Reply to reviewer #3 of *"Sensitivity of warm clouds to large particles in measured marine aerosol size distributions – a theoretical study"*

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We are grateful for the time and effort the reviewer invested in our work, and highly appreciate all of the constructive comments that helped us improve the paper. Below we address all the reviewer's comments point by point (our answers are marked in blue).

The influence of aerosol size distribution and chemical composition on precipitation formation and intensity is still a challenging question to answer, due primarily to the sophisticated microphysical processes dealing with particles with a wide range of sizes, and also to the interplays between dynamics and microphysics. In this study, the authors choose to focus on addressing aerosol-precipitation response in a warm cloud, using a detailed bin microphysical framework for both aerosols and cloud droplets while a somewhat simplified dynamical framework (an axisymmetric model). In addition, they have also assumed a uniform chemical composition for the included aerosol population (sea salt) to limit the aerosol activation in a one-dimensional (size) parametric space. In order to address the targeted issue more realistically, they have also adopted measured aerosol size distributions collected from locations with different atmospheric backgrounds.

An interesting finding of this study is the significant difference in aerosol-precipitation responses between a case with the so-called *Atlantic-1* aerosol profile with ultra large CCNs and cases with other measured aerosol profiles without evident fraction of such giant CCNs. With a careful design of their modeling simulations, the authors have been able to define the criterion size of large aerosol particles that can create significant impacts on precipitation. Overall speaking, the paper has been relatively well organized, the research findings are well presented, and conclusions are drawn with solid science evidence.

We thank the reviewer for the careful reading of our manuscript and the constructive remarks.

1) A clear missing information in the manuscript is the cloud droplets concentrations, especially the vertical profiles of number concentration of cloud droplets and raindrops.

… In addition, Fig. 2(d) presents a rather interesting feature in high concentration simulations using all the distributions except *Atlantic-1* where collision-coalescence overwhelmed the condensation growth in a relatively early stage. However, without information of vertical distributions of cloud mass, the reader would have problem to understand (1) why the collision-coalescence increases with time but in a rather slow pace comparing to the case of *Atlantic-1*, and (2) the depths of layer where cloud mass grew in various cases. Note that large droplets (i.e., raindrops) can still be moved upward by updraft and both condensation and collision-coalescence can proceed in either updraft or downdraft (as far as the parcel remains saturated). Therefore, knowledge of the vertical growth tracks of precipitating particles is critical to understand how the two major growing processes evolved.

Author reply: We thank the reviewer for these important comments. Here we address the reviewer's comments regarding the vertical profiles of number concentration of cloud droplets and raindrops, as well as the vertical distributions of cloud mass.

Following the reviewer's suggestion and to better explain our results, we added Text S7 and Figure S7 to the SI (see below) showing the time-height horizontal mean profiles of: cloud mass mixing ratio, droplet number concentration (N_d), vertical velocity (w), and relative humidity (RH) below the cloud base, for the *Atlantic—1* and *Pacific—6* MSDs, normalized to an aerosol concentration (N_{tot}) of 2629 cm^{-3}. We also added Text S8 and Figure S8 to the SI (see below as well) showing the time-height evolution of the horizontal mean profiles of the number concentration (N_r) and mass mixing ratio (M_r) of precipitating particles ($D_p > 80\ \mu m$) for the *Atlantic-1* and *Pacific-6* MSDs, normalized to $N_{tot} = 2629\ cm^{-3}$.

We chose to show the *Pacific—6* MSD results as a representative example since it represents well all the other four MSDs cases. As can be seen in Fig. R1 below, that shows the time-height evolution of the horizontal mean profiles of cloud mass mixing ratio for all the MSDs for $N_{tot} = 2629\ cm^{-3}$, there are only minor differences among the four clouds.

