

## Response to Anonymous Referee #2

**(Comment) This is a theoretical study of sensitivities of cloud droplet size distributions to initial aerosol loading. There are two unique aspects in this study: first, the authors limit their discussions on cloud top properties only; second, the sensitivity tests are thoroughly spaced over aerosol characteristics, including total number, median size, standard deviation of a log-normal distribution, and the hygroscopicity. This is a clearly structured manuscript with adequate figures and literature overview. The conclusions agree with various previous studies using different modeling tools and/or with different parameter choices. The main limitation of the current study is the use of a highly simplified kinematic model, albeit with detailed microphysical representations. I understand that there are tradeoffs to be made in order to carry out a large number of sensitivity tests. However, there should be a much more detailed discussions listing various limitations, and their associated errors, in both the kinematic framework and in handling aerosol activation processes. In addition, I think the scientific quality of the current manuscript can be improved with additional simulations and analyses. I will detail my suggestions in the follow section. There could be significant revisions if the authors decided to carry out some of the additional sensitivity studies.**

**(Answer) We would like to thank Anonymous Referee #2 for taking the time to analyze our work and suggest improvements. We agree that a more detailed discussion on the limitations of the modelling approach, including additional simulations to assess the influence of its shortcomings, would improve the scientific quality of the manuscript. Following the suggestions of both referees, we performed several modifications in the model. The new simulations allowed us to analyse the behavior of the sensitivities in diverse situations, providing a new perspective to the results. We are currently modifying the manuscript hoping to provide a deeper and clearer insight on the results already shown.**

In this document we provide detailed responses to the issues raised in the review, as well as a description of the new capabilities of the model.

### Major points:

**1. (Comment) There are significant limitations in using a kinematic model. In addition, some key aerosol activations processes in the model that have been simplified. The authors skimmed some of these limitations here and there in the manuscript. However, they have missed the most important aspect of the limitation discussions, that is, how these simplifications might affect their main conclusions. This is essential if the conclusions were to be useful for understanding aerosol-cloud interactions in the real world. I would suggest that the authors add a discussion section before the conclusion, to carry out some detailed, in-depth discussions. The following is the list of my suggested topics. Some of them are more obvious than others. Some of them are totally missing in the manuscript and need careful considerations.**

(Answer) In order to address the impact of the limitations in the modelling approach on the results, we have modified two key aspects in the model: the treatment of the aerosol and the computation of the vertical velocity.

#### I. Aerosol:

To better account for changes in the aerosol size distribution, we introduced a set of 19 bins for dry aerosols, with radii ( $r$ ) between  $0.0076$  and  $7.6 \mu\text{m}$ , according to Kogan (1991). We consider that the total number concentration of aerosols is log-normally distributed through those bins, at the beginning of the simulation, and can vary by advection, activation and regeneration after droplet evaporation. In-cloud aerosols can also vary by entrainment, which is explained later in this document.

At a given temperature and supersaturation, the critical dry size for aerosol activation is computed from the Köhler equation (Pruppacher and Klett, 1997). The initial bin for newly nucleated droplets is assigned according to its equilibrium size at 100% relative humidity, if  $r > 0.09w^{0.16}$  (Ivanova et al., 1977), where  $w$  represents the vertical velocity (m/s). According to Ivanova et al. (1977), for larger aerosols, the initial radius of the droplet will exceed  $r$  by a factor of  $k = 5.8w^{0.12}r^{0.214}$ , due to the time these particles take to reach its equilibrium size.

This method has been extensively employed (e.g., Yin et al., 2000ab, 2005; Altaratz et al., 2008; Hill et al., 2008; Mechum and Kogan, 2008) to substitute the explicit calculation of the diffusional growth of the aerosols from its dry sizes, which has a much higher computational demand. Leroy et al. (2007) analysed the influence of a similar assumption on the liquid and ice water content and the aerosol particles, drops and ice crystal spectra simulated by a 1.5D model. He found notable consistency between both approaches, even when the bin resolution was strongly decreased, as well as a reasonable sensitivity to the initial aerosol spectra.

The described approach has two major effects on the model. Previously, the activated droplets were always assigned to the smallest bin of the DSD, thus inducing a very narrow shape and spending a longer time to grow by diffusion until the collision-coalescence rate increases. In the current version, the newly activated droplets fill several bins of the DSD, which favors the development of wider DSDs and accelerates collection processes. Also, by using bins for the aerosol, we allow the PSD to evolve freely, which has a strong impact on the results. By fixing a log-normal size distribution, the previous version of the model guaranteed the continuous supply of larger aerosols for activation. Although the number concentration of aerosols decreased according to the amount of activated droplets, the assumed log-normal shape implied the presence of particles in the right tail of the PSD, which was actually removed. In the updated version, after activation, the tail of the PSD can only be filled again if new particles are advected, entrained or replenished due to droplet evaporation.

The aerosol regeneration is included here following the approach of Kogan et al. (1995) and Hill et al. (2008). It considers that large CCN particles grow to large cloud drops, which evaporates less efficiently than small droplets. Thus, small CCN will be released before large ones. As a result, the regenerated CCN are replenished to the aerosol bins starting by the smallest activated size, until the original number concentration in each bin is attained. If the number concentration of regenerated CCN is larger than the number concentration of "missing" aerosols (considering the initial PSD), which can happen by advection of droplets

to levels different than those where they were nucleated, the “excess” of CCN will be log-normally distributed according to the initially defined median radius and geometric standard deviation. A constraint is added to this scheme to conserve the domain-averaged aerosol size distribution.

This scheme provides a reasonable way to parameterize the aerosol regeneration without using a two dimensional probability density function to track the aerosols. It does not consider the processing of the aerosols inside the cloud, therefore, it could induce errors in the activation rate in situations where the collision-coalescence process is a significant sink of small aerosols and a source of larger aerosols (Lebo and Seinfeld, 2011). However, its use is justified in our case because of the occurrence of only low rates of evaporation. This evaporation takes place right above cloud-top, due to the advection of droplets to upper, unsaturated levels. Hence, even if the collision-coalescence significantly modify the size of the aerosol particles, when partial evaporation occurs, only the smallest droplets will deactivate. The collision-coalescence effect on the aerosol size distribution would have to be considered in cases with large evaporation rates, where even large droplets, containing the largest original or processed aerosols, deactivate.

## II. Vertical velocity

We introduced a new method for estimating the vertical velocity in the model. It is done by solving the simplified vertical momentum equation (Pruppacher and Klett, 1997), considering the buoyancy and the weight of the liquid water, as well as the reaction force on the parcel resulting from the acceleration of the air in the neighborhood (Turner, 1963):

$$\frac{dW}{dt} = \frac{g}{1 + \gamma} \left( \frac{T - T'}{T'} - w_L \right) - \frac{\mu}{1 + \gamma} W^2,$$

where  $\gamma \equiv m'/2m \approx 0.5$ .

and  $\mu$  is the entrainment rate.

