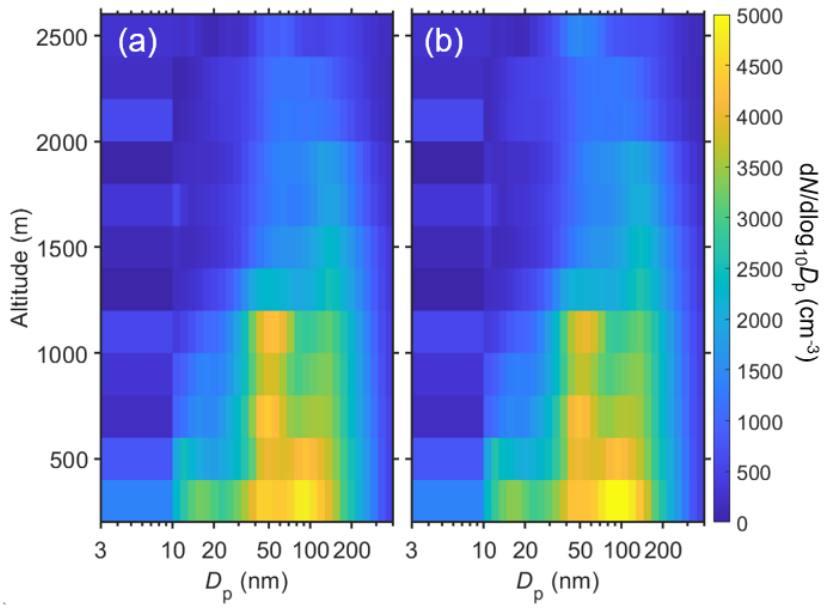
Manuscript No.: EGUSPHERE-2023-830
Title: Vertical profiles of trace gas and aerosol properties over the Eastern North Atlantic: Variations with season and synoptic condition

SUMMARY:
1. In the manuscript the authors study the vertical profile of aerosol particle properties over the Southern Great Plains (SGP) atmospheric observatory. The vertical profiles were measured using an instrumented Gulfstream I aircraft during two intensive campaigns in spring and summer 2016.
The observed vertical profiles are explained by the authors using aerosol dynamics and transport processes. The results add to the understanding of aerosol properties at SGP and motivate further studies. The text is clear, well-structured and the topic is scientifically relevant. I can recommend the manuscript for publication in ACP after the below comments have been addressed.
Response: We thank the reviewer for the positive feedback. We have thoroughly revised the manuscript based on the comments provided. The point-to-point responses can be found as follows.

COMMENTS:
2. Fig 1: It appears that the flights were done on average about two hours later during the spring campaign than during the summer campaign. Would the time difference contribute to a possible bias when comparing the time periods? Is it possible to check that the results don't change by comparing only overlapping times of day?
Response: The U.S. daylight-saving time of the year 2016 (when HI-SCALE was conducted) starts on March 13 and ends on November 6. The campaign was conducted between April 25 and September 23, which means that there was no change in the local time due to daylight saving. The reason the flights were earlier and shorter (in general) during the summer campaign is because of the hotter temperatures, which affect flight operations. It is more difficult to lift off with a full payload with hot temperatures (when the temperature is above ~ 32 ℃), and it consumes more fuel to fly. Therefore, the science team flew earlier in the morning during the summer campaign to have the coolest temperatures possible.

Regarding the impact of the flight time on temperature, the difference in time by a few hours is unlikely to cause significant bias. The boundary layer (BL) development is different for spring and summer, with BL likely developing faster during the warmer summer period. Moreover, time is one of the many factors influencing the vertical heterogeneity of aerosols, and our manuscript tries to identify these factors and analyze their influences. We also discussed the time effect in the manuscript regarding how boundary layer development affected aerosol vertical distribution in Section **3.4.1**.

To address the reviewer's comment, we plotted the averaged vertical size distributions during the summer IOP excluding the two late afternoon flights on September 15, 2016 and September 20, 2016 (panel (a) of the figure below), and the results are very similar compared to the results that include all flight times (panel (b) of the figure below).



3. Fig 2 a and b: Add a scale bar indicating horizontal distance.

Response: The scale bars indicating horizontal distance have been added to **Fig. 2**. The revised figure can be found below.