We added an explanation regarding this issue to the *Results* (**section 3, L169—L181):** "Under more polluted conditions, the trajectory of the *Atlantic—1* MSD cloud (black curve in Fig. 2d) on this phase space is similar (in shape) to the one in the cleaner case (black curve in Fig. 2c), but this cloud accumulates more mass, due to the larger droplet surface area (Fig. S5b—c). **However, the *Atlantic—1's* total droplet surface area is lower in comparison to the rest of the clouds (see Fig. S5c), and still, it condenses more mass, reaching ~11×10^7 *g* compared to ~8×10^7 *g* as the rest of the clouds. This can be explained by the nucleation of the GCCN and UGCCN that are present in the *Atlantic—1* MSD under polluted conditions, which on the one hand accumulate more mass (Fig. 2d) and drive a significantly higher number of raindrops at the growing stage of the cloud (Fig. S8), and on the other hand, results in a lower droplet number concentration (N_d) compared to the other clouds (Fig. S7). Therefore, the vertical distribution of mass of the *Atlantic—1* cloud is dominated by the precipitating particles, unlike the other clouds (Figs. S7 and S8). Note that while the total cloud mass of the *Atlantic—1* is larger than the one obtained by the other clouds, it is in the same order of magnitude. However, the mass of precipitating particles in the *Atlantic—1* cloud overwhelms the ones exhibited by the other MSDs. Contrastingly, the N_d of the other clouds is much higher than the one of the *Atlantic—1* cloud, allowing for collision-coalescence to begin toward the end of the condensational growth stage, or after the evaporation process has begun (e.g., *Pacific—4*, the trajectories turn back to the left before acquiring a vertical component), and to increase in a slow pace. For the *Atlantic—1* cloud, the accumulation of liquid water by nucleation and condensation occurs in parallel to the collection process that starts much earlier in this case (Fig. 2d).**"

We added ***Text S7. Time—height Diagrams of Cloud Mean properties*** to the SI **(L67—L76)**: "**To understand the vertical distribution of some of the cloud's key properties, we show the time evolution of the cloud mass mixing ratio, droplet number concentration (*N_d*), vertical velocity (*w*), and relative humidity (*RH*) below the cloud base for the *Atlantic—1* and the *Pacific—6* MSDs normalized to *N_{tot} = 2629 cm^{-3}*. We show the *Pacific—6* MSD case as a representative example to the other four MSD cases, since their results are very similar. It is clear that while the *Atlantic—1*'s mass is the same order of magnitude as the one of the *Pacific—6*, the total *N_d* is considerably smaller for the *Atlantic—1* cloud, and that the droplets are confined to the lower part of the cloud. These are big droplets that nucleated on the GCCN and the UGCCN in the *Atlantic—1* MSD. These droplets sediment out almost immediately after their formation, thus are not carried to higher levels in the cloud. The *Atlantic—1* starts to precipitate earlier than the other clouds (as discussed in the main text), while the cloud is still in its developing stage, updrafts prevail and the sub—cloud layer features low *RH* values**."

We added ***Text S8. Time—height Diagrams of Precipitating Particle's Growth*** to the SI **(L77—L84): "For clarifying the reasons behind the reduced surface rain amounts that are observed in the *Atlantic—1* MSD case, Fig. S8 shows the time-height evolution of the horizontal mean profiles of raindrops number concentration *N_r (D_p > 80 μm)*, and mass mixing ratio *M_r (D_p > 80 μm),* for the *Atlantic—1* and the *Pacific—6* MSDs, normalized to *N_{tot} = 2629 cm^{-3}*. The formation of raindrops is observed at a very early stage of the *Atlantic—1* cloud lifetime, compared to the timing of the rain formation in the *Pacific—6* case. In addition, it is clear that the high *N_r* around the *Atlantic—1*'s cloud base contains very little mass, but the drops are big enough to fall out. However, since the drops are small, and the sub—cloud layer is still dominated by updrafts (Fig. S7e—f) the majority of them evaporate before reaching the surface (efficient evaporation and longer fall time)."**