For a plume of radius  $R_J(z)$ , the entrainment rate can be expressed as  $\mu_J = C/R_J$ , where  $C \approx 0.2$  is the entrainment parameter. The equation for the radius of the plume is:

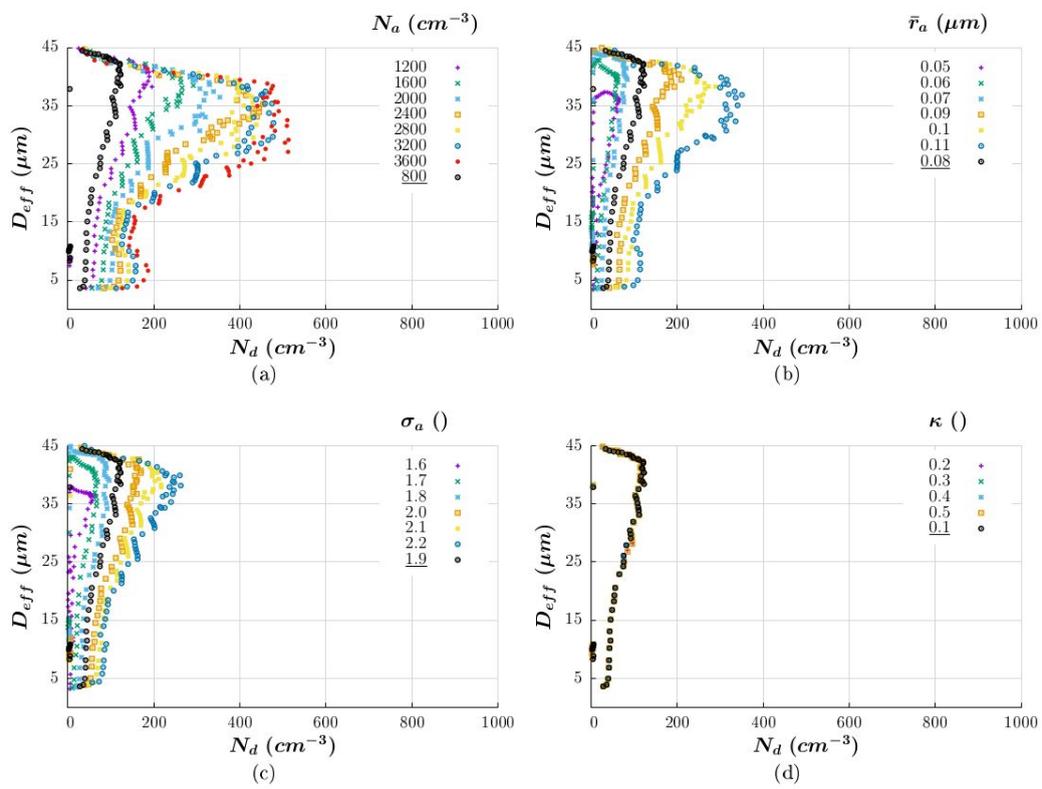
$$\frac{d \ln R_J}{dt} = \frac{1}{2} \left[ \mu_J W - \frac{d \ln \rho}{dt} - \frac{d \ln W}{dt} \right]$$

For the case with no entrainment,  $\mu_J = 0$  and we also neglect the acceleration of the parcel in the neighborhood, i.e., eliminate the second term and the factor  $1/(1+\gamma)$  in the first term in the right side of the vertical velocity equation.

The contributions of the entrainment in the equations for the evolution of the potential temperature, the water vapor mixing ratio and the aerosols is expressed as  $\mu_J(X-X')W$ , where  $X$  and  $X'$  represent the in-cloud and environment values for each one of the mentioned magnitudes, respectively.

For the purpose of representing a rising plume, we introduce a constant temperature perturbation at surface. The vertical profile of potential temperature and water vapor mixing ratio are taken from the Boa Vista sounding on 11/09/2014 at 12Z, the same as in the original tests, but no smoothing procedure is applied in this case.

The results obtained with the updated model, with no entrainment, are presented below:

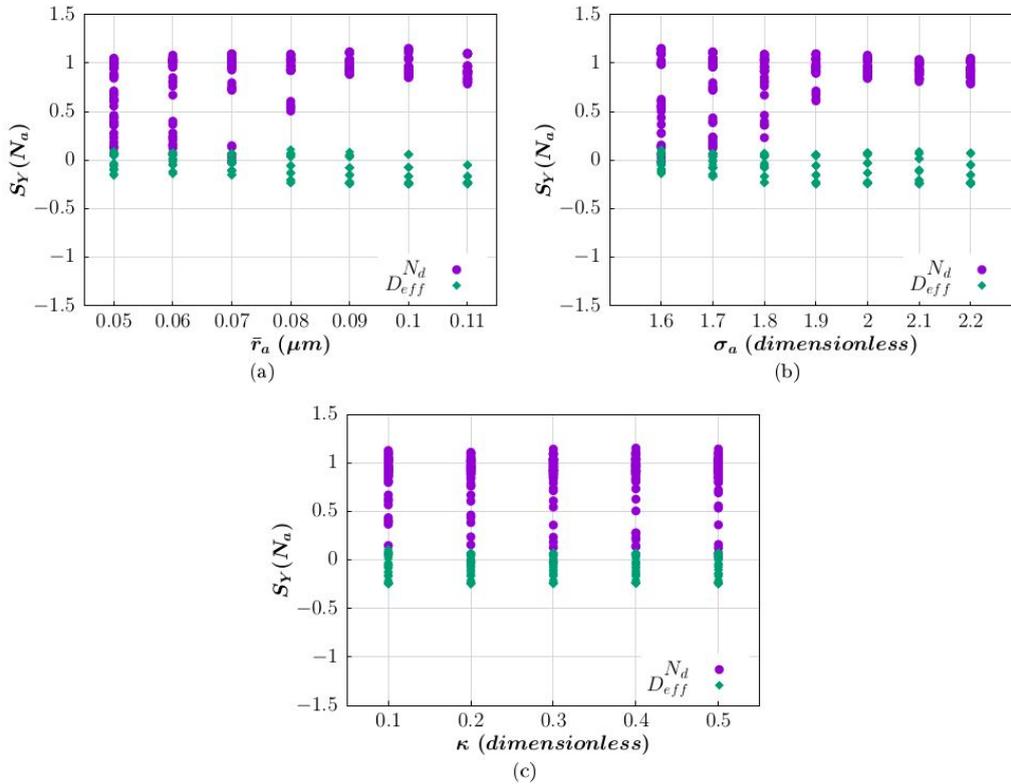


**Figure R1.** Illustration of the sensitivity of cloud-top bulk properties to (a) the aerosol number concentration ( $\text{cm}^{-3}$ ), (b) the median radius of the aerosol size distribution ( $\mu\text{m}$ ), (c) the geometric standard deviation of the aerosol size distribution (dimensionless), and (d) the aerosol hygroscopicity (dimensionless). The markers represent the averaged DSDs for the time steps when the cloud top remains at the same model level during its growth. The colors distinguish between simulations using different values of the parameter specified at the top of the graphs. The control simulation is represented by black markers in the figures.

Figure R1 shows a reduction of the droplet concentration ( $N_d$ ) and an increase of the effective diameter ( $D_{eff}$ ), compared to Fig. 3 in the original manuscript. It is a direct consequence of the modification in the treatment of the aerosol, as explained above. That is the reason why the values of the aerosol parameters are not the same than in the original tests. With the current configuration of the model, when the original values of the parameters are used, there is a very low nucleation rate and the cloud does not develop. It is reasonable, considering that once the aerosol is removed from activation, they are not spread over all sizes as in the previous version of the model, so that no more droplets are nucleated.

The trajectories in Fig. R1 keep the overall shape shown in Fig. 3 (main manuscript) until a critical point, where  $N_d$  start decreasing with height. This effect is due to the combination of two factors: the decrease of the nucleation rate and the increase of the collision-coalescence. Note that there is an inverse relation between  $N_d$  and  $D_{eff}$  at the critical

point in Fig. R1a, i.e., the smaller the aerosol number concentration, the smaller  $N_{d,crit}$  and the larger  $D_{eff,crit}$ . It also happens in the tests with varying  $r_a$  and  $\sigma_a$ , but only for the values larger than the control ones. It evidences that, in the cases with smaller  $r_a$  and  $\sigma_a$ , the decrease of the nucleation rate due to the lack of large aerosols is the dominant factor controlling the upper part of the trajectories. The “saturation” effect that appeared in the tests with varying aerosol number concentration, instead of the tests with the size-related parameters. It indicates the state at which all the supersaturation is consumed, and the system is therefore insensitive to the addition of more aerosols.