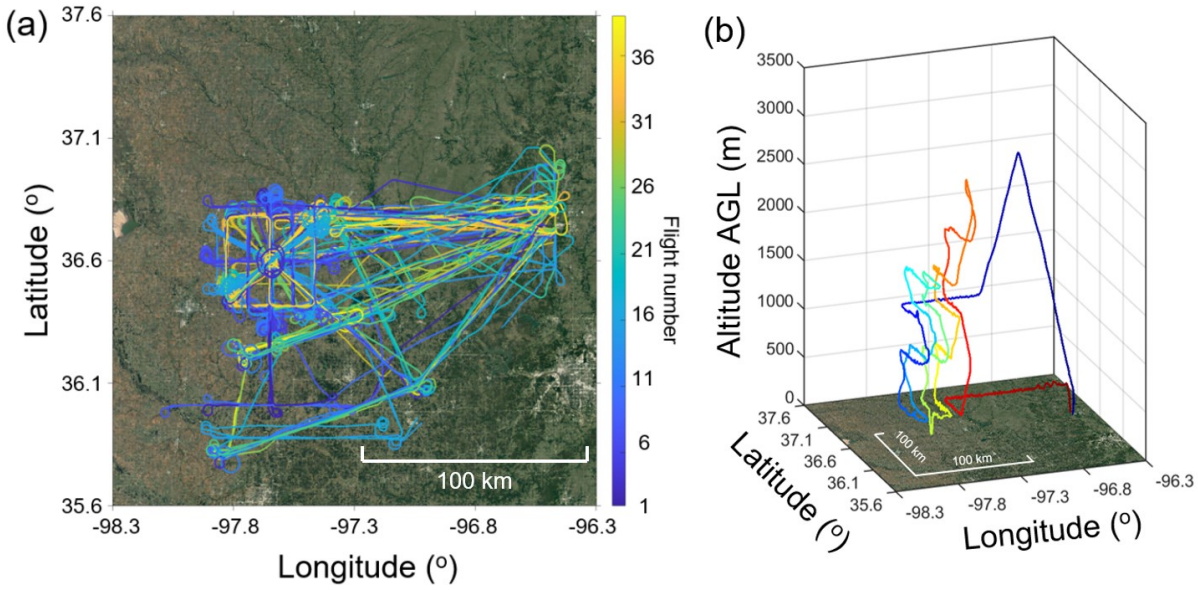


Fig. 2. (a) Flight patterns used during the HI-SCALE campaign. A total number of 38 flights were conducted over the SGP observatory. Different colors of the flight patterns represent different flight numbers. (b) An example of the flight pattern on May 3, 2016. The color represents the local time in hours. Maps are obtained from ©Google Maps.

4. Fig 3: For evaluating the source area better it would be helpful to see the distribution of back trajectories associated with each cluster.

Response: The trajectories associated with each cluster in **Fig. 3** are now added as **Figs. S3** and **S4**. The added figures can be found below.

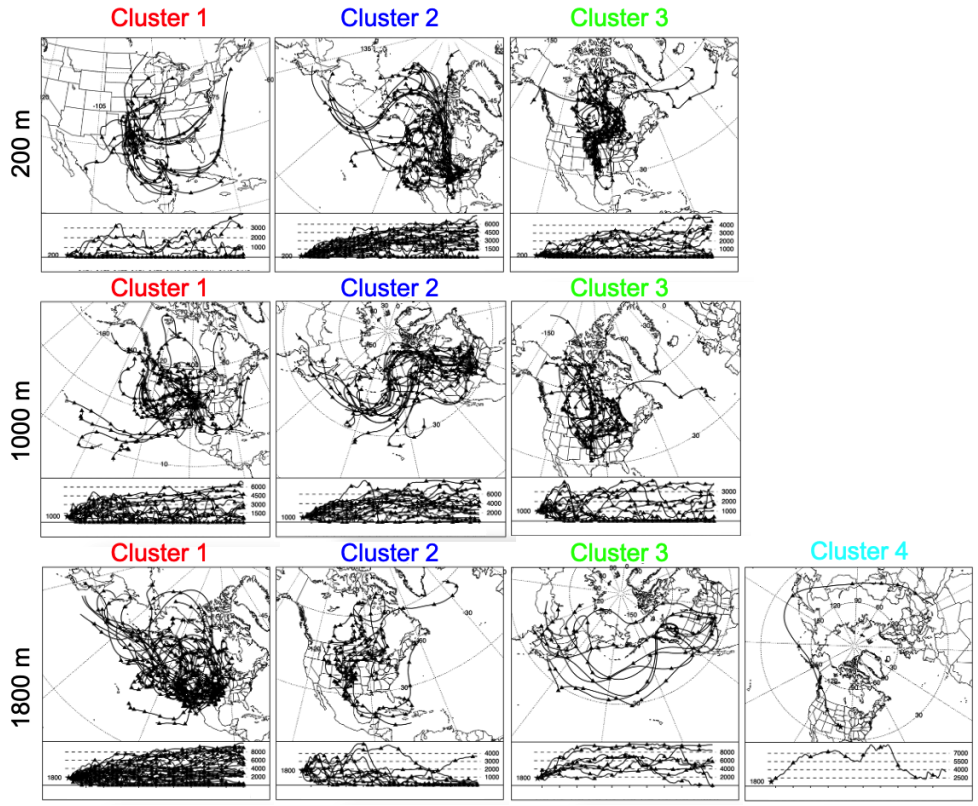


Fig. S3. Trajectories associated with each cluster during the spring campaign shown in Fig. 3.

