We thank RC1 and RC2 for their thorough reading of our manuscript and for their valuable comments.
We provide detailed responses to all comments below; the responses are shown in blue, and all revision are highlighted in red.

Review for “Regulation of Cloud Particle Spectra and Vertical Structure by Sub-Cloud Aerosols: Insights from Aircraft Observations and Numerical Simulations” by Liu et al.

Summary:
This study examines the impact of sub-cloud aerosol concentrations in different size ranges on cloud particle properties in liquid, mixed-phase, and ice clouds. The introduction and data sections are well-written. The numerical simulations are particular insightful and lend useful context to the process-level hypotheses made by the authors. The inclusion of the adiabatic cloud model in addition to analyzing flight data from numerous flights across hydrometeor phases is commendable and a step beyond studies that solely use either observational or modeling datasets.
That said, I have major concerns about the presentation and discussion of observations shown in Figure 2 and 3. More context on the flights (such as flight maps; context on where different phases were sampled), dataset classification (definitions of upper and lower layers, examples/highlights of an individual case or two), and objective analysis back by figures (see some brief comments on figure 2, 3) are sorely needed before the results are properly reviewed. In its current form, I am struggling to properly evaluate the conclusions drawn by the authors given the discrepancy between the figures and some of the text and the difficulty in examining the figures.
I recommend the authors rework the discussion and presentation of Figures 2 and 3 significantly before the manuscript can be considered for publication. In addition, there are some minor comments provided below.

We sincerely thank the reviewer for the careful evaluation of our manuscript and for the constructive comments. We appreciate the reviewer’s recognition of the strengths of the study, particularly the combination of aircraft observations and adiabatic cloud parcel model simulations, which helps provide process-level insight into aerosol–cloud interactions.
We also fully acknowledge the reviewer’s concerns regarding the presentation and interpretation of Figures 2 and 3, as well as the need for clearer descriptions of the flight context and dataset classification. Following these suggestions, we have made several substantial revisions to improve the clarity and robustness of the analysis.
Specifically, we have:
1. Added a map showing the aircraft flight tracks and observation regions, providing clearer geographical context for the measurements and indicating where different cloud phases were sampled.
2. Clarified the classification methods used in the analysis, including the criteria for high and low aerosol concentration groups and the definitions of normalized cloud layers (upper, middle, and lower).

3. Revised Figures 2 and 3 to improve their readability and interpretability, including clearer labeling, improved captions, and more explicit explanations in the text.
4. Expanded the discussion to better connect the observational evidence with the conclusions drawn in the study.
We believe that these revisions significantly improve the clarity of the figures and the transparency of the methodology, allowing the results and conclusions to be more clearly evaluated.
Detailed responses to each specific comment are provided below.

Major comments:
Abstract:

- The authors compare particle size/number in the upper and lower parts of the cloud layer. Firstly, "upper" and "lower" need to be defined objectively. Secondly, while differences in particle diameters are quantified, differences in droplet number concentration and spectral width (defined as 'broader size distributions') are not. From these results, it follows that the liquid/mixed-phase hydrometeor mass was considerably higher under high aerosol loading conditions? This should be clarified.

We sincerely thank the reviewer for the careful and constructive comments. We agree that improving the clarity of the abstract and the key figures (Figures 2 and 3) is essential for clearly presenting the results and supporting the conclusions. Following the reviewer's suggestions, we have made several substantial revisions to the manuscript.
We have clarified the definitions of "upper" and "lower" cloud layers. In the revised manuscript, cloud layers are objectively defined using the normalized cloud height (H), where H = 0 corresponds to the cloud base and H = 1 corresponds to the cloud top. The cloud was divided into three layers: lower layer ($H < 0.3$), middle layer ($0.3 \le H < 0.7$), and upper layer ($H \ge 0.7$).
We thank the reviewer for this helpful comment. Particle number concentration, spectral dispersion, and water content are mainly used to support the interpretation of the vertical variation in effective diameter, which represents the primary focus of this study. Therefore, these variables were not explicitly quantified in the abstract.
Nevertheless, we acknowledge their importance for a comprehensive interpretation. In the revised manuscript, we have provided a more detailed analysis of number concentration, spectral dispersion, and water content in Section 3.2.