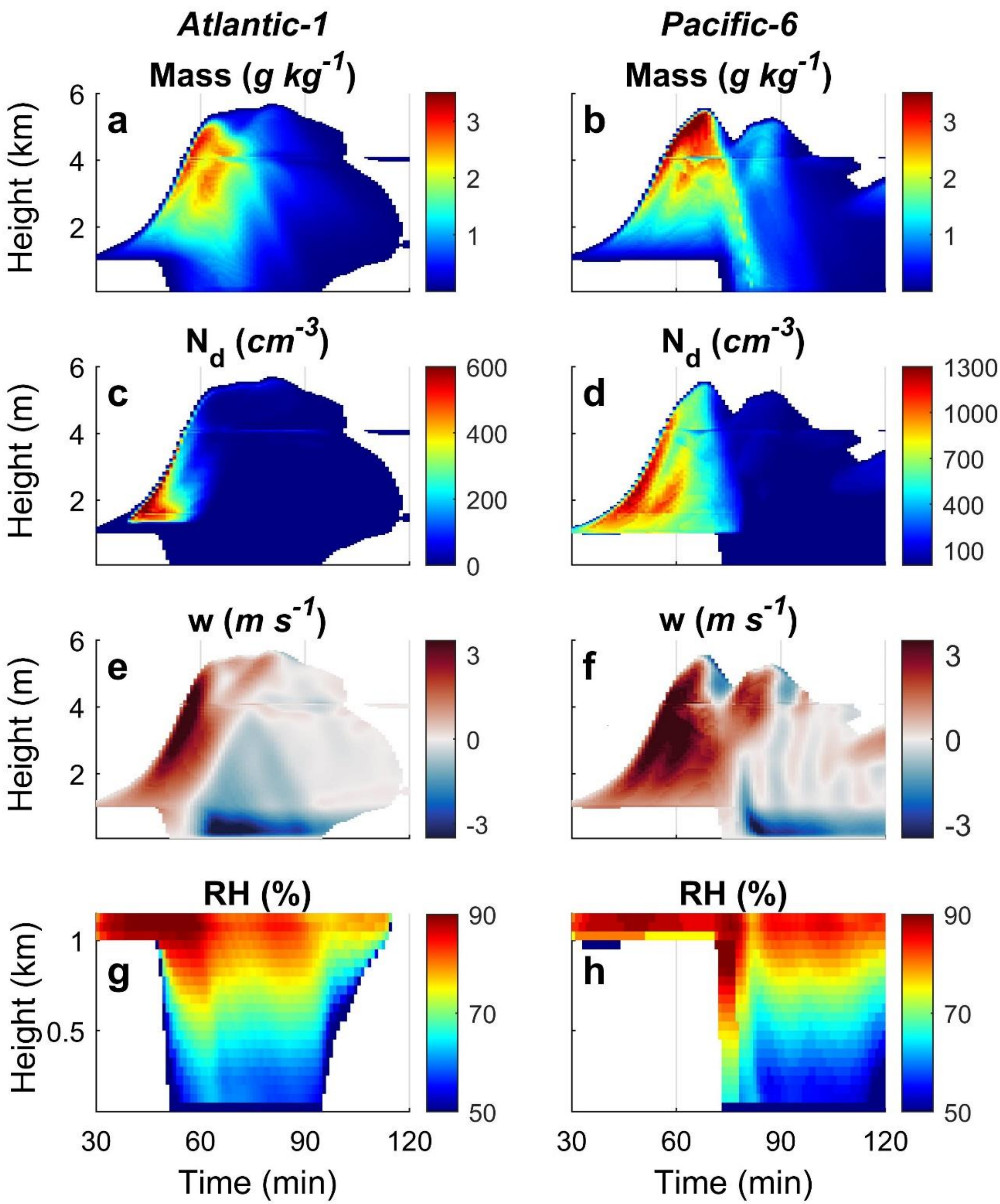

*Figure S7. Time-height diagram of the horizontal mean of **(a, b)** cloud mass mixing ratio (g kg^{-1}), **(c, d)** droplet number concentration (N_d, cm^{-3}), **(e, f)** vertical velocity (w, m s^{-1}), and **(g, h)** relative humidity (RH, %) below the cloud base, for the Atlantic—1 (left column) and Pacific—6 (right column) MSDs normalized to N_{tot} = 2629 cm^{-3}. Values are shown only for the cloudy (and rainy) pixels (mixing ratio > 10^{-3} g kg^{-1}). Note the different scales for the color bars in panels **(c)** and **(d)**.*

