Reply to reviewer #1 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).

In this paper, an axisymmetric cloud model with detailed bin–microphysics was initialized with six marine aerosol size distributions (MSD), measured in-situ in the Atlantic Ocean, Caribbean Sea and Pacific Ocean to study the effect of aerosol concentration and size distribution on warm clouds' properties. It shows that the cloud mass and precipitation change non-monotonically with the total concentration and size distribution of cloud condensation nuclei (CCN), especially when a tail of giant or ultragiant CCN (GCCN or UGCCN) is also included in the aerosol size distribution. The most interesting finding is the upper boundaries of the GCCN. This has not been reported in previous studies, to the best of my knowledge. The study is well within the scope of ACP and is generally well presented, except for a few places need to be clarified or corrected.

We thank the reviewer for the careful reading of our manuscript and this positive description of our work. We hope that the physical boundaries on GCCN will be helpful for the community.

Specific comments:

1) In the abstract, the statements and explanations are mainly based on the simulation results using the deepest thermodynamic profile, a more generalized statement or results including the shallower clouds should also be included, for a more complete picture.

Authors reply: We thank the reviewer for this important comment that helped us present our study in a more general way. We changed the abstract (and other parts in the paper, as described in answer no. 5) to describe the results of the different profiles.

The revised **abstract** reads: "Aerosol size distribution has major effects on warm cloud processes. Here, we use newly acquired marine aerosol size distributions (MSD), measured *in-situ* over the open ocean during the *Tara* Pacific expedition (2016—2018), to examine how the total aerosol concentration (N_{tot}) and the shape of the MSD change warm clouds' properties. For this, we used a toy-model with detailed bin-microphysics **initialized using three different atmospheric profiles, supporting the formation of shallow to intermediate and deeper warm clouds.** The changes in the MSDs affected the clouds' total mass and surface precipitation. In general, the clouds showed higher sensitivity to changes in N_{tot} than to changes in the MSD's shape, except for the case where the MSD contained giant and ultragiant cloud condensation nuclei (GCCN, UGCCN). For increased N_{tot} **(for the deep and intermediate profiles),** most of the MSDs drove an expected non-monotonic trend of mass and precipitation **(the shallow clouds showed only the decreasing part of the curves with mass and precipitation monotonically decreasing).** The addition of GCCN and UGCCN drastically changed **the non-monotonic trend**, such that surface rain saturated and the mass monotonically increased with N_{tot}. GCCN and UGCCN changed the interplay between the microphysical processes by triggering an early initiation of collision-coalescence. The early fall-out of drizzle in those cases enhanced the evaporation below the cloud base. Testing the sensitivity of rain yield to GCCN and UGCCN revealed an enhancement of surface rain upon the addition of larger particles to the MSD, up to a certain particle size, when the addition of larger particles resulted in rain suppression. This finding suggests a physical lower bound can be defined for the size ranges of GCCN and UGCCN."

2) I suggest each of filled circles in Fig. 2 (c)(d) to be marked with a time. The current figures are a little bit confusing.

Author reply: We thank the reviewer for this comment that helped us make the figure clearer. In the revised Fig. 2 (see below), we marked the filled circles on the black and blue curves, to indicate the simulated time (in *min*).

We also added specific time references to the part of the revised *Results* where Fig. 2 is interpreted to make it easier to follow the trajectories in the figure, **(section 3, L157—L161):** "At a later stage in the cloud's lifetime, the trajectories turn diagonally up **(~56 *min* into the simulation)**, showing that the collection process has begun. Finally, the clouds stop growing by condensation, reaching their maximum mass, and begin to evaporate **(~71 *min* into the simulation;** trajectories turn to the left). In the *Atlantic—1* MSD case, the collection process kicks in earlier, within 10 minutes of the cloud's lifetime **(~51 *min* into the simulation)**, due to the presence of GCCN in the MSD which initially form bigger droplets."