Under high sub-cloud aerosol loading, effective droplet diameters in the lower part($H<0.3$) of liquid and mixed-phase clouds are 2.4 and 2.8 μm larger, respectively, than those in the upper part ($H>0.7$).

In addition to number concentration and effective diameter, the liquid water content (LWC) also exhibits distinct vertical variations under different aerosol conditions (Figures 4d and h). Under high-aerosol conditions, LWC in the lower layer is higher than that in the upper layer by approximately 0.37 g m^{-3}, which is consistent with the vertical variations in spectral dispersion and effective diameter described above. A similar vertical structure is observed in mixed-phase clouds; however, LWC is further influenced by phase transition processes. Under high-aerosol conditions, LWC in the upper layer is lower than that in the lower layer by approximately 0.26 g m^{-3}.

- The authors should specify here if the quantitative comparisons are based solely on aircraft data or numerical simulations also.

We clarified that the quantitative comparisons in the abstract are based primarily on aircraft observational data, while the adiabatic parcel model simulations are used to interpret the underlying physical mechanisms.

Observations show that in liquid and mixed-phase clouds, droplet number concentration peaks occur at droplet diameters of 5–7 μm and 14–15 μm, whereas ice and mixed-phase clouds exhibit ice crystal peaks occur at particle sizes of about 125 and 1550 μm.

- Please review ACP submissions guidelines for authors at https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html. State importance and implications of the results in the abstract. The abstract ends abruptly with a result statement.

Following the ACP author guidelines, we revised the abstract to include a brief statement of the broader significance and implications of the results.

These results improve our understanding of aerosol–cloud interactions and provide important constraints for their representation in weather and climate models.

The two key figures in section 3 need some work to be better interpretable:
Figure 2: This figure needs better labels and an explanation of why two rows represent ice clouds. The discussions in text refer to upper and lower layers of clouds whereas the labels indicate “low, mid, high”?

For Figure 2, we improved the labeling and revised the caption to clearly explain the structure of the figure. The two rows showing ice crystals represent ice crystals observed in mixed-phase clouds and those in pure ice clouds, rather than two different types of ice clouds. This clarification has been added to the caption.

