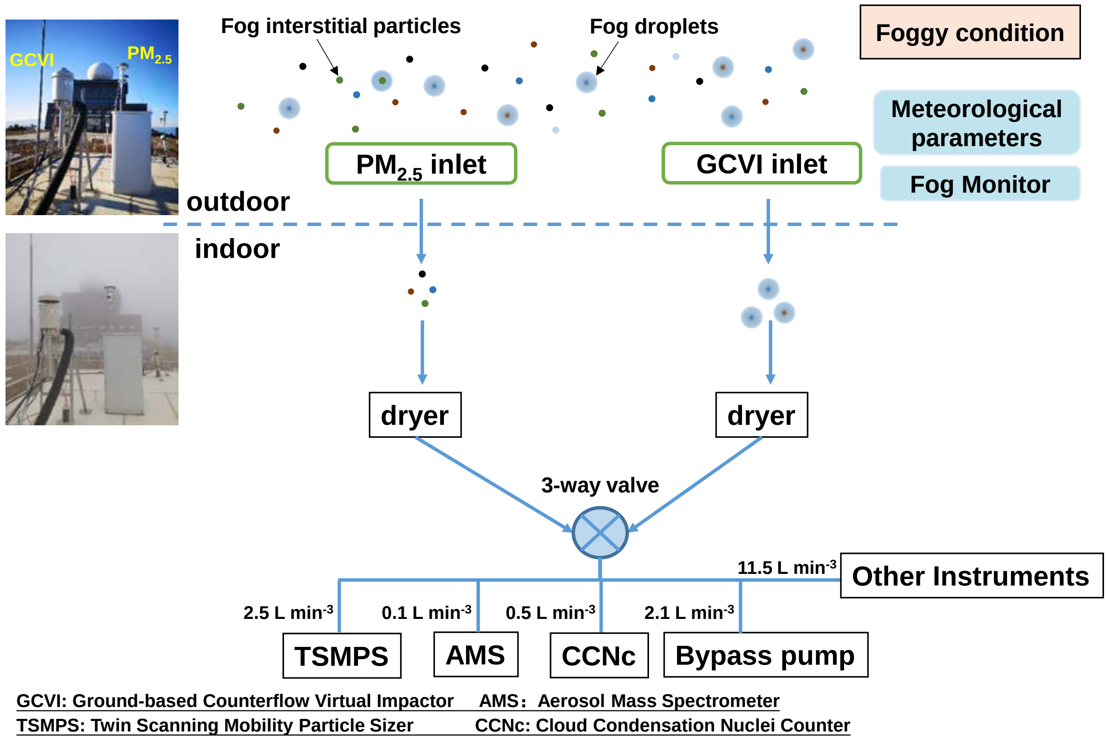


**Fig. S2. Schematic of the experimental setup at the Daming Mountain site. An automatic three-way switching system was placed between the sample inlets and instruments. Meteorological parameters and fog droplets were simultaneously measured on the roof of the observation container. The bypass pump only 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.**

6. In line 145, the threshold involved in the definition of fog requires a reference for support.

[*Response*] Suggestion adopted. We have added the relevant references in Lines 154-155 as follows:

Deng, Z., Zhao, C., Zhang, Q., Huang, M., and Ma, X.: Statistical analysis of microphysical properties and the parameterization of effective radius of warm clouds in Beijing area, Atmospheric Research, 93, 888-896, https://doi.org/10.1016/j.atmosres.2009.04.011, 2009.

Lu, C., Niu, S., Liu, Y., and Vogelmann, A. M.: Empirical relationship between entrainment rate and microphysics in cumulus clouds, 40, 2333-2338, https://doi.org/10.1002/grl.50445, 2013.

World Meteorological Organization: International Cloud Atlas - Manual on the Observation of Clouds and Other Meteors [WWW Document]. WMO-No. 407. URL https://cloudatlas.wmo.int/fog-compared-with-mist.html, 2017.