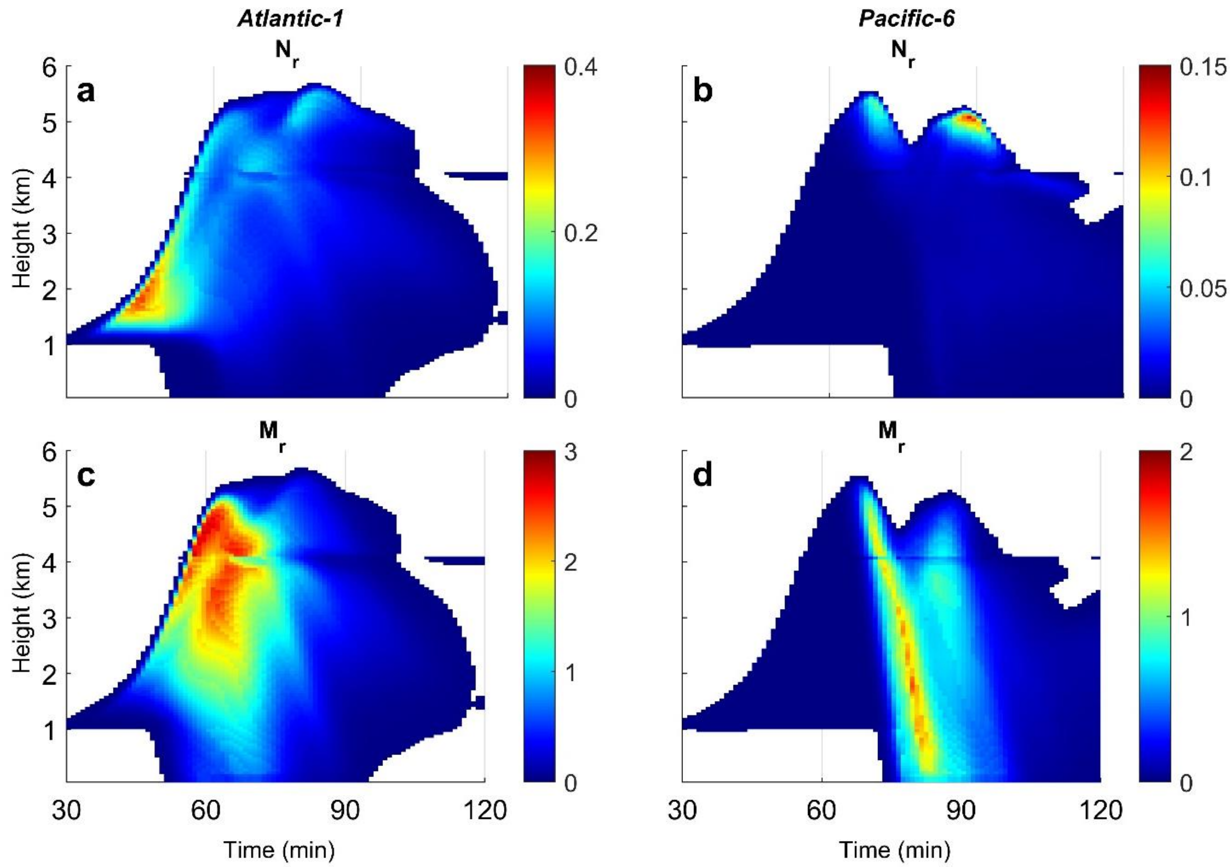

Figure S8. Time—height diagram of the horizontal mean of raindrops (D_p > 80 μm) ***(a, b)*** *number concentration (N_r, cm^{-3}) and* ***(c, d)*** *mass mixing ratio (M_r, g kg^{-1}), for the Atlantic—1 (left column) and Pacific—6 (right column) MSDs normalized to N_{tot} = 2629 cm^{-3}. Values are shown only for the cloudy (and rainy) pixels (mixing ratio > 10^{-3} g kg^{-1}). Note that the color bars have different limits for the Atlantic—1 and Pacific—6 clouds.*