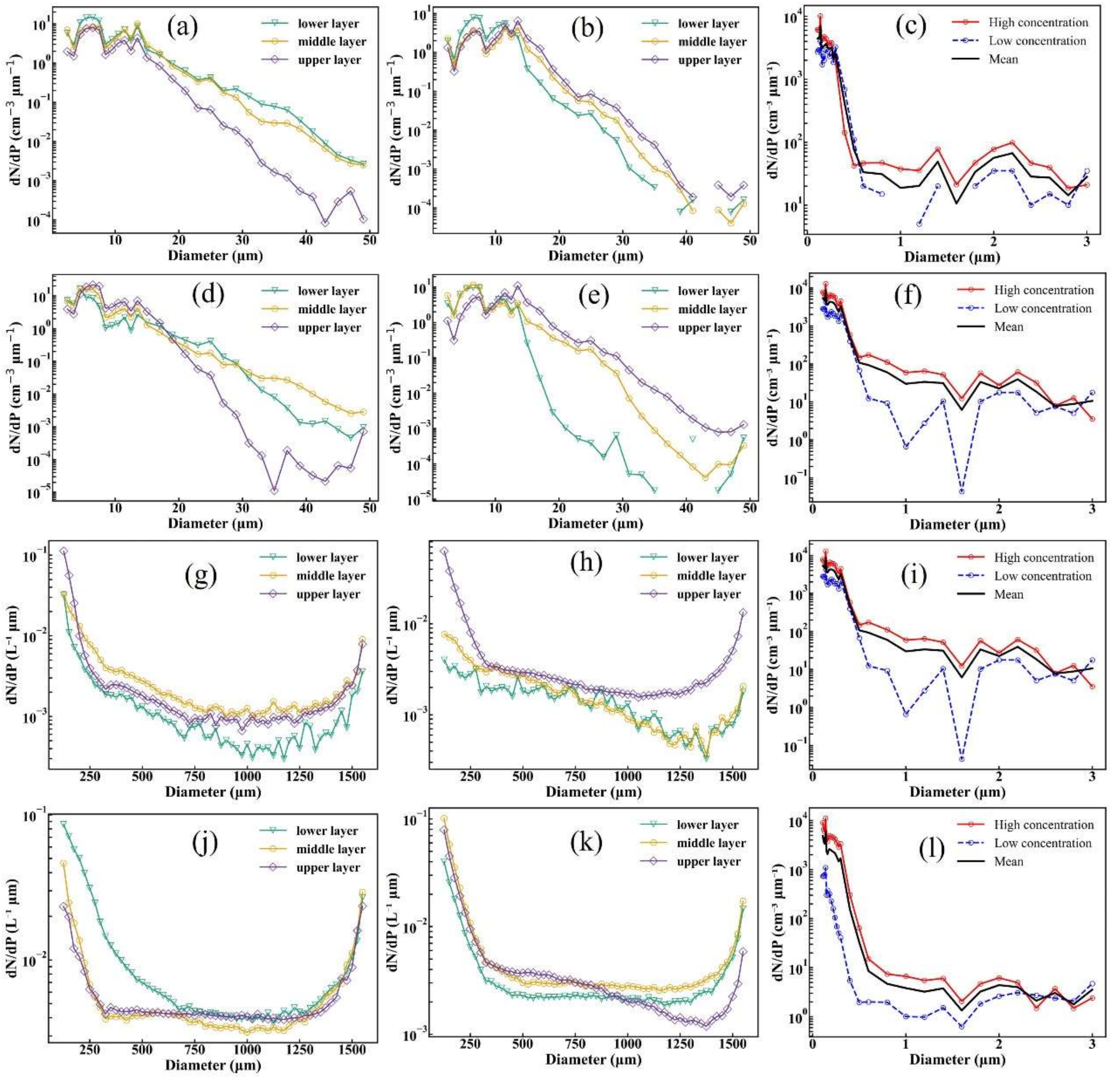

Figure 3. Size distributions of cloud particles in different regions of the observed clouds and aerosol size distributions below the clouds under varying aerosol concentrations. Figure 2. Size distributions of cloud particles in different regions of the observed clouds and aerosol size distributions below the clouds under varying aerosol concentrations. Panels (a–b) show cloud droplet spectra in warm clouds under high and low aerosol loading, respectively; (d–e) show cloud droplet spectra in mixed-phase clouds; and (g–h) show ice crystal spectra in mixed-phase clouds. Panels (j–k) present ice crystal spectra in ice clouds. In each panel, yellow, green, and purple curves represent the cloud top, middle, and base, respectively. Panels (c), (f), (i), and (l) show the corresponding aerosol size distributions below the clouds under high- and low-aerosol conditions, along with the mean distribution (black line).

Figure 3:

- This forms a key figure in the authors' analysis and interpretation of the aerosol impacts on particle distributions. As such the panels are very noisy and it is difficult to objectively analyse the results and their claims. Maybe a binning approach would be better – bin data over the upper, mid, and lower layer of the cloud profile?

For Figure 3, we agree that the original scatter plots contained substantial variability and were difficult to interpret. Following the reviewer's suggestion, we adopted a binning approach based on normalized cloud height, grouping the observations into vertical bins and displaying the mean values for each bin, which significantly improves the clarity of the vertical trends. In the revised manuscript, the original Figure 3 has been updated and is now presented as Figure 4, as shown below.