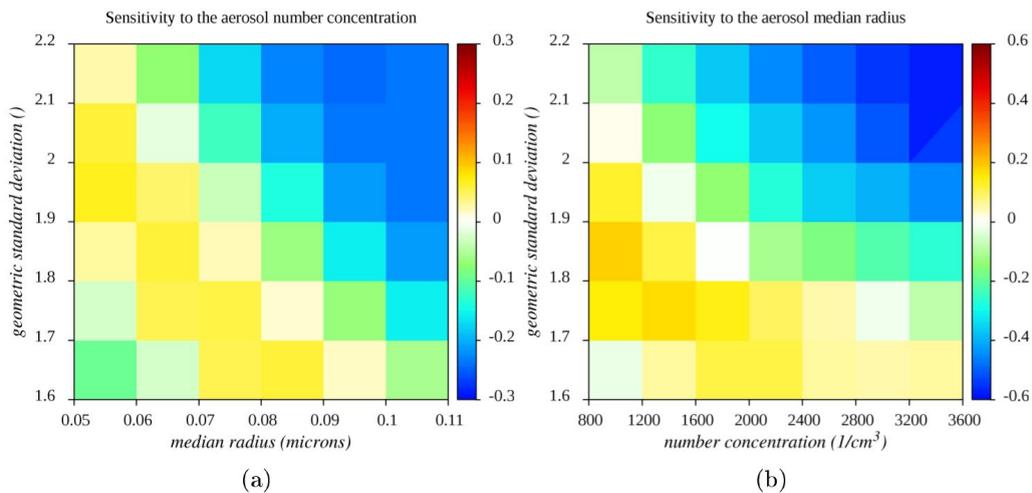
**Figure R2.** Sensitivities of the droplet number concentration and effective diameter to the aerosol number concentration ( $S_Y(N_d)$ ) as a function of (a) the median radius of the aerosol size distribution ( $\mu\text{m}$ ), (b) the geometric standard deviation of the aerosol size distribution (dimensionless) and (c) the aerosol hygroscopicity (dimensionless).

Figure R2 shows that the sensitivity of  $N_d$  to the aerosol number concentration can be almost null for small values of  $r_a$  and  $\sigma_a$ , while having almost no dependency on the aerosol hygroscopicity. It is consistent with the original tests, despite the difference in the values of the parameters tested. However, there is one effect that was not evident in the original tests: a secondary decrease in the sensitivity is found as the aerosol size distribution displaces toward larger aerosols and becomes wider. The latter effect is caused by the supersaturation depletion related to the enhanced activation of aerosols.

The variations in the sensitivity of the droplet effective diameter  $D_{eff}$  to the aerosol number concentration  $N_d$  are better illustrated in Fig. R3a. It can be observed that it reaches positive values for  $\sigma_a = -13.3r_a + 2.7$  approximately, and decreases otherwise. These positive values

are due to absence of water vapour competition. At those points, increasing the aerosol number concentration will create more droplets (note that the sensitivity of  $N_d$  to  $N_a$  is relatively high in that situation), increasing the vertical velocity by latent heat release, and therefore the supersaturation. But the increment in the number of droplets is not as intense as needed to cause a significant water vapour depletion, and since all the droplets will grow in the presence of such high supersaturations,  $D_{eff}$  is increased. On the other hand, for the smallest values of  $\sigma_a$  and  $r_a$ , the sensitivity is again negative. In that situation, only the largest aerosols in the right tail of the PSD are activated. Larger drops have a slower rate of growth by condensation, and the collision-coalescence rate may also be decreased due to less variety of fall speeds. Thus, even at high supersaturations, the growth of these droplets can be slower. In addition, when the total number concentration is increased and the shape of the distribution is maintained, the largest increments in the amount of aerosol occur near the center (mode values). Now, let's consider what happens in the right tail of the PSD, i.e., the aerosols that will be activated. In that situation, since the largest increments in number concentration occur toward the center of the distribution, the smaller sizes in the right tail will be favored, leading to a decrease in  $D_{eff}$  after activation. If the droplets growth rate is not as intense as to balance that trend, it will result in negative sensitivity.

Overall, Figure R3 shows that increases in both  $r_a$  and  $N_a$  have a tendency to produce lower  $D_{eff}$  (negative sensitivity). However, the effect is controlled by  $\sigma_a$ . For relatively narrow aerosol PSDs, increases in  $N_a$  or  $r_a$  have a lesser effect given the limited population of aerosols above the activation diameter. On the other hand, broader aerosol PSDs allow the  $r_a$  and  $N_a$  effects to go through. In the Amazon, the combination of aerosol sources (e.g. biogenic, biomass burning and urban) can lead to relatively broad aerosol PSDs, suggesting that it is more likely to find negative  $D_{eff}$  sensitivities. Cecchini et al. (2017) found an average  $S_{D_{eff}}(N_a)$  of  $-0.25$  from aircraft measurements.



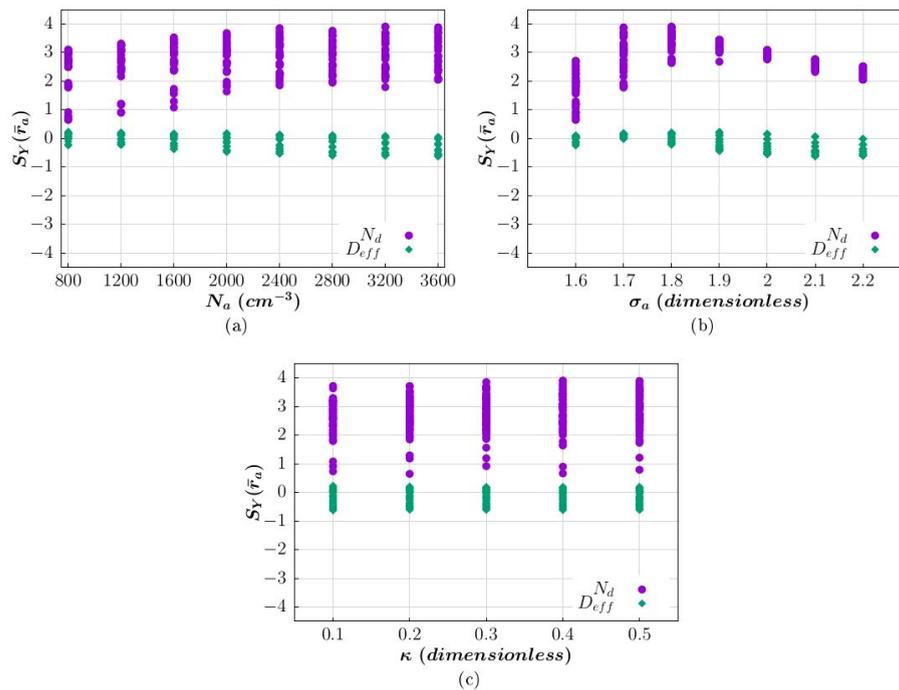
**Figure R3.** Sensitivity of the droplet effective diameter to (a) the aerosol number concentration ( $S_{D_{eff}}(N_a)$ ) and (b) the aerosol median radius ( $S_{D_{eff}}(r_a)$ ) as a function of other aerosol properties.