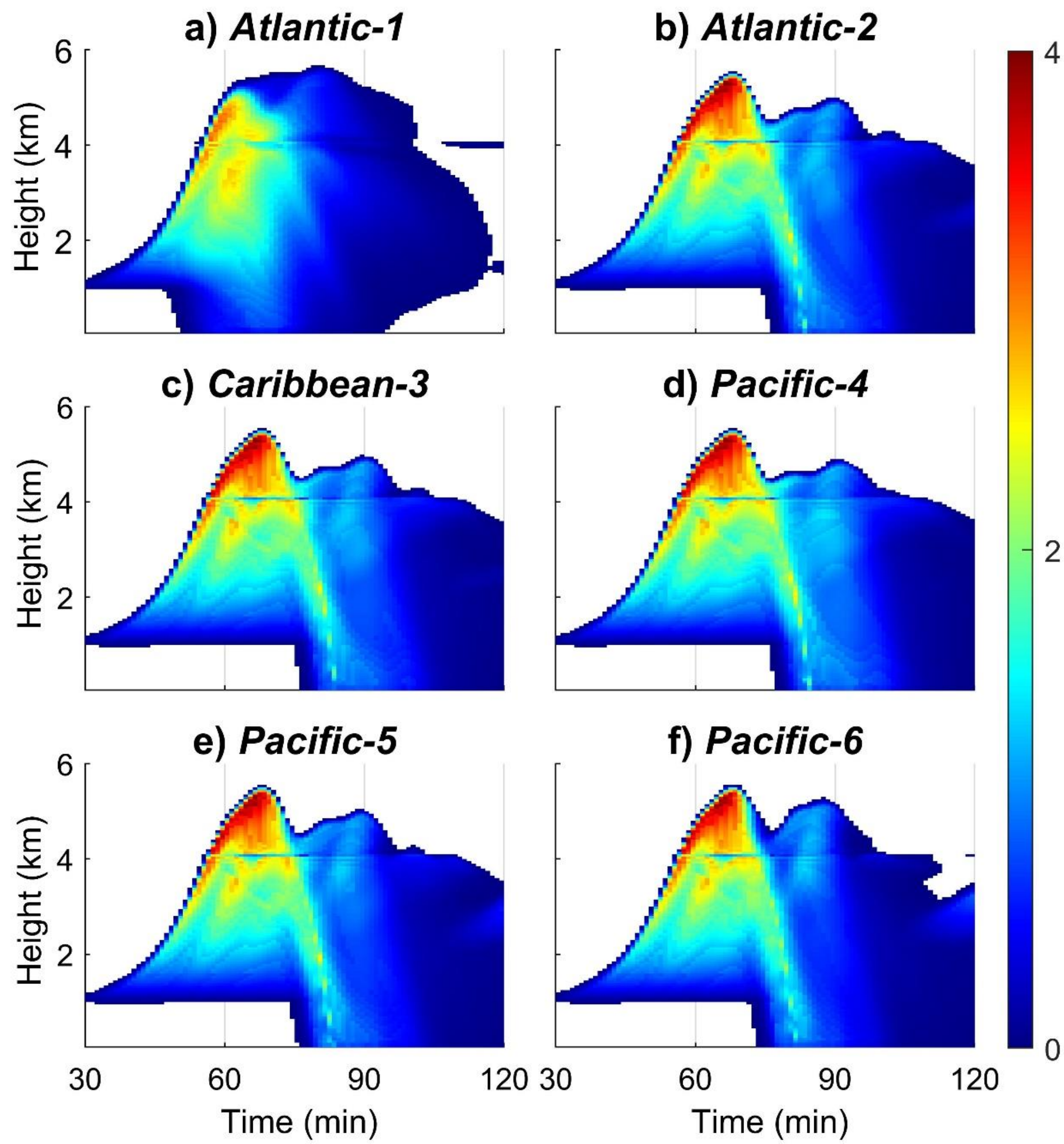

Figure R1. Time-height diagram of the horizontal mean profiles of cloud mass mixing ratio (g kg^{-1}) for all MSDs for an aerosol concentration of 2629 cm^{-3}. There are only minor changes among all MSD's besides the Atlantic—1. We, therefore, took Pacific—6 as a representative MSD.