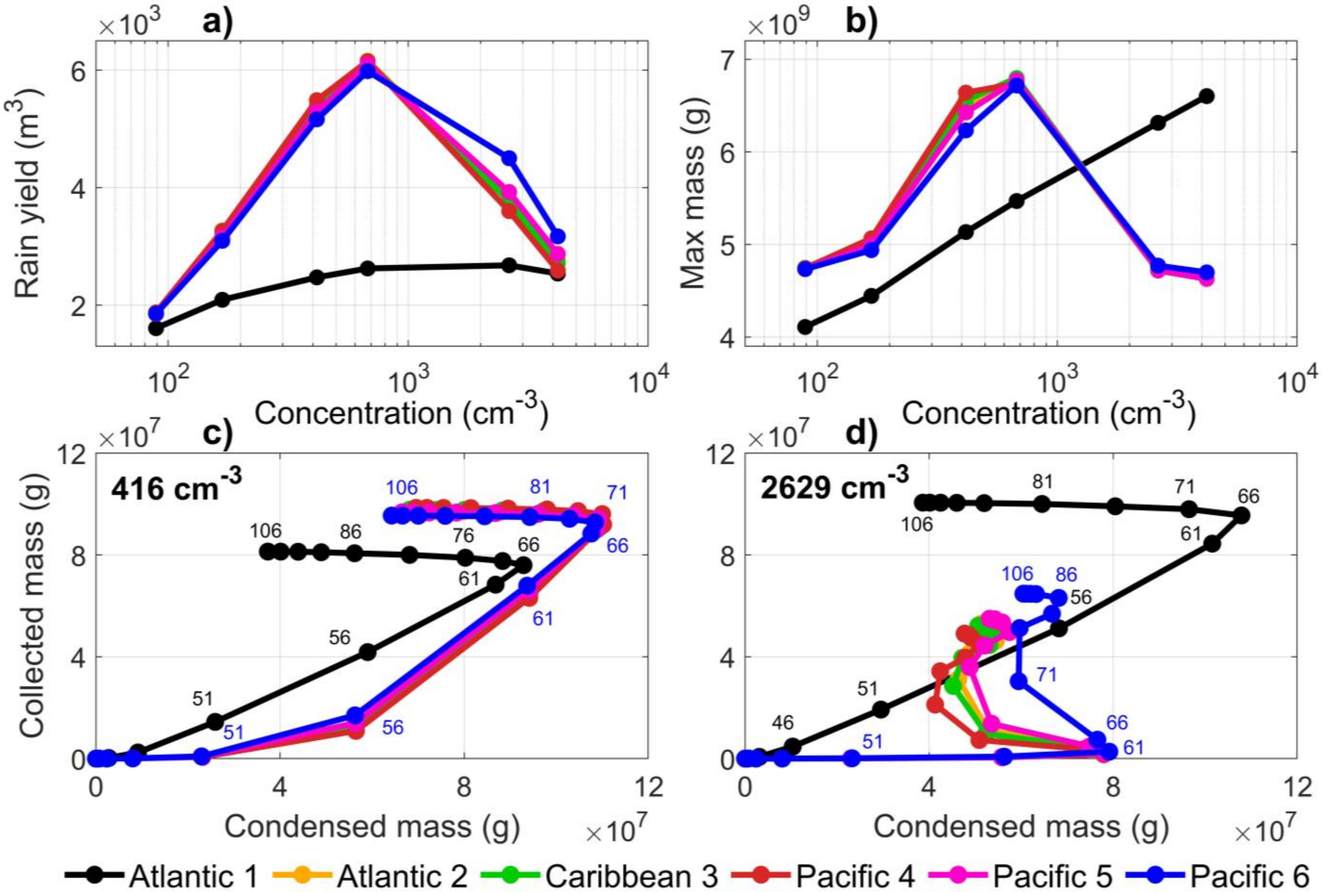

Figure 2. ***(a)*** *Surface rain yield and* ***(b)*** *cloud's maximum mass as a function of* $\mathbf{N_{tot}}$ *used in the simulation****, integrated over 150 minutes of simulations****. Each curve represents six simulations, done with a specific shape of the MSD normalized to different aerosol concentrations. The lower panels* ***(c, d)*** *show the time evolution of accumulated collected mass versus accumulated condensed mass.* ***The simulated time is noted along the black and blue curves for the Atlantic—1 and Pacific—6 MSDs, respectively.*** *The panels represent an aerosol concentration of 416 and 2629 cm*$^{-3}$ *(****c*** *and* ***d****, respectively).*

3) Line 183-184: "The *Atlantic-1* raindrops are considerably smaller than those produced by the other clouds (Fig. S6), and their evaporation is therefore greater": One fact might also be important is that while raindrops formed earlier in case *Atlantic-1*, the cloud is still in its developing stage, or the vertical velocity is still positive below cloud base, and the relative humidity is relatively low, so the raindrops spend more time and therefore evaporate more before reaching the surface. In other cases, rain was promoted by stronger downdrafts, and the relative humidity should also be higher. Therefore, it may provide more evidence to explain the differences in surface rain amount and evaporation mass between case *Atlantic-1* and other cases by analyze the below-cloud vertical velocity and relative humidity.

Author reply: We thank the reviewer for this important comment that allowed us to be more thorough in our explanation. We added to the SI a new figure (Fig. S7, attached below) that shows the time-height evolution of the horizontal mean values of cloud mass mixing ratio (Fig. S7a—b), droplet number concentration (N_d, Fig. S7c—d), vertical velocity (w, Fig. S7e—f), and relative humidity (RH, Fig. S7g—h) for all the cloudy (and rainy) pixels