The sensitivity of  $N_d$  to the aerosol median radius (Fig. R4) increases for high values of  $N_a$  and  $\sigma_a$ , in agreement with our previous results, but unlike in the original test, has a very small dependency on the aerosol hygroscopicity. Also, the absolute values of  $S_{N_d}(r_a)$  in this version

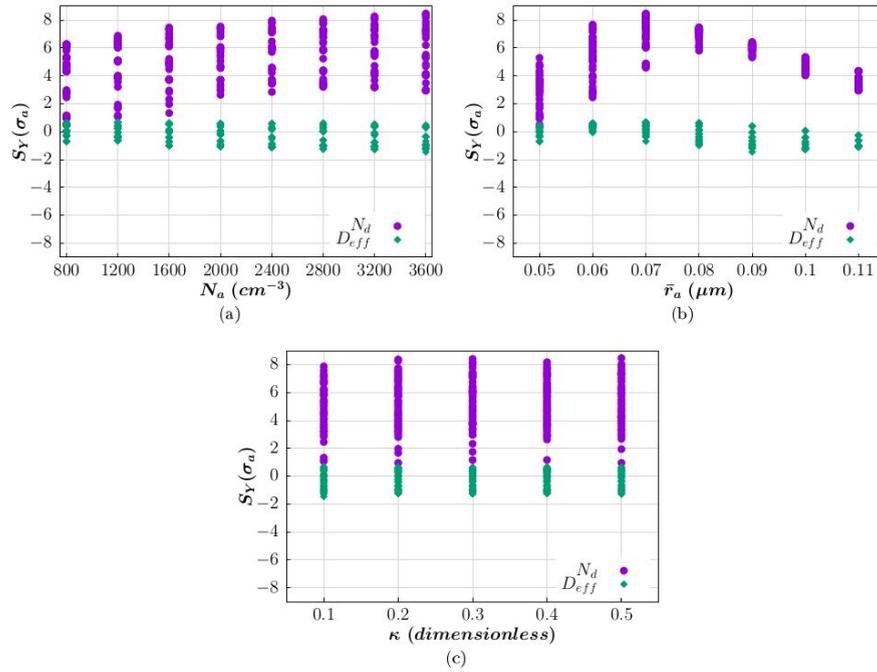
can be more than twice as large as in the original tests. The influence of the depletion of suitable-sized aerosols and water vapor is again visible for the smaller and larger values of  $\sigma_a$ , respectively, generating a maximum sensitivity at  $\sigma_a \approx 1.7$ . It reflects also in the behavior of  $S_{Def}(r_a)$ , which response to varying  $N_a$  (Fig. R3a) is similar to the response of  $S_{Def}(N_a)$  to varying  $r_a$  (Fig. R3a).

Like in the original tests, the sensitivity to the geometric standard deviation of the aerosol size distribution (Fig. R5) doubles in absolute value and shows a behavior similar to  $S_Y(r_a)$ .

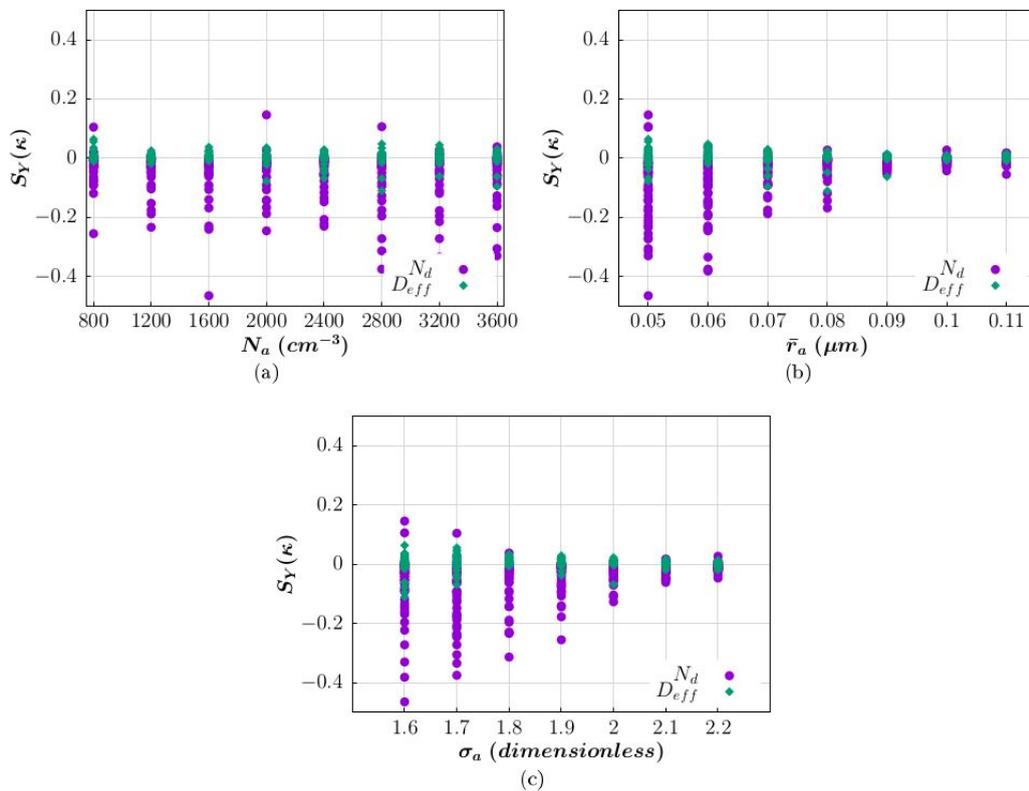
The low values of the sensitivity on the aerosol hygroscopicity (Fig. R6) are consistent with its small influence on the sensitivities of the other parameters, as mentioned above. The trend of its absolute value is similar to the one in the original tests, but the sign of the sensitivities is mostly the opposite. It is reasonable, in this version of the model, because higher values of  $\kappa$  define smaller critical radii for activation. Although at first it would increase the droplet number concentration, it also contributes to a faster depletion of the larger aerosols, leading to a reduction in the nucleation rate afterward.



**Figure R4.** Sensitivities of the droplet number concentration and effective diameter to the median radius of the aerosol size distribution ( $S_Y(r_a)$ ) as a function of (a) the aerosol number concentration ( $cm^{-3}$ ), (b) the geometric standard deviation of the aerosol size distribution (dimensionless) and (c) the aerosol hygroscopicity (dimensionless).



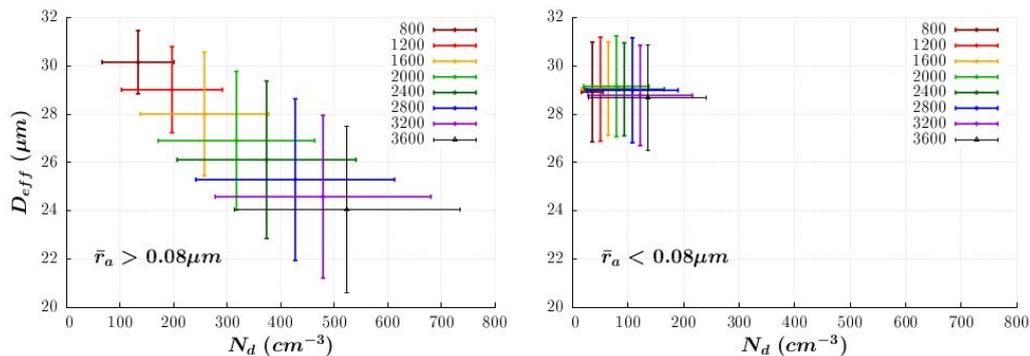
**Figure R5.** Sensitivities of the droplet number concentration and effective diameter to the geometric standard deviation of the aerosol size distribution ( $S_Y(\sigma_a)$ ) as a function of (a) the aerosol number concentration ( $\text{cm}^{-3}$ ), (b) the median radius of the aerosol size distribution ( $\mu\text{m}$ ) and (c) the aerosol hygroscopicity (dimensionless)



**Figure R6.** Sensitivities of the droplet number concentration and effective diameter to the aerosol hygroscopicity ( $S_Y(\kappa)$ ) as a function of (a) the aerosol number concentration ( $\text{cm}^{-3}$ ),

(b) the median radius of the aerosol size distribution ( $\mu\text{m}$ ) and (c) the geometric standard deviation of the aerosol size distribution (dimensionless).