2) The authors have discussed the correlation between sub-cloud evaporation and rainfall at surface. With a knowledge of sub-cloud raindrop population including total number and size distribution this would be much easier to understand.

Author reply: Thank you for this comment. The revised Fig. S6 (see below) shows the droplet size distribution below the cloud base at the time of maximum surface rain rate for the six different MSDs normalized to N_{tot} = 2629 cm^{-3}. The total number concentration of droplets was added for the different curves. The figure clearly shows that the *Atlantic—1* MSD has more small raindrops and less big raindrops than the other MSDs.

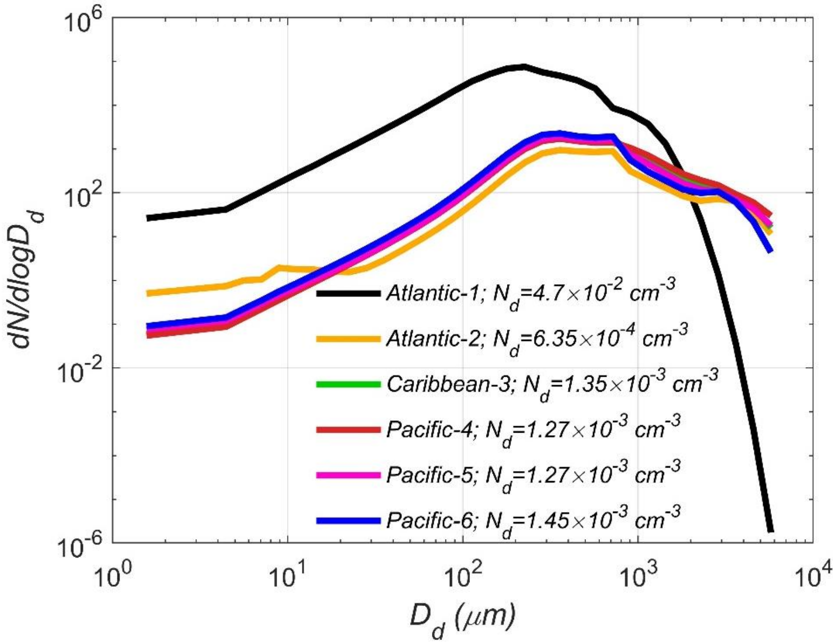

Figure S6. Droplet size distribution below the cloud base at the time of maximum surface rain rate for the six different MSDs normalized to N_{tot} = 2629 cm^{-3}. The total droplet number concentration (N_d, cm^{-3}) is noted in the legend for each MSD.

3) It is understood that the authors wanted to focus on the aerosol and cloud microphysical connections. Nevertheless, the feedback of dynamics, even in a rather simplified dynamical framework still plays a role in determining the growth of precipitating particles. The authors mentioned very briefly about cold downdraft and also analyzed sub-cloud evaporation. Perhaps a more in-depth analysis would provide a better understanding of the role of dynamical feedback in, e.g., leading to the results presented in Fig. 3 and 4.

Author reply: We thank the reviewer for highlighting this important point. As described in answer no. 1 above, we are now showing the vertical velocity in the revised version of the SI in Fig. S7e—f. As the reviewer stated, the dynamics is strongly coupled to the microphysics, and hence it has a crucial effect on the growth of precipitating particles and now it is presented in the paper to create a full picture. The early fall-out of the smaller *Atlantic—1* rain drops, while the cloud is still in its developing stage, results in the *Atlantic—1* raindrops falling through updrafts that dominate the cloud and the sub-cloud layer at this stage (Fig. S7g—h). This causes a longer fall and together with the fact that the *Atlantic—1* sub—cloud layer is drier, promotes the efficient evaporation of the already smaller raindrops of the *Atlantic—1* cloud. This results in the reduced surface rain as shown in Figs. 3 and 4 of the main text.