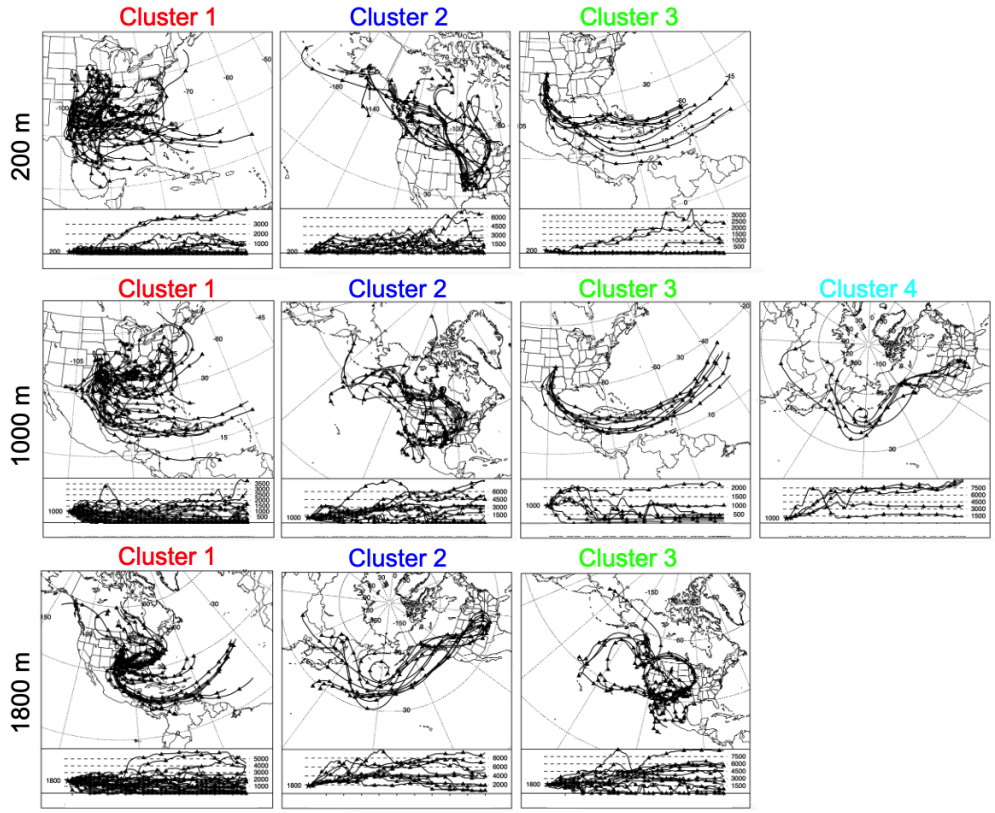


Fig. S4. Trajectories associated with each cluster during the summer campaign shown in Fig. 3.

5. Fig 4: Add a size bin for 3-10 nm size range from the two onboard CPCs.

Response: The size bin for 3 to 10 nm particles is now added in the revised **Fig. 4**. The figure caption is also revised to reflect the methods for calculating the size distributions. The revised figure can be found below.

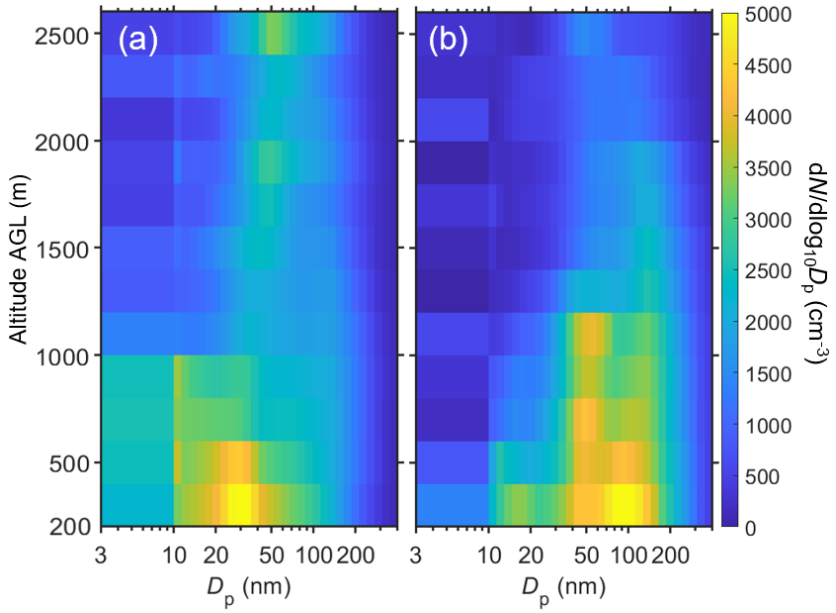


Fig. 4. Vertical profiles of the mean aerosol size distributions during the (a) spring campaign and (b) summer campaign. Note that the size distributions between 3 and 10 nm are calculated based on the difference of concentrations monitored by the two CPCs, and the size distributions above 10 nm are obtained by the FIMS. The size distributions are normalized to standard temperature and pressure (273.15 K and 101.325 kPa; STP).

6. Fig 5: Add the vertical profile of 3-10 nm number concentration calculated from the CPCs. Also add a legend showing that red is summer and blue is spring (same for Fig. 6).