7. The information in the figures should be clearly explained. For instance, there is a lack of explanation for Dp in Figure 1; Q1 and Q2 are not explained in the title of Figure 6. Please check other figures.

[*Response*] Suggestion adopted. Here, $D_p$ in Figure 1 represents the diameter of droplet or particle. To avoid any confusion between them, we use $D_d$ and $D_p$ to denote the diameters of fog droplets and aerosol particles, respectively. Explanations for $SS_{Q1}$ and $SS_{Q1}$ have been added to the revised figure caption. We have also checked others figures thoroughly.

8. In line 158, there is an "s" at the end of "speeds." Is speed a countable noun?

[*Response*] Revised.

9. Water Vapor Consumption in Line 218

The hygroscopic growth of aerosols affects the water vapor mixing ratio, but temperature directly influences the saturated water vapor mixing ratio, not water vapor itself. The authors mention only water vapor consumption in line 218. Please reorganize the explanation to clarify the mechanism behind the relatively high supersaturation.

[*Response*] Thanks for the reviewer's suggestion. In the revised manuscript, we have revised the interpretation of the positive correlation between estimated *SS* and altitudes as below:

"This can be partly attributed to the lower aerosol number concentration and temperature at high altitudes (Liu et al., 2020b), which reduce excess water vapor consumption in clouds and fog, as well as the equilibrium vapor pressure (Baccarini et al., 2020; Shen et al., 2018), thereby promoting supersaturation."

10. Definition of Activation Ratio in Line 243

The authors define the Activation Ratio (AR) as "the CCN number concentration at a supersaturation setting of 0.2% relative to the total particle concentration." Why was 0.2% chosen? Please provide a reference to justify this choice.

[*Response*] Thanks for pointing this out. To avoid excessive *SS* variation in the CCNc column, the four *SS* setpoints were sequentially scanned from low to high and then back from high to low. Consequently, the number of data points for the intermediate *SS* values is twice that of the endpoint *SS* values. Meanwhile, CCNs with weaker activation are more likely to remain un-activated under low *SS* conditions. Based on the above considerations, the case of *SS* = 0.2% was selected in Fig. 6 to discuss the relationship between them. We have added the results for other *SS* setpoints to the supplement information (Fig. S10), and it can be seen that they present a phenomenon that is basically consistent with the results discussed for *SS* 0.2%. The relevant descriptions had added in the revised manuscript. Please see Lines 303-308.