To clarify this point in the main text, we added the following paragraph to the *Results* **(section 3, L200—L205)**: "The *Atlantic—1* raindrops are considerably smaller than those produced by the other clouds (Fig. S6; see below), and their evaporation is, therefore, **more efficient**. **Moreover, the rain falls below the cloud base earlier, compared to the other MSDs cases, while the cloud is still in its developing stage, meaning that the cloud and the sub—cloud layers are dominated by updrafts, and the sub—cloud**

layer is consequently drier (see Fig. S7). The combination of the small raindrops with their early fall out that last longer (due to the updrafts prevailing at this stage), results in greater rain evaporation below the cloud base for the *Atlantic—1* MSD."

We also added the following to the *Summary* **(section 4, L272—L275):** "This results in **the** fast formation of large drops and thus, an early fall-out of drizzle, **while the cloud is still in its developing stage, updrafts prevail and the sub—cloud layer is drier. The combination of a sub—cloud layer that is dominated by updrafts and features lower *RH* values, further promotes longer fall time for the small rain drops and an efficient evaporation below the *Atlantic—1* cloud base."**

Some minor comments.

4) Page 5, Figure 2(a) and (b): it would be helpful to provide the integration length of each simulation shown in these two figure panels in the figure caption.

Author reply: We added the integration length of the simulation (150 *min*) to the revised Fig. 2 caption.

5) Page 5, Ln 112: I understand the purpose of normalizing every distribution to match a given total concentration is for the convenience to identify the role of certain characteristics of size distributions such as shape in influencing the formation of precipitation. However, it is expected that the shift of the distributions to meet often much higher concentrations would increase the number of GCCN or even UGCCN. Could the authors provide such numbers even in the supplementary materials as a table or so? In addition, I don't remember this has been discussed in the manuscript, e.g., why the increase of GCCN still had no effect on the overall rain formation and growth for all cases including Atlantic-2 other than Atlantic-1.

Author reply: We thank the reviewer for this comment. As we normalize the MSDs to higher aerosol concentrations, we indeed shift the distributions such that they contain bigger particles (see Fig. S1 below). However, most of the MSDs (all except from the *Atlantic—1* and *Atlantic—2*) did not contain any GCCN or UGCCN even in the case of the highest N_{tot}. In the paper we define a physical threshold for GCCN (D_p~$5\ \mu m$) and UGCCN (D_p~$14\ \mu m$). These thresholds are marked on the revised panels of Fig. S1, for each N_{tot}. The *Atlantic—1* and the *Atlantic—2* are the only MSDs that contain GCCN, and the *Atlantic—1* is the only MSD that contains UGCCN. Even though the *Atlantic—2* contains some GCCN, they are present at low concentrations ($1.6x10^{-6}$—$7.5x10^{-5}\ cm^{-3}$, for the lowest and highest N_{tot}, respectively). These concentrations of GCCN are low in comparison to e.g., the GCCN concentration of the *Atlantic—1* ($1.9x10^{-5}$—$5.7x10^{-4}\ cm^{-3}$), and are not sufficient to cause any effect on the *Atlantic—2* rain formation and growth.

We added a clarification regarding this matter to *Methods* **(section 2.2., L109—111):** "**As we normalized the MSDs to higher values of N_{tot}, the MSDs shifted such that they contained larger particles (Fig. S1). However, only the *Atlantic—1* and *Atlantic—2* MSDs contained GCCN, and the *Atlantic—1* was the only MSD that contained UGCCN even in the case of the highest aerosol concentration.**"

We also extended ***Text S1. Normalized MSDs*** to include the following paragraph **(L17—L21):** "**The *Atlantic—1* and the *Atlantic—2* are the only MSDs that contained GCCN, and the *Atlantic—1* is the only MSD that contained UGCCN. Note that even though the *Atlantic*—2 contained some GCCN, they were present at low concentrations (*$1.6x10^{-6}$—$7.5x10^{-5}$ cm^{-3}*, for the lowest and highest N_{tot}, respectively). These concentrations of GCCN are low in comparison to e.g., the GCCN concentration of the *Atlantic—1* (*$1.9x10^{-5}$—$5.7x10^{-4}$ cm^{-3}*), and are not sufficient to cause any effect on the *Atlantic—2* rain formation and growth.**"

6) Page 7, Ln 152: "bigger droplets resulted in a lower total droplets' surface area. . .", the sentence is somewhat ambiguous since such a result is not obvious, an explanation would be helpful here.