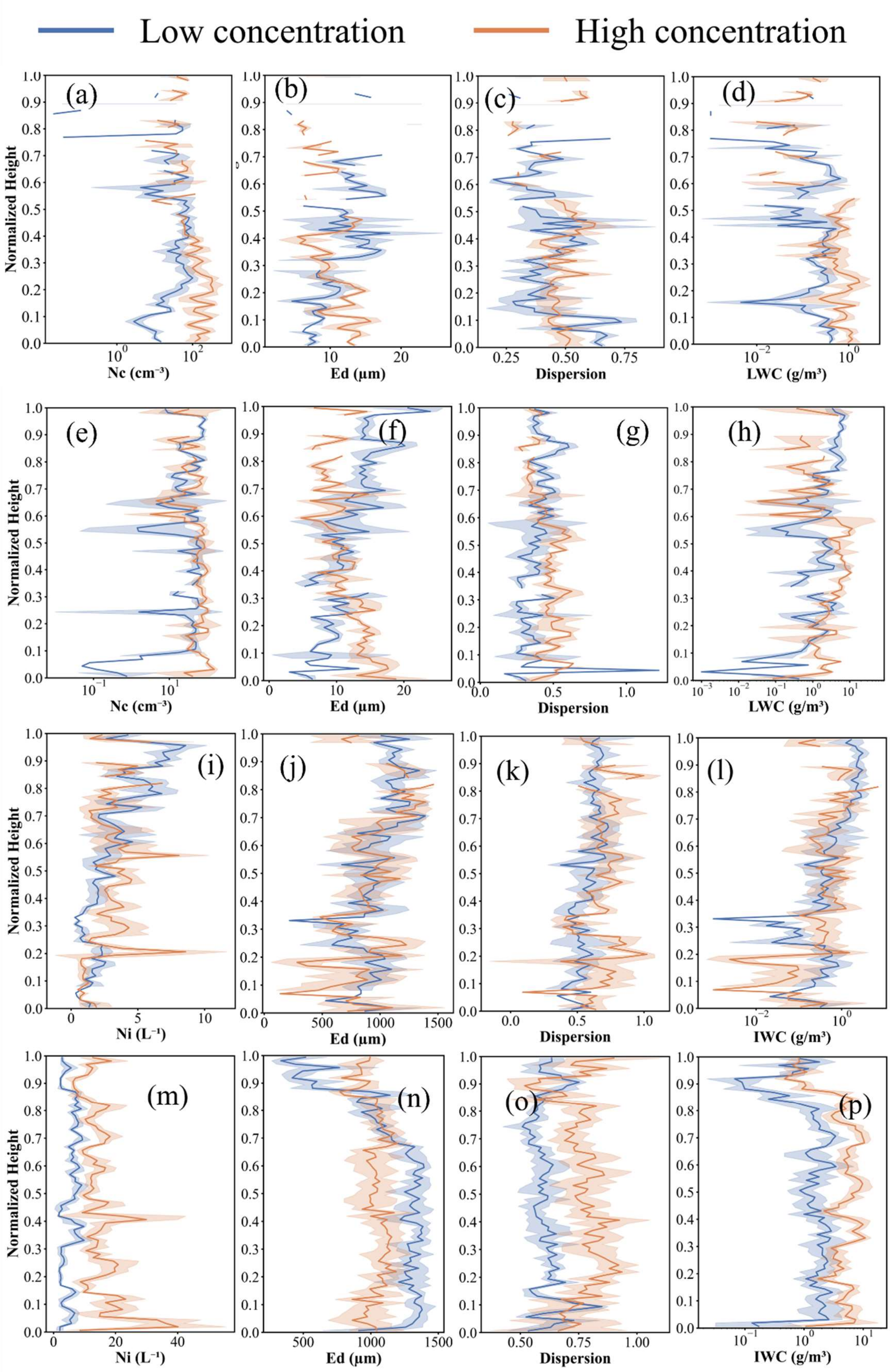

Figure.4 Vertical distributions of different cloud particle characteristics. (a–d) water cloud droplets, (e-h) mixed-phase clouds droplets, (i-l) ice crystals in mixed clouds,

(m–p) ice crystals in ice clouds. For each cloud type, the panels from left to right represent number concentration, effective diameter, dispersion, and water content, respectively.

- Line 208: Apart from maybe panel m, I'm really struggling to see how high aerosol conditions have higher particle concentrations. In fact, near cloud base, panel a shows low droplet concentrations near cloud base under conditions of high aerosol concentrations.

We thank the reviewer for carefully examining the figure. After rechecking the original plotting script, we found that the color assignment for the high- and low-aerosol conditions in panel (a) was inadvertently reversed when adjusting the color scheme to make the figure color-blind friendly. As a result, the curves representing the two aerosol regimes were displayed incorrectly, which led to an apparent contradiction with the description in the text.
This issue has now been corrected in the revised figure. After fixing the color assignment, the particle number concentrations under high-aerosol conditions are generally higher than those under low-aerosol conditions, consistent with the interpretation provided in the manuscript.

- Line 210: I'm struggling to see this claim represented in Fig. 3a, e. In fact, I see the opposite trend in Fig. 3a as blue colors representing low aerosol concentration conditions have lower values near cloud top.