Response: We have now included the analysis of $N_{3\text{-}10}$ in the updated **Fig. 5** (also shown below). $N_{3\text{-}10}$ showed a consistent trend with $N_{10\text{-}20}$, meaning that these aerosols are associated with the nucleation-mode aerosols generated from NPF events. To include the contribution of $N_{3\text{-}10}$ to total aerosol concentration, we have also changed the analysis and used $N_{>3}$ (instead of $N_{>10}$) to represent total aerosol concentration. Because of this change, the discussion regarding the vertical profiles of total aerosol concentration has changed, because now, springtime $N_{>3}$ is consistently higher than summertime $N_{>3}$ at all altitudes below 2500 m AGL due to the large contribution of nucleation mode aerosols to total aerosol number concentrations. These changes have been included in both the discussions regarding **Fig. 5** and the conclusion. Legends are now added to differentiate spring and summer campaigns in **Figs. 5, 6, S6** and **S7**.

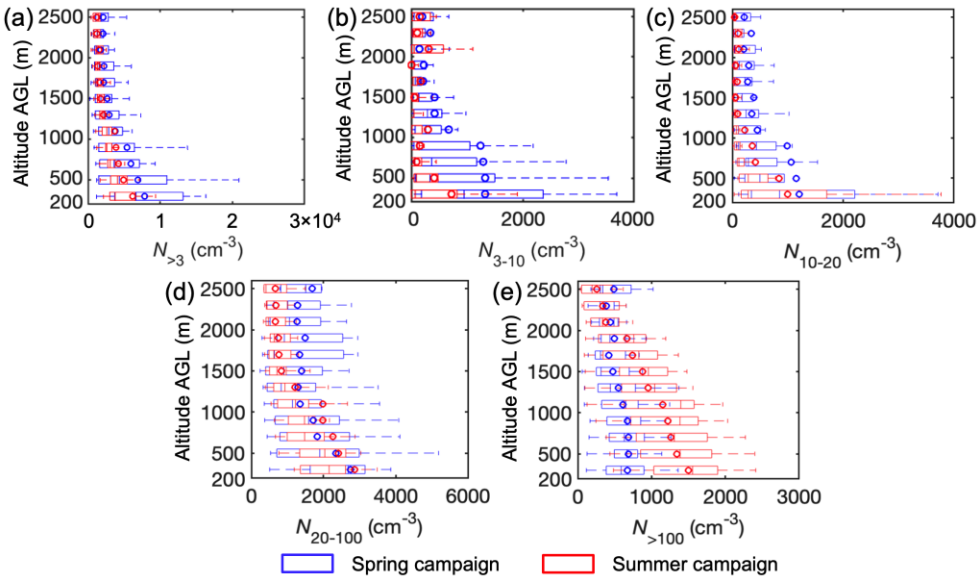


Fig. 5. Vertical profiles showing the concentrations of particles (a) larger than 3 nm ($N_{>3}$), (b) with sizes between 3 and 10 nm ($N_{3\text{-}10}$), (c) with sizes between 10 and 20 nm ($N_{10\text{-}20}$), (d) with sizes between 20 and 100 nm ($N_{20\text{-}100}$), (e) with sizes above 100 nm ($N_{>100}$) over the SGP site during the spring campaign (blue) and summer campaign (red). The line and circle markers represent the median and mean of the data, and the edges of the box indicate the 25th and 75th percentiles, respectively. The concentrations are normalized to standard temperature and pressure (273.15 K and 101.325 kPa; STP).

7. In addition to the vertical profiles of number concentrations and mass concentrations could you include a figure with average vertical profiles of the meteorological variables such as potential temperature as well as LWC. Alternatively add them as subplots to Fig. 5.

Response: We have now included the boxplots showing the potential temperature and LWC (figure below). However, as they are not directly related to the aerosol measurements, we have now included the figure in the supplementary materials as **Fig. S2**. The following discussion has been added to Section 3.1 in the manuscript:

> "**Figure S2** shows the vertical profiles of potential temperature and LWC during the spring and summer campaigns. Note that overall, there is minimal LWC at altitudes above 1500 m (**Fig. S2b**). However, the inversion of the potential temperature may occur even when there is no LWC, meaning that the profile of LWC in **Fig. S2b** could only qualitatively show the BL height."

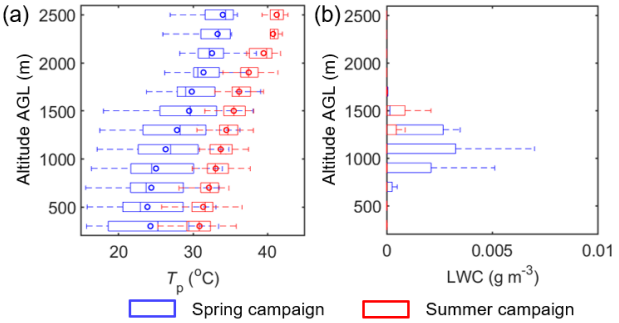


Fig. S2. Vertical profiles of (a) potential temperature (T_p) and (b) liquid water content (LWC) during the spring and summer campaigns.

8. For the NPF case studies (Figs. 7-9) it would be useful to include horizontal track of the aircraft colored by the 3-10 nm number concentration. It was mentioned that several vertical profiles were made during each flight, it would be good to see all of them. Also inlcude the number size distribution as a function of time measured at the surface for further comparison.