Author reply: We thank the reviewer for this comment.

We clarified this in the revised *Results* **(section 3, L161—L164):** "The bigger droplets **formed by the nucleation of GCCN** resulted in a lower droplets' surface area **for a given total water mass (compared to the one that would have formed from droplets nucleated on smaller CCNs)** for the *Atlantic-1* MSD case and also compared to the other MSDs (Fig. S5). **The lower total droplets' surface area** was then further reduced by the early initiation of the collection process."

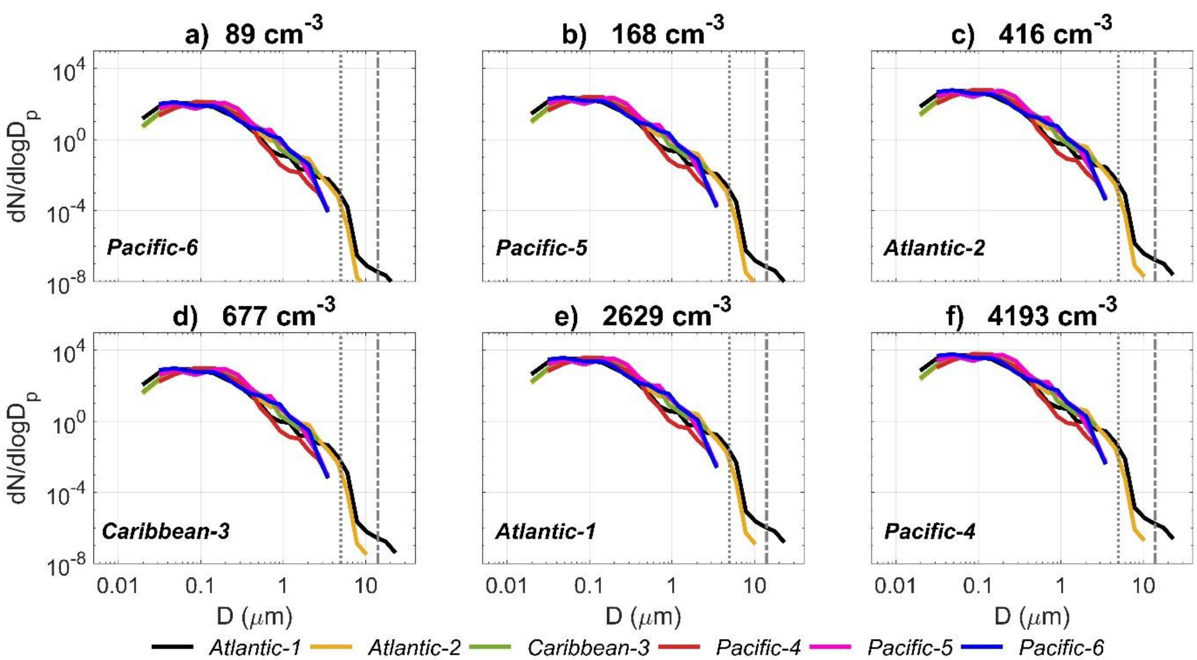

*Figure S1: All of the MSDs used in the model. Each panel shows the MSDs normalized to the specific total aerosol concentration of the MSD noted in the lower left corner. The panels are organized from clean **(a)** to polluted **(f)** conditions.* ***Dotted and dash-dotted verticals line indicated the threshold for GCCN (D_p~5 µm) and UGCCN (D_p~14 µm), respectively.***