Finally, aiming to complete the comparison with the original tests, we computed the variability of the cloud droplet bulk properties to emulate the information in Fig. 8 in the main manuscript. Figure R7 shows that the variability of the droplet number concentration and effective diameter (represented by the size of the bars -the standard deviation- in the figure) does not present a significant dependence on the aerosol size, in this case. Instead, the variability is a function of  $N_d$  and  $D_{eff}$  on their own. In other words, the difference between both graphics resides on the position of the points -for smaller aerosols,  $N_d$  will be lower and  $D_{eff}$  will be larger, than for large aerosols-, and that location defines their standard deviation, i.e., points located at the left upper corner in Fig. R7a have approximately the same standard deviation than points at the same location in Fig. R7b.



**Figure R7.** Mean and standard deviation of the time-averaged values of  $N_d$  and  $D_{eff}$  at the cloud top for each simulation.

**a) (Comment) Will the conclusions change if a full dynamic model were used?**

(Answer) The simulations performed here represent an idealized cloud resulting from observed humidity and temperature profiles and from either a prescribed or prognosed vertical velocity. The control simulation with the original version of the model was previously validated by comparing the evolution of the cloud-top against in-situ observations (see the response to Anonymous Referee #1). Also, the agreement of our results with previous studies regarding the sensitivity to aerosol properties indicates some reliability in our methodology. However, even if we assume it represent a realizable situation, corresponding to an average behavior, it does not include the variety of possibilities existing in real cases. Important processes such as turbulent entrainment and dynamic feedbacks can introduce a significant departure from the idealization we are considering, as Anonymous Referee # 2 pointed out. Full dynamical models account for dynamics feedbacks and several subgrid processes that could enhance or reduce the range of sensitivities that are demonstrated here. Nevertheless, the qualitative behavior of our main results, i.e., the dependency of the cloud sensitivity to the aerosol properties according to its position in the full parameter space, might not change. For example, Gettelman (2015) simulated

several warm rain cases with the KiD and climatological cases with a global model, using a double-moment microphysics scheme, in order to analyze the sensitivity of the aerosol-cloud interaction to cloud microphysics. They found that the test in the KiD were consistent with the global sensitivity tests. This is an aspect we intend to study in a following work, to build on the present results.

**b) (Comment) If the initial sounding and/or vertical velocity profile changed, will it change the conclusions?**

(Answer) In order to address this question, we performed several sets of simulations for increased/decreased values of the potential temperature, the water vapor mixing ratio and the vertical velocity using the original version of the model. The initial profiles of temperature and water vapor were modified by adding/subtracting 0.5K and 0.5g/kg, respectively, at all heights. The vertical velocity was modified by means of the maximum updraft speed parameter ( $W$  in equation 1 in the manuscript) to take values of 4m/s and 6m/s. For each one of this modifications, a set of simulations were performed in a way similar to the tests illustrated in Fig. 3 in the manuscript, i.e., when varying one aerosol parameter, the others were fixed at its control values. Then, we calculated the sensitivity  $S_{Nd}(X_i)$  and  $S_{Deff}(X_i)$  according to equation 2 in the manuscript. The results are summarized in Table 1 and Table 2 below, where the sensitivity for each condition is specified, together with the difference of the sensitivity compared to the control case (“diff” columns) and the percentage this difference represents compared to the control value (“%” columns).

Table 1. Sensitivity of the droplet number concentration to the aerosol parameters specified in the first row.

	N <sub>a</sub>			r <sub>a</sub>			sigma <sub>a</sub>			kappa		
	S <sub>Nd</sub>	diff	%	S <sub>Nd</sub>	diff	%	S <sub>Nd</sub>	diff	%	S <sub>Nd</sub>	diff	%
control	1,0026			0,5464			0,3551			0,0313		
q <sub>v</sub> -0.5 g/kg	1,0017	-0,0009	0,09	0,5048	-0,0416	7,61	0,2600	-0,0951	26,77	0,0247	-0,0066	21,05
q <sub>v</sub> +0.5 g/kg	0,9834	-0,0192	1,91	0,6180	0,0717	13,12	0,5404	0,1853	52,18	0,0438	0,0125	39,90
Theta -0.5 K	0,9911	-0,0115	1,15	0,5926	0,0462	8,46	0,4595	0,1044	29,41	0,0395	0,0081	25,92
Theta +0.5 K	1,0004	-0,0022	0,22	0,5173	-0,0290	5,31	0,2836	-0,0715	20,15	0,0296	-0,0017	5,54
W=4	0,9867	-0,0158	1,58	0,5867	0,0403	7,38	0,5423	0,1872	52,72	0,0317	0,0003	1,01
W=6	1,0100	0,0075	0,75	0,5692	0,0229	4,18	0,3144	-0,0407	11,45	0,0314	0,0000	0,11

Table 2. Sensitivity of the droplet effective diameter to the aerosol parameters specified in the first row.

	N <sub>a</sub>			r <sub>a</sub>			sigma <sub>a</sub>			kappa		
	S <sub>Deff</sub>	diff	%	S <sub>Deff</sub>	diff	%	S <sub>Deff</sub>	diff	%	S <sub>Deff</sub>	diff	%
control	-0,3522			-0,2245			-0,1433			-0,0125		
q <sub>v</sub> -0.5 g/kg	-0,3611	0,0089	2,52	-0,2104	-0,0140	6,25	-0,0783	-0,0650	45,37	-0,0177	0,0052	41,46
q <sub>v</sub> +0.5 g/kg	-0,3617	0,0095	2,69	-0,2518	0,0273	12,17	-0,1891	0,0458	31,97	-0,0203	0,0078	61,95
Theta -0.5 K	-0,3568	0,0045	1,29	-0,2441	0,0197	8,76	-0,1694	0,0261	18,19	-0,0151	0,0025	20,19
Theta +0.5 K	-0,3590	0,0068	1,93	-0,2136	-0,0108	4,82	-0,0930	-0,0503	35,08	-0,0137	0,0012	9,51
W=4	-0,3411	-0,0112	3,17	-0,2400	0,0156	6,93	-0,2008	0,0575	40,14	-0,0157	0,0032	25,69
W=6	-0,3617	0,0095	2,70	-0,2180	-0,0065	2,89	-0,1162	-0,0271	18,89	0,0545	0,0419	334,75

In Tables 1 and 2, the red (blue) color of cells indicates increased (decreased) sensitivity relative to the control simulation, excepting the sensitivity of the droplet

effective diameter to the hygroscopicity parameter ( $\kappa$ ) at  $W=6$ , to which we will refer later. Yellow cells are for percentage differences higher than 5%, for reference.