Response: We have added the tracks (including both the horizontal and vertical ones) of the G-1 aircraft during these three flights, and the tracks are colored by the ratio between $N_{>3}$ and $N_{>10}$ (**Figs. S9** to **S11**). We are showing **Fig. S9** below as an example. We chose to plot the ratio ($N_{>3}/N_{>10}$) because there were periods where the difference in the concentration was small, but the aerosol population was dominated by $N_{3\text{-}10}$. Using $N_{>3}/N_{>10}$ could show the scale and influence of the NPF events. Together with the flight tracks, we also included the surface measurements of aerosol size distributions for comparison against the flight measurements (**Figs. S9** to **S11**). Since the SMPS data corresponding to the case in **Fig. 9** is not available, we showed the normalized size distribution data measured by the UHSAS. From the size distributions, we found that the flight measurements near the surface had a general agreement with the surface measurements. We further discussed these figures when introducing the NPF cases in Section 3.3.1. Please also see the response to comment #10.

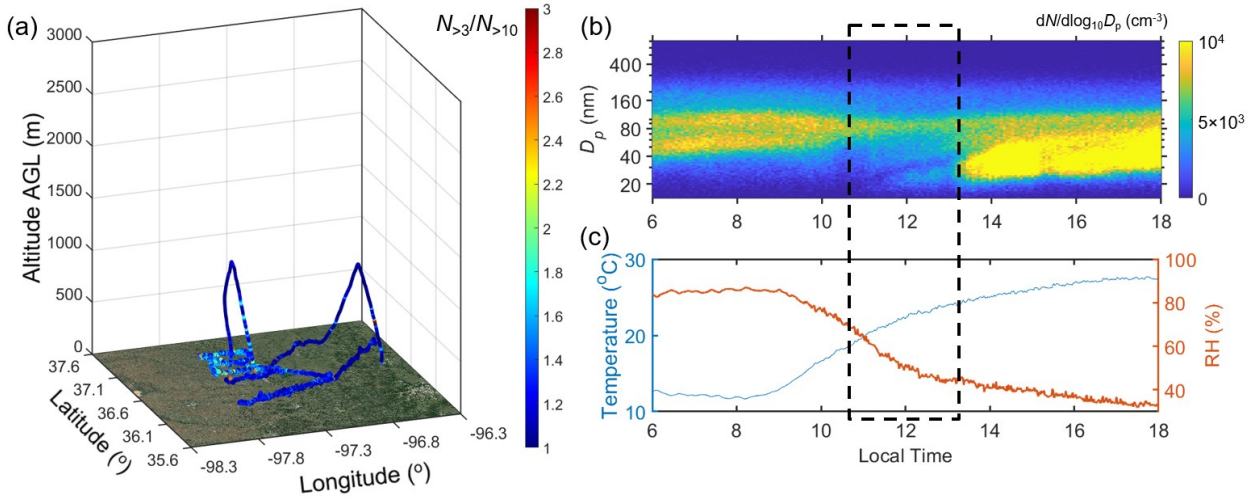


Fig. S9. (1) Track of the G-1 aircraft on September 11, 2016 colored by the ratio between $N_{>3}$ and $N_{>10}$. Map is obtained from ©Google Maps. (2) Aerosol size distribution and (3) temperature and relative humidity (RH) measured at the SGP observatory on September 11, 2016. The dashed box shows the corresponding time for the aircraft measurement.

9. Can you be sure that the observed heterogeneity in the vertical profiles is not actually due to horizontal heterogeneity? This is especially relevant in the case studies. If horizontal heterogeneity cannot be ruled out you should mention it in the text.

Response: The horizontal heterogeneity could play a role in the analysis, as indicated in Fast et al. (2022), which characterized the subgrid variability of aerosol properties using grid spacings of 3, 9, 27, and 81 km. However, as indicated by that study, the averaged aircraft-measured aerosol properties within the 81 km cell were similar to many aerosol properties measured at the SGP observatory, suggesting that averaging aircraft measurements over large spatial scales could reduce the influence of horizontal heterogeneity on aerosol properties. Since this study analyzed the averaged aerosol properties collected during the entire campaign, we believe that horizontal heterogeneity is reduced. However, we could not rule out the influence of horizontal

heterogeneity when analyzing the vertical profiles in a case-by-case manner. This discussion is now added to the Conclusion as:

> "We should note that the horizontal heterogeneity could influence the analysis in this study, as indicated in Fast et al. (2022), which characterized the subgrid variability of aerosol properties using grid spacings of 3, 9, 27, and 81 km. However, as indicated by that study, the averaged aircraft-measured aerosol properties within the 81 km cell were similar to many aerosol properties measured at the SGP observatory, suggesting that averaging aircraft measurements over large spatial scales could reduce the influence of horizontal heterogeneity on aerosol properties. Since this study analyzed the averaged aerosol properties collected during the entire campaign (which covers a large special scale), we believe that horizontal heterogeneity is reduced. However, we could not rule out the influence of horizontal heterogeneity when analyzing the vertical profiles on a case-by-case manner. Nonetheless, analyzing the vertical profiles of aerosols could significantly improve our understanding of the sources and sinks of aerosols in the atmosphere and greatly compliments ground observations."

