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Quantifying the aerosol effect on droplet size distribution at cloud-top

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Abstract. This work uses the number concentration-effective diameter phase-space to test cloud sensitivity to variations in the aerosol population characteristics, such as the aerosol size distribution, number concentration and hygroscopicity. It is based on the information from the top of a cloud simulated by a bin-microphysics single-column model, for initial conditions typical of the Amazon. It is shown that the cloud-top evolution can be very sensitive to aerosol properties, but the relative importance of each parameter is variable. The sensitivity to each aerosol characteristic varies as a function of the tested parameter and is conditioned by the base values of the other parameters. The median radius of the aerosols showed the largest influence on this sensitivity. We show that all aerosol properties can have significant impacts on cloud microphysics, especially if the median radius of the aerosol size distribution is smaller than 0.05 μm.

1 Introduction

Because of their role as cloud condensation nuclei (CCN) and ice nucleating particles, aerosols can affect the cloud optical properties (Twomey, 1974) and determine the onset of precipitation (Albrecht, 1989; Braga et al., 2017; Rosenfeld et al., 2008; Seifert and Beheng, 2006) and ice formation (Andreae et al., 2004; Fan et al., 2007; Gonçalves et al., 2015; Khain et al., 2005; Koren et al., 2010; Lee et al., 2008; Li et al., 2011). Aerosols also play an indirect role in the thermodynamics of local cloud fields through the suppression of cold pools and enhancement of atmospheric instability (Heiblum et al., 2016). However, knowledge about the characteristics of the effects of atmospheric aerosols on clouds and precipitation is still lacking and remains an important source of uncertainty in meteorological models.

Many studies have been dedicated to quantifying the effect of aerosols on clouds through sensitivity calculations, using both modeling and observational approaches. Knowing the real values of each parameter that characterize the aerosol is difficult. Also, detailed modeling of droplet nucleation implies a high computational cost. Thus, sensitivity studies intend to determine whether the variability of some characteristics of the aerosol population can be neglected without introducing significant errors in the description of clouds.

A major debate refers to the relative importance of aerosol composition against size distribution and total number concentration (McFiggans et al., 2006). Several studies suggest that accurate measures of aerosol size and number concentration are more

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important to obtain a relatively accurate description of cloud droplet populations (Feingold, 2003; Dusek et al., 2006; Ervens et al., 2007; Gunthe et al., 2009; Rose et al., 2010; Reutter et al., 2009). However, other observations/simulations show that, under certain circumstances, neglecting the variability of the aerosol composition prevent realistic estimations of the aerosol effect on clouds (Hudson, 2007; Quinn et al., 2008; Cubison et al., 2008; Roesler and Penner, 2010; Sánchez Gácita et al., 2017). This circumstantial sensitivity is commonly found in the literature and it refers not only to aerosol composition, but also to other meteorological/aerosol conditions (McFiggans et al., 2006). For instance, Feingold (2003) showed that the influence of aerosol parameters over the droplet effective radius (r_e) varies as a function of aerosol loading. Under clean condition, r_e is mostly determined by the liquid water content and the aerosol number concentration (N_a), with decreasing dependence on the aerosol size distribution, aerosol composition and vertical velocity (w). However, under polluted conditions, all of them contribute significantly to r_e. Reutter et al. (2009) obtained that the variability of the initial cloud droplet number concentration (N_d) in convective clouds is mostly dominated by the variability of w and N_a. They found that the hygroscopicity parameter (κ) appears to play important roles at very low supersaturations in the updraft-limited regime of CCN activation. Also, a significant sensitivity of N_d on the aerosol size distribution parameters was found for some situations belonging to each one of the $w - N_a$ regimes. Karydis et al. (2012) used a global meteorological model to obtain the sensitivity field of N_d to w, uptake coefficient, κ and N_a. They state that, overall, N_d is predicted to be less sensitive to changes in κ than in N_a, although there are regions and times where they result in comparable sensitivities.

To further evidence the importance of aerosol composition on clouds, Ward et al. (2010) consider the Reutter et al. (2009) environmental regimes but vary the log-normal median aerosol radius ($\bar{r}_a$) to examine the behavior of the sensitivity to κ. Their results compare well with the Reutter et al. (2009) regime designation when using the same value of $\bar{r}_a$. However, they show that w/N_a, or supersaturation-based regimes, cannot fully predict the compositional dependence of CCN activity, it also varies significantly as a function of $\bar{r}_a$. It is remarkable that for small aerosols ($\bar{r}_a < 0.06\ \mu m$), composition affects CCN activity even in the aerosol-limited regime.