It can be observed in the tables that the sensitivity to the aerosol concentration remains almost unaltered in all cases, with percentage differences relative to the control of less than 3.17%. The sensitivities to the aerosol median radius and geometric standard deviation result enhanced when the water vapor mixing ratio and temperature are increased (which corresponds to a decreased potential temperature,  $\theta$ ), and reduced otherwise. They are also increased for smaller vertical velocities. For the larger value of the vertical velocity, the sensitivity to these size-related parameters tend to be smaller, despite the fact that  $S_{N_d}(r_a)$  is larger for  $W=6$  than for  $W=5$  (control value) by a 4.18%. However, its value is still smaller than for  $W=4$ . Whether that behavior can be related to the existence of a minimum in the sensitivity of  $N_d$  for  $W=5$  is something that needs a much more detailed analysis, with a larger number of simulations for variations in  $W$ . Nevertheless, given the relatively low value of the percentage difference in both  $S_{N_d}$  and  $S_{D_{eff}}$ , it can also be related to other factors in the calculations, such as those arising from the definition of the cloud-top, for example, or from the error in the fit procedure to calculate  $S$ . On the other hand, the sensitivities to the aerosol hygroscopicity are very small due to the values of the control parameters, as already commented in the manuscript. The influence of the variations in  $q_v$  and  $\theta$  on the sensitivities to  $\kappa$  is the same than for the size-related parameters, except that it seems to exist a minimum of  $S_{D_{eff}}$  at the control values of the initial profiles. As mentioned before, a deeper analysis would be necessary to understand its causes. The vertical velocity has almost no influence on  $S_{N_d}(\kappa)$ , but a relatively huge influence on  $S_{D_{eff}}(\kappa)$ . The sensitivity of the droplet effective diameter to  $\kappa$  is increased by a factor of three at  $W=6$ , compared to its value at  $W=5$  (control). Also, besides increasing its modulus, it changes the sign of the sensitivity parameter, meaning that increasing the aerosol hygroscopicity would increase the droplet effective diameter. That behavior is opposite to the expected response, considering that a  $\kappa$  is inversely proportional to the critical radius for droplet activation. However, for a given number of aerosol particles, if the updraft is strong enough, the large rate of nucleation can deplete the aerosol content causing the supersaturation to be “used” for increasing droplet sizes thereafter. In that situation, increasing  $\kappa$  would accelerate the aerosol depletion, favoring the increase of  $D_{eff}$  from then on.

These tests include only a subset of the entire parameter space. To a deeper understanding of the effects of the environmental conditions on the cloud sensitivity to aerosols, it would be necessary to perform a analysis similar to that we present in the manuscript, i.e., simulate all the possible combination of the parameters values over its interval of realizable values.

We are currently performing new tests including variations in the initial profiles, with the updated version of the model. Since the evolution of the variables are better coupled now, it would be interesting to know whether the above results maintain.

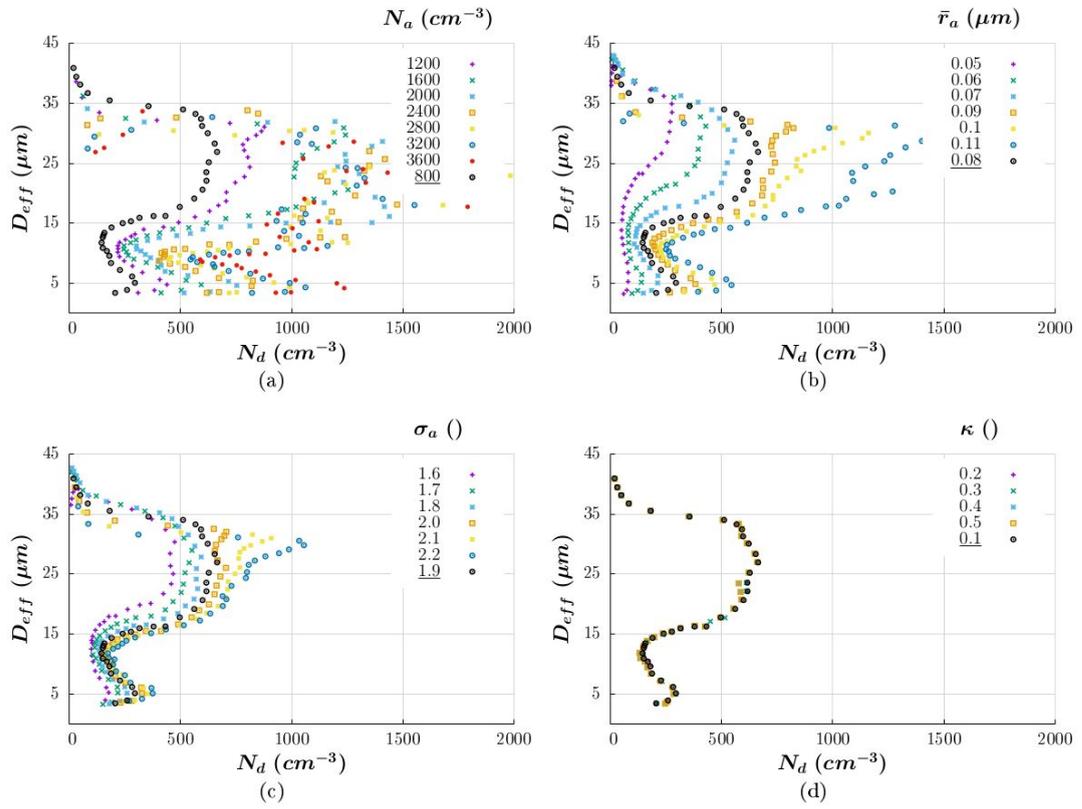
**c) (Comment) A small cumulus with cloud top below 6km seem to be the closest real world resemblance of the kinematic model setup. A key piece that is missing is the entrainment of environment air, together with additional**

aerosols, into such a small cumulus. This is not discussed at all in the manuscript. The entrainment could come from the cloud bottom, side of the cloud, and most challenging, from the cloud top. Since the focus of this study is the cloud top properties, the variations in the cloud top entrainment along might change the existing conclusions. I think that the entrainment can be added fairly easily in the kinematic framework, with pre-determined entrainment rates and vertical variations. I suggest that the authors repeat their calculations with various entrainment rates, repeat the analysis, and see if the conclusions remain the same. I am particularly interested in how the cloud top properties change if entrainment from the top is added. I believe these additional simulations will improve the scientific quality of this study significantly.

(Answer) As explained above, we introduced a parameterization for the effect of lateral entrainment in the simulations. We consider that the column in the model is located in the center of a plume with radius  $R(z)$ , which mixes homogeneously with the radially entrained air at each level  $z$ . The entrainment affects the vertical velocity, the temperature, the humidity and the amount of aerosols in the column. Past studies in the Amazon have assumed that the entrainment mixing in Amazonian clouds is close to the extreme inhomogeneous case, given that the droplet effective radius remain relatively constant horizontally (e.g. Freud et al., 2011). However, the recent studies of Pinsky et al. (2016) and Pinsky and Khain (2018) indicate that homogeneous and inhomogeneous mixing can be indistinguishable for polydisperse DSDs, especially for wide distributions. Additionally, those studies show the inadequacy of previous in-situ techniques to identify mixing type. Based on this finding, we will stick to the homogeneous case in the present study as a first approximation. Further studies would be needed to assess the effects of inhomogeneous mixing and this comparison is beyond the scope of this manuscript.