We thank the reviewer for carefully examining the figure. As mentioned in the response to the previous comment, we identified an error in the color assignment of panel (a) when adjusting the figure to a color-blind-friendly scheme. The curves representing high- and low-aerosol conditions were inadvertently reversed, which caused the apparent inconsistency between the figure and the description in the text. This issue has been corrected in the revised figure. After correcting the color assignment, the vertical variations in Fig. 3a and 3e are consistent with the interpretations presented in the manuscript.

- Line 216: I don't see an increase in Ed with height for low aerosol concentrations.

We thank the reviewer for this helpful comment. The apparent inconsistency arises from the same issue noted above, namely an error in the color assignment introduced when the figure was adjusted to a color-blind-friendly scheme. In the original figure, the curves representing high- and low-aerosol conditions were inadvertently reversed, which affected the visual interpretation of the vertical variation in effective diameter (Ed) under low-aerosol conditions.
This issue has now been corrected in the revised figure. After correcting the color assignment, the vertical variation of Ed under low-aerosol conditions is consistent with

the corresponding description in the manuscript.

- Panels o, p should be reversed as dispersion is in column 3 in the preceding rows.

We appreciate the reviewer for identifying the panel ordering issue. The positions of panels (o) and (p) were inadvertently reversed in the original figure. This has now been corrected in the revised manuscript so that the variables are consistently arranged across all rows.

Minor comments:
Abstract:

- Please maintain consistency in terminology. Authors use the terms "sub-cloud aerosol loading", "low-aerosol conditions", and "sub-cloud aerosol concentrations", in three consecutive sentences to mean the same thing (I assume).

We thank the reviewer for pointing out this inconsistency. In the revised manuscript, we have standardized the terminology and now consistently use "sub-cloud aerosol loading" when referring to aerosol conditions below the cloud base.

- Is there a difference between "mixed-phase clouds" and "ice clouds" on line 21?

We thank the reviewer for pointing out this ambiguity. In the original sentence, the phrase "both cloud types" refers specifically to liquid clouds and mixed-phase clouds.

- Is the full stop before "at low aerosol concentrations" intended?

We thank the reviewer for identifying this punctuation issue. The full stop before "at low aerosol concentrations" was not intended. It has been removed in the revised manuscript and the sentence has been corrected accordingly.

Line 14-15: Please specify radius or diameter.

We thank the reviewer for this helpful suggestion. In the revised manuscript, we have clarified that the particle size referred to in this sentence corresponds to particle diameter.

In liquid and mixed-phase clouds, droplet number concentration peaks occur at droplet diameters of 5–7 µm and 14–15 µm, whereas ice and mixed-phase clouds exhibit ice crystal peaks occur at particle sizes of about 125 and 1550 µm.

Line 17: Please rephrase and/or break sentence into two parts, it is hard to understand.

Following the reviewer's suggestion, the sentence has been rewritten and divided into two shorter sentences to improve clarity.

Under high sub-cloud aerosol loading, effective droplet diameters in the lower part of liquid and mixed-phase clouds are 2.4 and 2.8 μm larger, respectively, than those in the upper part. The lower cloud layers also exhibit higher droplet number concentrations and broader size distributions.

Line 50: Can the authors list a couple of the findings corresponding to these citations?

We thank the reviewer for this helpful suggestion. In the revised manuscript, we have added brief descriptions of the main findings from the cited studies in this section to provide clearer context for the referenced work.

In China, aircraft-based cloud observations, which have been conducted since the 1960s, have become fundamental data sources, yielding numerous significant findings (Chang et al., 2019; Feng et al., 2021; Wang et al., 2014; Zhao et al., 2018). For example, in South China's Guangxi region, Liu et al. (2025) analyzed nine aircraft flights conducted in 2020 to investigate the diurnal variation of stratocumulus clouds and the impact of aerosols on cloud microphysical properties. They found that aerosol number concentration and cloud droplet effective radius exhibited the typical Twomey effect and highlighted the important influence of boundary-layer height on the microphysical structure. Based on 35 aircraft flights conducted over the North China Plain during 2019–2021, Ke et al. (2025) further investigated the influence of boundary-layer–cloud coupling processes on cloud microphysical variables.

Line 54: "Jae et al. (Yeom et al. (2025)" needs to be corrected.

Thank you for identifying this reference error. The citation has been corrected.

Line 76: Some of the uncertainties, specifically those the authors intend to address in this work should be mentioned here.