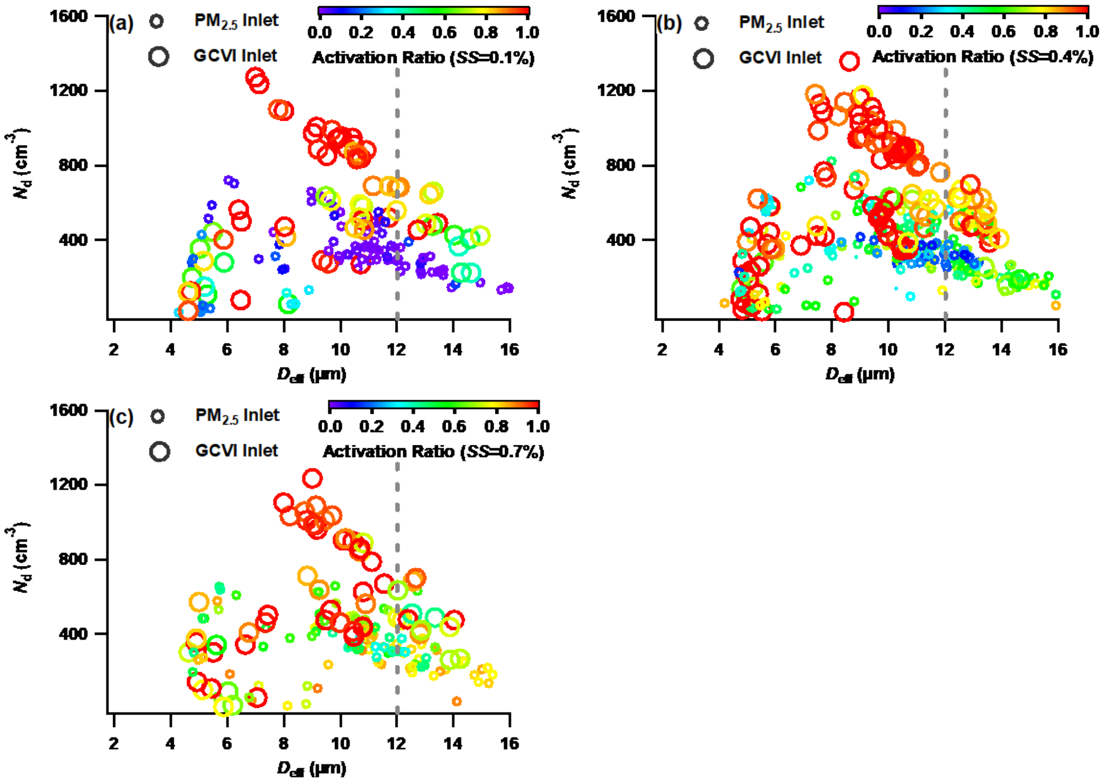


Fig. S10. Differences in CCN activity between fog residual particles (GCVI inlet) and fog interstitial particles ($PM_{2.5}$ inlet), and their variations with fog microphysical parameters: (a) *SS*=0.1%, (b) *SS*=0.4%, and (c) *SS*=0.7%. The gray dash line indicates significant collision-coalescence processes occurring when $D_{eff}$ exceeds 12 μm.

11. In line 270, why was 880 nm used in this study? Please provide a reference or explanation.

[*Response*] The wavelength used in the visibility meter is 880 nm. In order to make the *VIS* derived from the Mie theory is comparable with the *VIS* measured by the visibility meter, the same wavelength was used in the *VIS* calculation. We clarified it in the revised manuscript. Please see Lines 180-183.

12. In lines 296-299, the "≤" symbol is not in Times New Roman font.

[*Response*] Revised.

13. Introduction

In line 68, the authors focus on polluted regions. The criterion for distinguishing between polluted and clean backgrounds is aerosol mass concentration, but the authors do not use this threshold to determine whether the observation site is polluted or clean. Describing the background as having high or low aerosol loading would be more accurate. If the authors wish to continue using the terms "polluted" and "clean," they should provide criteria to support these distinctions.

In lines 67-68, The authors emphasize the impact of interactions between aerosols and fog microphysics on visibility ("their impacts on visibility degradation"). However, only the effect of aerosols on visibility is highlighted. What about the influence of interactions between aerosols and fog on visibility? Additionally, while the effect of aerosols on fog microphysics is analyzed in the manuscript, the effect of fog on aerosols is not addressed (Qian et al., 2023). The interactions between aerosol and fog should be more prominently discussed.

[*Response*] Thanks for your suggestion. The term "polluted region" here referred to the megacity cluster of the YRD region mentioned later in this sentence. The paper did not discuss clean or polluted weather conditions. In Section 3.4, the terms "low aerosol concentration condition" and "high aerosol concentration condition" are used, but their definitions were not provided. Following the referee's suggestion, we have added descriptions for the classification criteria in Lines 219-224.

For the interactions between aerosol and fog on visibility, we have discussed the effects of aerosol concentration on $N_d$ and evolution of fog droplets size distribution. These fog microphysical parameters significantly influence visibility, as discussed in Section 3.5. Additionally, we acknowledge that the effect of fog on aerosols is crucial for understanding the interactions between aerosols and fog. After participating the fog process, the chemical composition, mixing state, and morphology of aerosol particles would be changed (Schroder et al., 2015; Roth et al., 2016; Qian et al., 2023). At downstream of the GCVI inlet, the TSMPS, AMS and SP2 were also installed to measure physicochemical properties of fog residual particles. The results of these measurements will be used to analyzed the effects of fog on aerosol particles in a subsequent paper.