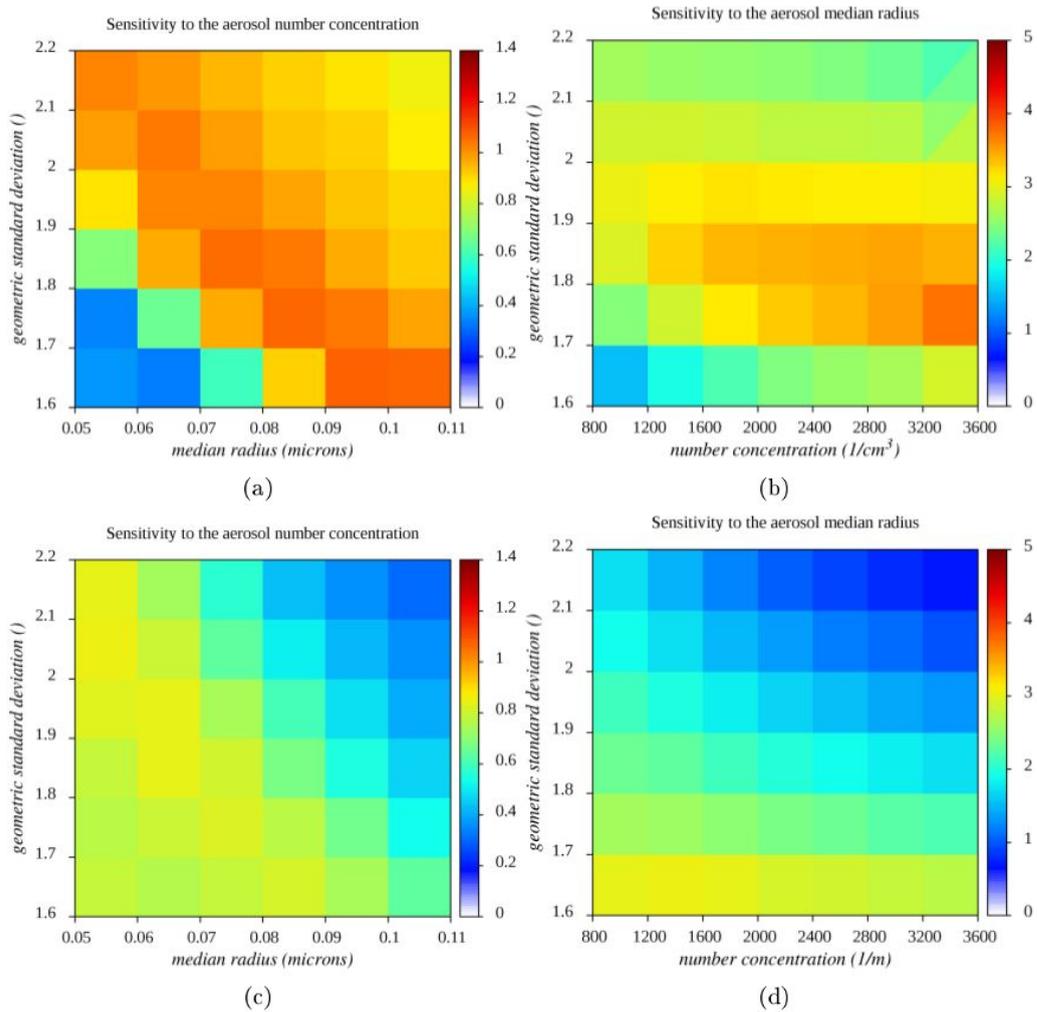
Some cloud-top mixing is resolved in the model grid. However, it can be affected by the numerical diffusion and dispersion introduced by the scheme that solves the advective terms. In the updated version of the model, we use the Lax-Friedrichs first order, conservative scheme to compute the advection of temperature, water vapor and aerosol, and the ULTIMATE scheme to solve the advection of hydrometeors. A first order upwind scheme is used for solving the vertical velocity equation. The choice of the schemes was done by trial and error, in an attempt to minimize the cloud-top dispersion. However, the representativeness of the mixing induced by such an advection at cloud top must be analyzed carefully, and is out of the scope of this paper. For now, we limit our analysis to the results with and without lateral entrainment, as a proxy for the effect of the dilution caused by mixing with the air in the neighbourhood of the clouds.

Since the entrainment decreases the buoyancy of the rising air, including this process significantly reduces the cloud-top height. In order to obtain a thicker cloud, we increased the temperature perturbation at surface, compared to the no entrainment simulations.



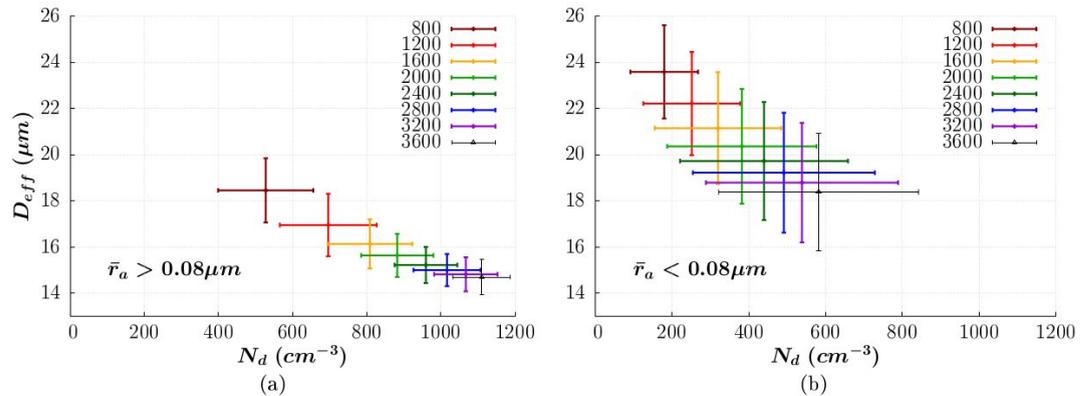
**Figure R8.** Similar to Fig. R1 but for the tests with entrainment.

Figure R8 shows the cloud-top trajectories for several combinations of the aerosol properties in the case including the entrainment. Compared to the no-entrainment cases (Fig. R1), there is an increase of  $N_d$  and a decrease of  $D_{eff}$ , better approaching the behavior reflected in the original tests. The inverse relation for  $N_d$  and  $D_{eff}$  at the critical point (point where there is a change in the monotonicity of  $N_d$ ) holds for all the combinations of the size-related parameters, which evidences the neutralization of the aerosol depletion effect in this case.



**Figure R9.** Sensitivity of the droplet number concentration to (a, c), the aerosol number concentration ( $S_{Nd}(N_a)$ ) and (b, d) the aerosol median radius ( $S_{Nd}(r_a)$ ) as a function of the other aerosol properties. (a,b) tests without entrainment, (c,d) tests with entrainment.

In Fig. R9, it can be seen that the effect of the entrainment in the sensitivity to the aerosol properties is to change the relative importance of the aerosol and water vapor depletion effects. In the no-entrainment case (Fig. R9a,b), the effect of the aerosol depletion predominates over the effect of the water vapor depletion, for smaller-sized and less numerous aerosols. On the other hand, the effect of the consumption of the supersaturation by large, numerous aerosols, is more evident in the case including entrainment (Fig. R9e,f). It is caused by both the increase of nucleation and the mixing with the entrained, drier air. In that case, even small, sparser aerosols do not cause a significant reduction of the sensitivities, because of the supply of aerosols by entrainment.



**Figure R10.** Similar to Fig. R7 but for the tests with entrainment.

Figure R10 illustrates the variability of the bulk properties of the droplet size distribution for the tests with entrainment. It shows that, in a better agreement with the original tests, the variability of  $N_d$  and  $D_{eff}$  is considerably larger for the simulations with smaller aerosols.

**d) (Comment) Prognostic aerosol activation is another significant limitation of the current study. On P4, L24, the authors stated that they use “a 0.25 factor that attempts to accommodate for the fact that not all CCN will grow to the size of the first droplet bin.” Please discuss in details how the factor of 0.25 was chosen, how this factor could affect aerosol activation and cloud droplet spectra, and how it will affect the sensitivities.**

(Answer) We agree that we were not clear about its meaning, origin and importance. We limited to just mention it, considering that it is a feature of the parameterization (Stevens et al., 1996). We also didn't express its function correctly, in order to do so, it is necessary to clarify that this artifice only applies to the mass increment in the first bin, not to its number concentration. The mass of each bin is a key feature in the TAU scheme, since it employs the method of moments for the calculations of vapor deposition and collection. However, given that the first bin contains very small droplets, the application of this factor does not significantly influence the results. In the new version of the model, the mentioned factor is not considered.