10. Fig 9: If the particles were formed in the upper parts of the well-mixed layer you would expect them to be more vertically mixed since it takes several hours for the particles to grow to such sizes. Do you see at the surface when the layer was mixed down?

Response: We presume that the reviewer is referring to **Fig. 8**, as it is the figure describing the NPF in the upper BL. It is a good idea to consider the time needed for the growth of the newly formed particles. The NPF layer in the upper BL is indeed relatively well mixed, as indicated by the similar magnitudes of $N_{3\text{-}10}$ between 600 and 1000 m AGL in **Fig. 9b**. The surface measurements related to the example NPF events are now included in **Fig. S10**. As shown in **Fig. S10**, the SGP observatory did not observe the formation of particles below 30 nm. Instead, particles of ~30 nm appeared at 16:00 local time (flight measurement was conducted between 12:40 and 15:30 local time), suggesting that 30 nm particles were transported vertically due to the mixing of the upper BL air to the surface. We have added this discussion in the manuscript as:

> "The spatial scale of this NPF event is shown in **Fig. S10a**, where NPF occurred in the upper BL, while $N_{>3}/N_{>10}$ near the surface is relatively low. This is indicated by the surface measurements of aerosol size distributions, which did not observe the formation of particles below 30 nm. Instead, particles of ~30 nm appeared at 16:00 local time (flight measurement was conducted between 12:40 and 15:30 local time), suggesting that 30 nm particles were transported vertically due to the mixing of the upper BL air to the surface."

11. Lines 339-441: Another explanation might be that the particles were formed in the residual layer and mixed down (Lampilahti et al., 2021). Precursor gases could be present in the residual layer as well (Beck et al., 2022).

REFERENCES:

Lampilahti et al. (2021) https://doi.org/10.5194/acp-21-7901-2021

Beck et al. (2022) https://doi.org/10.5194/acp-22-8547-2022

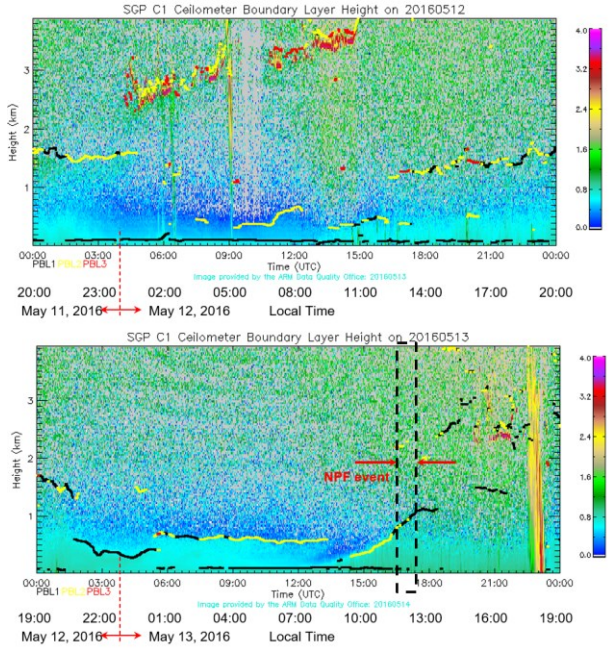
Response: We thank the reviewer for providing these references. It is indeed probable that NPF occurred in the residual layer, and the newly formed particles were further mixed down to the BL. However, due to the narrow vertical range of the residual layer above the BL, the current

aircraft cannot segregate the aerosol properties inside of this layer from the BL. We have included this discussion as:

> "It may be because of the following reasons: (1) the low S_{tot} plays a dominating role in promoting NPF, and a SO_2 mixing ratio of 0.5 ppb is already sufficient for initiating NPF; (2) NPF events occurred in the residual layer above the BL and the newly formed particles were further mixed down to the upper BL (Lampilahti et al., 2021; Beck et al., 2022), where the SO_2 in the upper BL is not related to the NPF events; (3) SO_2 was depleted in the upper BL due to its oxidation to sulfate that promoted the NPF event in the upper BL; (4) different nucleating precursors, such as amines and extremely low volatility organic compounds (ELVOCs), were involved in this NPF event."

12. Fig 10: S_tot and number size distribution show dicontinuities between 1300-1500 m and between 2000-2200 m. Gradient in potential temperature also slightly increases around or before these altitudes indicating weak inversions. Is it possible that the BL reached 2 km on a previous day and the sub-10 nm particles are in fact inside the residual layer? Cases in Figs. 9-10 might both be residual layer NPF events observed before (Fig 10) and after (Fig 9) entrainment into the well-mixed layer.