14. There are large uncertainties in the aerosol–cloud interactions (ACIs) (Fan et al., 2016). If the conclusion provides novel insights into ACIs based on the findings related to interactions between aerosols and fog, it could significantly enhance the manuscript's appeal and attract more attention.

[*Response*] Thanks for your suggestion. In the conclusion, we described the influence of pre-existing aerosol levels on the peak $N_d$ of each fog event and highlighted a secondary activation process that occurred during fog evolution. This process led to the formation of numerous small fog droplets, thus reducing the effective diameter. We acknowledge the effects of fog droplets on aerosol particles are also important for better understanding the interactions between aerosols and fog. Elaborate analysis for these measurements is prepared for a subsequent paper.

## References


Fan, J., Wang, Y., Rosenfeld, D., and Liu, X.: Review of Aerosol–Cloud Interactions: Mechanisms, Significance, and Challenges, J. Atmos. Sci., 73, 4221-4252, https://doi.org/10.1175/jas-d-16-0037.1, 2016.

Roth, A., Schneider, J., Klimach, T., Mertes, S., van Pinxteren, D., Herrmann, H., and Borrmann, S.: Aerosol properties, source identification, and cloud processing in orographic clouds measured by single particle mass spectrometry on a central European mountain site during HCCT-2010, Atmos. Chem. Phys., 16, 505-524, 10.5194/acp-16-505-2016, 2016.

Qian, J., Liu, D., Yan, S., Cheng, M., Liao, R., Niu, S., Yan, W., Zha, S., Wang, L., and Chen, X.: Fog scavenging of particulate matters in air pollution events: Observation and simulation in the Yangtze River Delta, China, Sci. Total Environ., 876, 162728, https://doi.org/10.1016/j.scitotenv.2023.162728, 2023.

Schroder, J. C., Hanna, S. J., Modini, R. L., Corrigan, A. L., Kreidenwies, S. M., Macdonald, A. M., Noone, K. J., Russell, L. M., Leaitch, W. R., and Bertram, A. K.: Size-resolved observations of refractory black carbon particles in cloud droplets at a marine boundary layer site, Atmos. Chem. Phys., 15, 1367-1383, 10.5194/acp-15-1367-2015, 2015.

Shen, X., Sun, J., Ma, Q., Zhang, Y., Zhong, J., Yue, Y., Xia, C., Hu, X., Zhang, S., and Zhang, X.: Long-term trend of new particle formation events in the Yangtze River Delta, China and its influencing factors: 7-year dataset analysis, Science of The Total Environment, 807, 150783, https://doi.org/10.1016/j.scitotenv.2021.150783, 2022.

Sun, T., Che, H., Qi, B., Wang, Y., Dong, Y., Xia, X., Wang, H., Gui, K., Zheng, Y., Zhao, H., Ma, Q., Du, R., and Zhang, X.: Aerosol optical characteristics and their vertical distributions under enhanced haze pollution events: effect of the regional transport of different aerosol types over eastern China, Atmos. Chem. Phys., 18, 2949-2971, 10.5194/acp-18-2949-2018, 2018.

Wang, Y., Niu, S., Lu, C., Lv, J., Zhang, J., Zhang, H., Zhang, S., Shao, N., Sun, W., Jin, Y., and Song, Q.: Observational study of the physical and chemical characteristics of the winter radiation fog in the tropical rainforest in Xishuangbanna, China, Sci. China, Ser. D Earth Sci., 64, 1982-1995, https://doi.org/10.1007/s11430-020-9766-4, 2021.

Wang, Y., Li, J., Fang, F., Zhang, P., He, J., Pöhlker, M. L., Henning, S., Tang, C., Jia, H., Wang, Y., Jian, B., Shi, J., and Huang, J.: In-situ observations reveal weak hygroscopicity in the Southern Tibetan Plateau: implications for aerosol activation and indirect effects, npj Clim. Atmos. Sci., 7, https://doi.org/10.1038/s41612-024-00629-x, 2024.