**e) (Comment) Since aerosols are represented prognostically, there is no sink term for them in the microphysical calculations. In reality, aerosols are removed in clouds through both activation and wash out. Please discuss how this simplification will affect the conclusions.**

(Answer) As explained earlier in this document, in the original version of the model, the total number concentration of aerosols was modified by activation, advection, and regeneration although fixing its size distribution. In the updated version, the introduction of bins for the aerosol number concentration allows to represent the evolution of the aerosol size distribution as well, and aerosols are also modified by

entrainment and mixing. Washout is not included, since the amount of precipitation produced in the simulations is negligible.

**f) (Comment) Aerosol sizes also grow with increasing supersaturation, and consume certain amount of water vapor supply. This is not considered in the model. How important is this process?**

(Answer) The consumption of water vapor by pre-activated aerosols is not considered in the model. The only sink of water vapor we consider is the droplet activation. We assume that aerosols smaller than the activation size don't represent a significant sink of water vapor, given the great availability of humidity over the Amazon.

**2. (Comment) There are significant vertical variations in simulated cloud properties, as shown in Fig. 2. It will be beneficial to conduct the same sensitivity calculations in Fig. 3 for vertically averaged cloud properties, and compare them with the cloud top properties. The results can also be compared with Cecchini et al (2017).**

(Answer) Cecchini et al. (2017) used the measurements of aircraft penetrations at the top of growing cumulus to analyse the sensitivity of the droplets population to the aerosol loading, vertical velocity and cloud-top height (taken as a proxy for cloud evolution).

In order to compare our results with Cecchini et al. (2017), we calculated the sensitivity of  $N_d$  and  $D_{eff}$  to the aerosol number concentration at intervals of 200m above cloud base ( $H$ ). For consistency with their results, we consider the average and standard deviation of the sensitivity values for all the subsets ( $H, r_a, \sigma_a, \kappa$ ):

$$S_{N_d}(N_a) = 0.82 \pm 0.55$$
$$S_{D_{eff}}(N_a) = -0.19 \pm 0.08$$

The mean sensitivities are very close to the values reported by Cecchini et. al. (2017), although with higher standard deviations due to the much more detailed nature of the simulations as compared to the aircraft measurements.

Minor points:

**1. (Comment) P2, L23: “Must of the previous studies” should be “Most. . .”;**

(Answer) This error was corrected in the manuscript.

**2. (Comment) P3,L28: “1 s” should be “1s”, so is “1200 s”;**

(Answer) This error was corrected in the manuscript.

**3. P8, L5: “Thus the width of the aerosol spectrum can be more important for droplet activation than. . .”. I don't agree with this statement.**

(Answer) The intended meaning of this statement is that, since the sensitivity is higher, a given change in the geometric standard deviation of the PSD would modify the DSD more than a proportional change in the other magnitudes. However, the effect of varying a parameter will be determined by its range of possible values. As the variations in the aerosol median radius, total number concentration and composition can be larger, its impact will be more significant.

**4. (Comment) Calculations in Fig. 6 have different units. One cannot compare numbers with different units.**

(Answer) By definition, the sensitivity is dimensionless. That is the reason why the graphs in Fig. 4,5,6,7 can be compared between each other.

**5. (Comment) Fig. 3: What is the meaning of individual point with the same color? Are they averages over certain time period, or across certain height levels, or something else?**

(Answer) Same color points in Fig. 3 apply for averages of the DSDs according to cloud-top height. The text was modified in the manuscript to clarify the meaning of this graph.

**6. (Comment) It will be nice if the zero lines are labeled in Figs. 4-7, so the positives and negatives can be clearly separated.**

(Answer) These figures will be modified in the corrected manuscript.

## **References**

ALTARATZ, O. et al. Aerosols' influence on the interplay between condensation, evaporation and rain in warm cumulus cloud. *Atmospheric Chemistry and Physics*, v. 8, n. 1, p. 15-24, 2008.

CECCHINI, Micael A. et al. Sensitivities of Amazonian clouds to aerosols and updraft speed. *Atmospheric Chemistry and Physics*, v. 17, n. 16, p. 10037-10050, 2017.

FREUD, E.; ROSENFELD, D.; KULKARNI, J. R. Resolving both entrainment-mixing and number of activated CCN in deep convective clouds. *Atmospheric Chemistry and Physics*, v. 11, n. 24, p. 12887-12900, 2011.

GETTELMAN, A. Putting the clouds back in aerosol–cloud interactions. *Atmospheric Chemistry and Physics*, v. 15, n. 21, p. 12397-12411, 2015.

HILL, A. A.; DOBBIE, S.; YIN, Y. The impact of aerosols on non-precipitating marine stratocumulus. I: Model description and prediction of the indirect effect. *Quarterly Journal of the Royal Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography*, v. 134, n. 634, p. 1143-1154, 2008.

IVANOVA, E. T. Method of parameterizing the condensation process of droplet growth in numerical models. *Izv. Akad. Sci. USSR Atmos. Ocean Phys.*, v. 13, p. 821-826, 1977.

KOGAN, Yefim L. The simulation of a convective cloud in a 3-D model with explicit microphysics. Part I: Model description and sensitivity experiments. *Journal of the atmospheric sciences*, v. 48, n. 9, p. 1160-1189, 1991.

KOGAN, Y. L. et al. Modeling of stratocumulus cloud layers in a large eddy simulation model with explicit microphysics. *Journal of the atmospheric sciences*, v. 52, n. 16, p. 2923-2940, 1995.

LEBO, Z. J.; SEINFELD, J. H. A continuous spectral aerosol-droplet microphysics model. *Atmospheric Chemistry and Physics*, v. 11, n. 23, p. 12297-12316, 2011.

LEROY, Delphine; WOBROCK, Wolfram; FLOSSMANN, Andrea I. On the influence of the treatment of aerosol particles in different bin microphysical models: a comparison between two different schemes. *Atmospheric Research*, v. 85, n. 3-4, p. 269-287, 2007.

MECHEM, David B.; KOGAN, Yefim L. A bulk parameterization of giant CCN. *Journal of the Atmospheric Sciences*, v. 65, n. 7, p. 2458-2466, 2008.

PINSKY, Mark et al. Theoretical investigation of mixing in warm clouds—Part 2: Homogeneous mixing. *Atmospheric Chemistry and Physics*, v. 16, n. 14, p. 9255-9272, 2016.

PINSKY, Mark; KHAIN, Alexander. Theoretical analysis of mixing in liquid clouds—Part IV: DSD evolution and mixing diagrams. *Atmospheric Chemistry & Physics*, v. 18, n. 5, 2018.

PRUPPACHER, Hans R.; KLETT, James D. *Microphysics of Clouds and Precipitation*: Reprinted 1980. Springer Science & Business Media, 2012.

STEVENS, Bjorn et al. Elements of the microphysical structure of numerically simulated nonprecipitating stratocumulus. *Journal of the atmospheric sciences*, v. 53, n. 7, p. 980-1006, 1996.

YIN, Yan et al. The effects of giant cloud condensation nuclei on the development of precipitation in convective clouds—A numerical study. *Atmospheric research*, v. 53, n. 1-3, p. 91-116, 2000a.

YIN, Yan et al. Seeding convective clouds with hygroscopic flares: Numerical simulations using a cloud model with detailed microphysics. *Journal of Applied Meteorology*, v. 39, n. 9, p. 1460-1472, 2000b.

YIN, Y.; CARSLAW, K. S.; FEINGOLD, G. Vertical transport and processing of aerosols in a mixed-phase convective cloud and the feedback on cloud development. *Quarterly Journal of the Royal Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography*, v. 131, n. 605, p. 221-245, 2005.