Response: Based on the description, we presume that the reviewer is discussing the event in **Fig. 9** instead of **Fig. 10**. This is an interesting point. To address this comment, we examined the BL height on the previous day and during the day of the event shown in **Fig. 9** (NPF in FT). The figure below shows the BL height determined by the ceilometer. As can be seen from the figure, the BL height for May 12, 2016 (the day before the NPF event shown in **Fig. 9**) reached around 1700 m, and a nocturnal boundary layer was developed overnight. On May 13, 2016, the NPF event was observed on G-1 between 12:00 and 13:00 local time at the altitudes between 1800 and 2200 m. Therefore, this NPF event is more likely an event in the lower FT and not influenced by the residual layer. Moreover, if the newly formed particles are entrained from the residual layer to the upper BL, these newly formed particles should be observed in a wider altitude range in **Fig. S11**.



However, we cannot completely rule out the impact from the residual layer NPF as the BL height (1700 m) on May 12, 2016 is close to the NPF region (1800 to 2200 m) on May 13, 2016. This discussion has been added to the manuscript as follows:

> "We should note that it is also possible that these newly formed particles may be generated from NPF in the residual layer developed overnight (a mechanism similar to the discussion regarding NPF in the upper BL), as the BL height reached 1700 m on the previous day before the development of the nocturnal boundary layer. If so, the NPF cases shown in **Figs. 8** and **9** may be both occurring in the residual layer but observed before (**Fig. 9**) and after (**Fig. 8**) entrained into the BL. Given these NPF events occur in the higher altitudes and there are limited observations regarding NPF in these locations, the mechanisms associated with NPF events require more investigation."

12. Lines 429-430: Show the ground-based number-size distribution measurements. Does it support the analysis of what happens between the morning and the afternoon flights in the well-mixed part of the BL.

Response: We have now included the size distributions measured at the SGP observatory as **Fig. S12** (also shown below). The ground observation agrees with the measurements conducted in the morning and afternoon flights, where the size of the accumulation mode aerosols increased, and at the same time, aerosols below 30 nm emerged. We have added the following discussion in Section 3.4.1:

> "Surface measurements conducted on the same day agreed in general with the flight observations (**Fig. S12**), showing a gradual growth in the size of the accumulation-mode aerosols during the day and the emergence of aerosols below 30 nm between 14:00 and 15:00 local time."

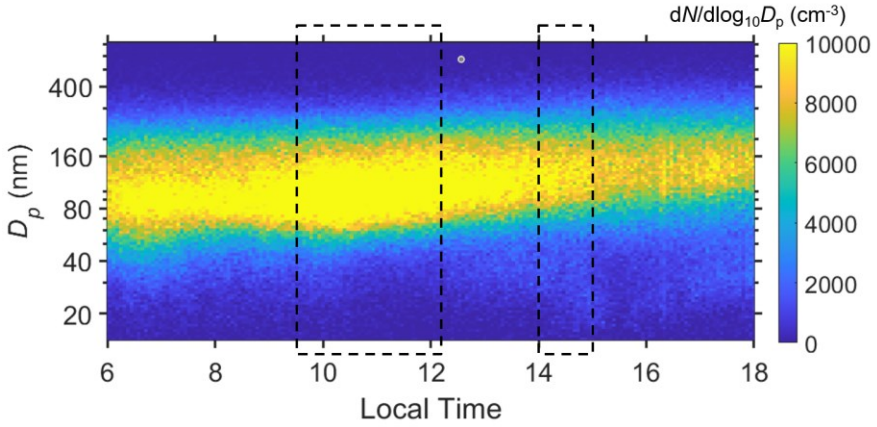


Fig. S12. Aerosol size distribution measured at the SGP observatory on September 4, 2016. The black dashed boxes correspond to morning and afternoon flight times.

13. Line 467: "FL" -> "FT"

Response: Corrected. Thank you.

14. Fig 13: Show the time evolution of the number concentrations in the relevant size ranges (N_<50 and N_>100) at the surface.

Response: We have now included the concentrations of aerosols above 100 nm ($N_{>100}$) and below 80 nm ($N_{<80}$) during this event (**Fig. 13d**, also shown below). We chose 80 nm as the size

cut because the Aitken mode had an upper size boundary of ~ 80 nm during the MCS event. We added the discussion:

> "We further calculated the concentrations of aerosols above 100 nm ($N_{>100}$) and below 80 nm ($N_{<80}$) during this event. There was an increase in $N_{<80}$ between 21:10 and 23:00 (increased from ~ 600 to ~1000 cm^{-3}, **Fig. 13d**, while $N_{>100}$) remained relatively constant during this period. This change in aerosol properties may be explained by the horizontal transport of air mass, but at the same time, the vertical transport of FT air to the BL may also contribute to the change in aerosol properties."