We thank the reviewer for this helpful suggestion. In the revised manuscript, we have expanded this section to highlight the remaining uncertainties in previous studies, particularly regarding the vertical variations of cloud microphysical properties under different aerosol conditions. We have also clarified the specific aspects that this study aims to address, including the relationships between sub-cloud aerosol concentrations and cloud particle characteristics in different cloud phases.

Despite these advances, the effects of aerosols on cloud microphysical properties remain uncertain. It is still unclear whether higher aerosol concentrations necessarily lead to higher cloud particle number concentrations and smaller effective diameters.

Moreover, it remains an open question whether aerosol impacts differ among clouds of different phase states. In this study, we combine extensive aircraft observations from 16 flights over the North China Plain between 2019 and 2021 with results from an adiabatic cloud model to systematically investigate the spectral distributions, vertical structures, and aerosol-induced responses of cloud particles across clouds of different phase states.

Line 87: There needs to be consistency in the information provided for different probes – please provide size range, number of bins (channels), range of particle sizes used in the study, uncertainty estimates if available, for each probe. Also please delineate between primary measurement and derived quantities. Perhaps a table with all these details might be suitable.

We thank the reviewer for this helpful suggestion. In the revised manuscript, we have provided a more consistent description of the aircraft probes used in this study. Specifically, we now include the measurement size range, number of channels, and the particle size ranges used in the analysis for each probe.
Following the reviewer's recommendation, we have also added a summary table that compiles the key information for each probe to improve the clarity and readability of this section.

Table.1 Summary of airborne probes used in this study.

Instrument	Measurement range (μm)	Number of channels	Variables used in this study
PCASP	0.1–3.0	30	Aerosol size distribution, aerosol number concentration
CDP	2–50	30	Droplet size distribution, droplet number concentration (Nc), effective diameter (Ed), spectral dispersion, liquid water content (LWC)
CIP	25–1600	64	Ice particle size distribution, ice crystal number concentration (N_{ice}), effective diameter, ice water content (IWC)

Line 98: I'm very confused on how many flights were used in this study because the introduction states 21 flights, then here the authors state 35 flights but the paragraph ends with a total of 29 flights, and Table 1 describes "sorties" which are not defined yet and there are only 16 sorties with multiple sorties on a single date.

We appreciate the reviewer's careful reading and for pointing out the inconsistencies in the description of the flight numbers. After rechecking the dataset, we found that the number 21 flights mentioned in the introduction was incorrect and has been corrected in the revised manuscript.

In this study, a total of 16 aircraft flights with cloud observations were used in the analysis. However, during some flights, observations were conducted in different locations and encountered different cloud types, which were treated as independent cases in the analysis. After applying quality control and filtering procedures to avoid counting mixed-phase and ice clouds from the same cloud cluster within a single flight, the dataset resulted in 29 independent cloud cases used in the statistical analysis.
In addition, the term “sortie” has now been explicitly defined in the revised manuscript to clarify that it refers to an individual aircraft mission during the field campaign.
The corresponding descriptions in the manuscript and Table 1 have been revised to ensure consistency throughout the paper.

Table 1: Previously, it was mentioned that data from two flight platforms were used but Table 1 only lists K350 flight sorties?

We thank the reviewer for pointing out this potential confusion. The observational dataset includes 35 flight sorties conducted by two different aircraft platforms. However, after data screening and quality control, only the observations from the K350 aircraft were selected for the analysis presented in this study.
Therefore, Table 1 lists only the K350 aircraft sorties used in the analysis. Observations from the other aircraft platform were used only as supporting information and were not included in the statistical analysis.

Line 105: “number concentration distribution” is awkward, I believe you mean to say the “number distribution function”, which is a term used in previous studies using airborne cloud probe datasets.

We thank the reviewer for this helpful suggestion. We agree that the phrase “number concentration distribution” was not sufficiently clear. In the revised manuscript, we have replaced this expression with “number concentration per size interval” to more accurately describe the particle number distribution derived from the probe measurements.

The number concentration per size interval for each size channel of the CDP, CIP, and PCASP instruments was calculated as follows:

$$N_i = \frac{n_i}{1000 V_i \Delta r_i}$$

where N_i represents the number concentration density of the i-th channel for each instrument (cm^{-3}·μm^{-1}),n_i is the number of particles in the i-th channel, V_i is the sampled volume for the i-th channel (L), and Δr_i is the bin width of the i-th channel (μm).