of the *Atlantic—1* and *Pacific—6* MSDs for an aerosol concentration of 2629 cm^{-3}. Note that the *RH* panels show only the sub—cloud layer to address the reviewer's comment. Focusing on the *w* and *RH* panels, they show clearly that the reduced surface rain of the *Atlantic—1* case is indeed due to a combination of the smaller raindrops, and their early fallout while the cloud is still in its developing stage. Therefore, the sub—cloud layer is dominated by updrafts and low *RH* values in comparison to the *Pacific—6* MSD (presented in this figure as a representative of all other MSDs).

We added an explanation 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 (Fig. S7). The combination of the small raindrops with their early fall out that lasts 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 explanations to the *Summary* **(section 4, L265—L268):** "This results in **the** fast formation of large drops and the early fall-out of drizzle **while the cloud is still in its developing stage, such that 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 raindrops and an efficient evaporation below the *Atlantic—1* cloud base."**

4) Figure 4(e) and 4(f) show a little bit strange here and do not add more support to the main body of the text, may be removed?

Author reply: While we agree that these two images are exceptional in this paper, we believe they are a valuable addition. They emphasize the uniqueness of this work that relies on *in-situ* measurements as the initial conditions for the modeled aerosol's MSDs, while also showing the type and size of big aerosol (GCCN and UGCCN) that are present in the marine boundary layer. However, following the reviewer's comment and to better explain them we separated panels e—f from Fig. 4 and put them in a separate new Figure (Fig. 5, see below) in the *Summary* (section 4) of the revised manuscript, where we want to demonstrate which type of GCCN and UGCCN were measured over the open ocean.