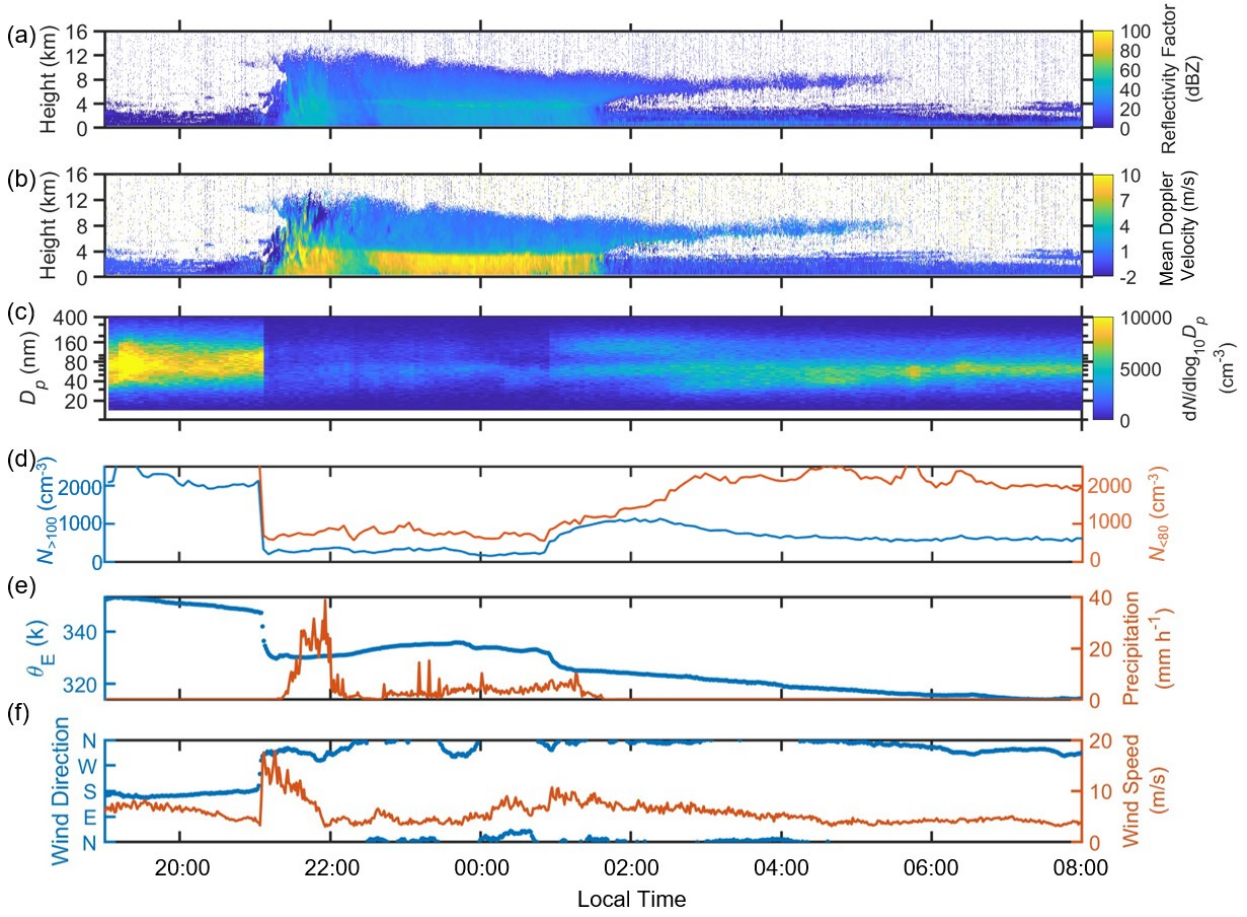


Fig. 13. Ground measurements of aerosol, cloud, and meteorological properties during the convective downward transport event from September 9 to 10, 2016. (a) Cloud reflectivity measured by RWP. (b) Mean Doppler velocities measured by the radar wind profiler (RWP). A positive value indicates downward motion. (c) Aerosol size distributions measured by a scanning mobility particle sizer (SMPS). (d) Concentration of aerosols with sizes above 100 nm ($N_{>100}$) and below 80 nm ($N_{<80}$). (e) Equivalent potential temperature and precipitation rate measured by the weather station. (f) Wind direction and wind speed measured by the weather station.

15. Line 525: Does the calculated particle exchange agree with the ground-based number concentration measurements during the convective event?

Response: Thank you for the suggestion. We calculated the increase of surface-measured aerosol concentration due to the downdraft, assuming that the BL is well mixed and there are no other sources or sinks of aerosols. This can be done via dividing the total exchange by the BL

height (assumed to be 1 km), which yields an increase in the aerosol concentration of 450 cm^{-3}. Note that this value is similar to that observed in the increase of $N_{<80}$ between 21:30 and 23:00 (from ~600 to ~1000 cm^{-3}), suggesting that the increase of the Aitken-mode aerosols during MCS may be related to the convective downdraft. We have added the following discussion to Section 3.4.3:

> "Assuming that the BL is well mixed and there are no other sources or sinks of aerosols, we could estimate the impact of this event to the total aerosol concentration measured at the surface. This can be done via dividing the total exchange (4.50×10^7 cm^{-2}) by the BL height (assumed to be 1 km), which yields an increase of the aerosol concentration of 450 cm^{-3}. Note that this value is similar to that observed in the increase of $N_{<80}$ between 21:30 and 23:00 (from ~600 to ~1000 cm^{-3}), suggesting that the increase of the Aitken-mode aerosols during MCS may be related to the convective downdraft."

We thank the reviewer for his/her comments. This has improved the quality of the manuscript and we look forward to this manuscript being accepted.