Line 117: should “n_i” be replaced by “N_i times delta r_i”? and does d_i not represent the central diameter for the ith size channel as stated previously? “droplet diameter” is misleading given that the diameter for individual droplets is not estimated.

We thank the reviewer for this careful and helpful comment.First, “n_i” in the equation represents the particle counts recorded in the i-th size channel by the probe, which are used to calculate the number concentration per size interval. Therefore, “n_i” is the directly measured quantity and is not replaced by “N_i times delta r_i”.
Second, we confirm that d_i presents the central diameter of the i-th size channel, as defined by the probe bin configuration. To avoid ambiguity, we have clarified this definition in the revised manuscript.
Finally, we agree that the phrase “droplet diameter” may be misleading because the probes measure particle counts within predefined size bins rather than estimating the diameter of individual droplets. we have modified the wording to refer to the bin-center diameter of each size channel, defined as the midpoint between the upper and lower bounds of the size bin.

Line 118: what is the value of n?

We thank the reviewer for pointing out that the definition of the parameter n was not clearly stated. In the revised manuscript, we have clarified that n represents the number of particle size channels used in the calculation, which is 30 in this study. This definition has been explicitly added in the Methods section.

Line 125: mention that m is the number of channels? What is the value of m?

We thank the reviewer for pointing this out. In the revised manuscript, we have clarified that
m represents the number of particle size channels used in the calculation, and its value is 60 in this study. This definition has been explicitly added in the Methods section.

Line 128: the parameter “effective radius” was defined much earlier in a study by Hansen and Travis, 1974, which may be an appropriate reference here. Again, the authors may want to maintain consistency across the equations (this is the first equation that has the i subscript for the Greek symbol for summation).

We thank the reviewer for this helpful suggestion. In the revised manuscript, we have added the reference to Hansen and Travis (1974) when introducing the definition of the effective radius. In addition, we have revised the equations to ensure consistent notation throughout the manuscript, including the use of the summation index in the equations.

Line 141: The authors need to define N_c and N_i first. The acronym “N_i” has already been used as a variable in an equation above, so I recommend using “N_{ice}”. Was there no “N_{ice}” threshold used to define liquid clouds? Why not include LWC and IWC as determinates of cloud phase? For an ice cloud threshold for temperature, I would imagine that for values up to 5 C, there may be some mixed phase segments. Please clarify.

We thank the reviewer for these helpful comments. First, we agree that the definitions of N_c and N_i should be introduced more clearly. In the revised manuscript, we have explicitly defined N_c as the droplet number concentration and N_{ice} as the ice crystal number concentration when they first appear in the text. To avoid confusion with the previously defined N_i representing the number concentration in the i-th size channel, the notation N_{ice} has been adopted throughout the manuscript.
Second, regarding the cloud phase classification, the identification of liquid, mixed-phase, and ice clouds in this study is based on particle number concentration thresholds combined with temperature conditions. In addition, liquid water content (LWC) and ice water content (IWC) are derived from the observed particle size distributions rather than directly measured. Therefore, in this study, cloud phase classification is based solely on number concentration and temperature thresholds.
Finally, we clarify that the temperature threshold used in this study is −5 °C, rather than +5 °C. This threshold, together with particle observations, is used to distinguish ice clouds from mixed-phase clouds.

Line 143: Sections 2.3 and 2.4 have the same heading, please adjust appropriately.

The duplicated section heading has been corrected in the revised manuscript, and the section titles have been adjusted accordingly.

Technical corrections:
Line 56: (2025) -> (2025). Similar error repeats throughout the manuscript. Please edit following the journal's recommended format.

The citation format has been corrected throughout the manuscript in accordance with the ACP formatting guidelines, including the correction from "(2025)" to "(2025)" and similar inconsistencies elsewhere in the text.

Line 61: do you mean ice nucleating particle concentrations?

Yes, the intended meaning here is ice nucleating particle (INP) concentrations. We have clarified this terminology in the text to explicitly refer to ice nucleating particles.

Line 69: Please define E3SM as was done for other models.

E3SM has been defined at its first occurrence as the Energy Exascale Earth System Model, consistent with the treatment of other model names in the manuscript.

Line 117: "channel" is written twice.

The duplicated word “channel” has been removed.