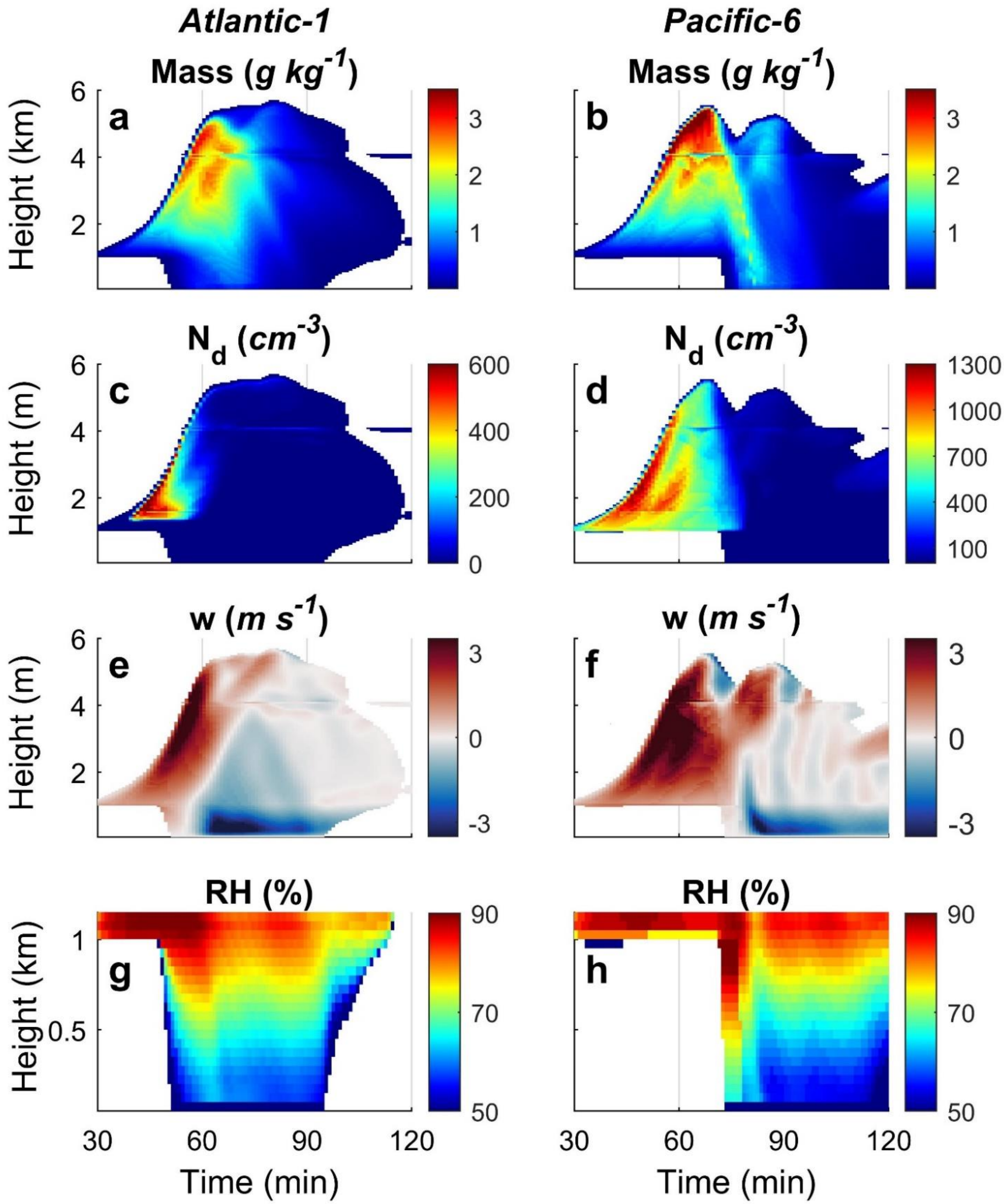

Figure S7. Time-height diagram of the horizontal mean of ***(a, b)*** *cloud mass mixing ratio (g kg^{-1}),* ***(c, d)*** *droplet number concentration (N_d, cm^{-3}),* ***(e, f)*** *vertical velocity (w, m s^{-1}), and* ***(g, h)*** *relative humidity (RH, %) below the cloud base, for the Altantic-1 (left column) and Pacific-6 (right column) MSDs normalized to an aerosol concentration of 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)****.*

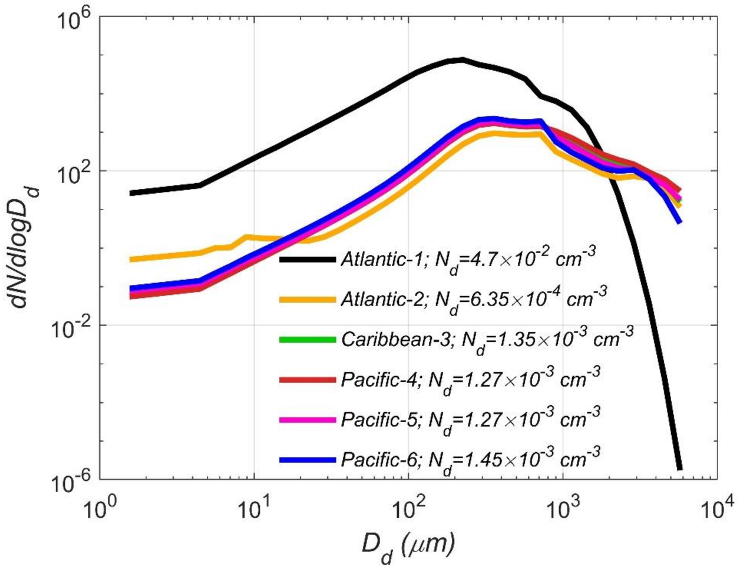

Figure S6. Droplet size distribution below the cloud base at the time of maximum surface rain rate for the six different MSDs normalized to N_{tot} = 2629 cm^{-3}. The total droplet number concentration (N_d, cm^{-3}) is noted in the legend for each MSD.

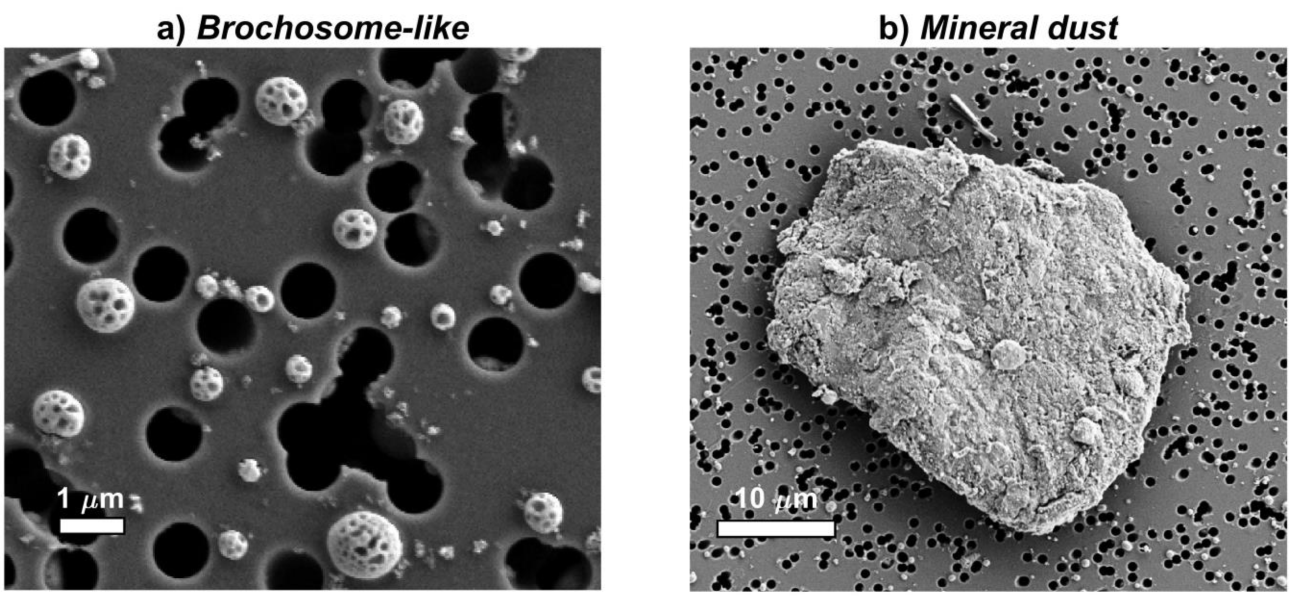

Figure 5. Scanning electron microscope images of Brochosome-like particles (a), and mineral dust (b) collected during the same period as the Atlantic-1 MSD measurement.

5) In this paper, only results from the simulation with the most unstable thermodynamic profile are analyzed in detail. For shallower clouds, the rain yield and the max. cloud mass show monotonic change with CCN concentration and no significant changes with GCCN. So the conclusion should be generalized to reflect how the results change for other thermodynamic situations.

Author reply: We thank the reviewer for this comment. Based on this and to give a more general picture of our results, we changed the abstract to describe the other profile's results as well (see answer no. 1

above). In addition, we added more explanations regarding the intermediate and shallow profiles to other parts of the manuscript.

Results **(Section 3, L121—126):** "The general shape of the five curves is similar **for the deep and intermediate profiles**, and exhibits a non-monotonic trend (see Fig. 2a—b **and Fig. S2a—b, respectively**): an increase in total rain yield and the cloud's maximum mass as a function of aerosol loading, up to a maximum optimal aerosol concentration (N_{op}), followed by a decrease. All five curves have a similar N_{op} of around $N_{tot} = 677\ cm^{-3}$ **($N_{tot} = 416\ cm^{-3}$ for the intermediate profile)** for both surface rain yield and maximum cloud mass. **For the shallow profile, the five MSD curves preset only the decreasing branch, with a minor decrease in rain yield and cloud mass with increasing aerosol loading.**"

Results **(Section 3, L135—139):** "For the cases of shallower cloudy-layers, where the clouds are more subjected to entrainment effects, the ascending branch of the curves is less pronounced (**intermediate profile,** Fig. S2a—b) or non-existent (**shallow profile,** Fig. S2c—d). We, therefore, focus on the deepest atmospheric profile, which better demonstrates the full effect of the competition and interactions between the microphysical processes in the clouds, **and refer to Text S2 in the SI for the intermediate and shallow profiles."**

Summary **(Section 4, L250—254):** "We focused on the deepest profile, since it best captured the effect of competing microphysical cloud processes, and showed that surface rain yield and cloud's maximum mass are affected in a non-monotonic way by changes in N_{tot}, and the shape of the MSDs for most of the cases. **This was also the case for the intermediate profile results, while the shallow one only showed the decreasing branch of this non-monotonic trend, due to more dominant entrainment effects.**"

Text S2. Additional Atmospheric Profiles in the SI was extended and now includes the following (L29—L36): "We examined the surface rain yield and the cloud's maximum mass as a function of N_{tot}. The results of the deepest cloud profile are shown in the main text (Fig. 2a—b), and the other two profiles are shown in Fig. S2. **The trends of the *Atlantic—1* surface rain yield and cloud's maximum mass curves for the intermediate profile are similar to the ones of the deeper profile. The only difference is that the rain yield values of the *Atlantic—1* are higher than the ones produced by the other MSDs for $N_{tot} > 677\ cm^{-3}$. All the curves show a lower N_{op} compared to the deepest profile curves. Under the shallow thermodynamic profile, the *Atlantic—1* rain yield curve shows a similar trend to all other MSD cases, while producing the highest rain values. As for the trend in cloud mass, the *Atlantic—1* shows a monotonic increase (similar to the deep and intermediate profiles).**"

Technical corrections:

1) Line 143, change "accumulating" to "accumulated";

Author reply: Changed.

2) Line 206: Figure 4b,d should be "Figures 4b-d.

Author reply: Corrected.