Most of the previous studies are based on the information from cloud-base. However, given the possibility of occurrence of cloud-top nucleation (Sun et al., 2012), it would be useful to assess the evolution of the cloud-top droplet size distribution (DSD), along with the cloud-base DSD, for exploring the aerosol first indirect effect. In a growing cumulus, the cloud-top represents the beginning of the cloud development at each level, including cloud-base –because, in the initial stage of the cloud life-cycle, both the base and the top coincide in space. Thus, the characteristics of the DSD at cloud-top will strongly impact the evolution of the cloud, modulating the rates of microphysical process onward and therefore determining the structure of the cloud. As Cecchini et al. (2017) pointed out, studies should take into account the altitude above cloud base. The authors showed that, on average, droplet growth with cloud evolution is comparable in absolute value and is opposite to the aerosol effect. They determined that the aerosol effect on DSD shape inverts in sign with altitude, favoring broader droplet distributions close to cloud base but narrower DSDs higher in the clouds.

With ample water vapor supply, high temperatures and a wide spectrum of aerosol conditions, the troposphere over the Amazon constitutes an ideal scenario to study aerosol-cloud-precipitation interaction. Because they belong to the “aerosol-limited regime", characterized by strong updrafts and a low aerosol background concentration (Reutter et al., 2009), Amazonian

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clouds are found to be very sensitive to aerosols (Andreae et al., 2004; Braga et al., 2017; Cecchini et al., 2017; Fan et al., 2018; Reid et al., 1999). At the same time, clouds control both the removal and production of atmospheric particles over the Amazon basin. According to Andreae et al. (2018), the production of new aerosol particles from biogenic volatile organic material, brought up by deep convection to the upper troposphere, is the dominant process supplying secondary aerosol particles in the pristine atmosphere. Then, those particles can be transported from the free troposphere into the boundary layer by strong convective downdrafts or even weaker downward motions in the trailing stratiform region of convective systems (Wang et al., 2016).

Here we propose to explore the cloud sensitivities to several aerosol properties, by simulating some characteristics of Amazon clouds. We focus on the information from cloud-top, during the warm stages of cloud life-cycle, using a sample strategy that also includes the information from the cloud-base at the initial stage of development of the cloud. Our approach is similar to Ward et al. (2010), but it is not limited to analyze the hygroscopicity sensitivity. Instead, we extended the discussion to the sensitivity to the aerosol median size and number concentration too, and consider their effects on both droplet size and concentration.

2 Modelling approach

The model employed here consists of a bin microphysics parameterization (Feingold et al., 1988; Tzivion et al., 1987, 1989) coupled to a single-column Eulerian framework, the Kinematic Driver (KiD) (Shipway and Hill, 2012), with prescribed w. Thus, at each time step, every grid-point parcel receives an influence from two sources: advection and microphysics processes.

The KiD prognostic variables are potential temperature (K) and water vapor, hydrometeor and aerosol mixing ratios (kg kg^{-1}). It uses the Exner pressure as a fixed vertical coordinate and the total variance-diminishing scheme (Leonard et al., 1993) as the default advection scheme. Its prognostic variables are held on "full" model levels, while w and the density are held on both "full" and "half" levels such that the grid can be used as a Lorenz-type (Lorenz, 1960) or Charney-Phillips-type (Charney and Phillips, 1953) grid.

The KiD model was conceived as a kinematic framework to compare different microphysics parameterizations without addressing the microphysics-dynamics feedbacks. Thus, obtaining precise quantitative simulations with KiD cannot be expected; nevertheless, it can provide important insights about the responses of the simulated cloud to changes in the parameters of the microphysics scheme.

In our simulations, a 1 s time step was used for both dynamics and microphysics algorithms during an integration time of 1200 s (20 min). For the vertical domain, a 120-level grid was defined with a 50-m grid spacing from 0 m to 6000 m of altitude.

As initial conditions, vertical profiles of potential temperature and water vapor mixing ratio from an in situ atmospheric sounding[1] were provided (Fig. 1a). We used the 12Z sounding, on September 11, 2014, from Boa Vista-RR, Brazil. The sounding data were interpolated to match the model resolution and then smoothed to represent a more general situation.

[1] http://weather.uwyo.edu/upperair/sounding.html

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Here, the vertical velocity field ($w(z,t)$) was constructed based on the idea of having a layer of positive buoyancy, where a parcel updraft velocity would increase with height until reaching the negative buoyancy layer. The defined time dependence for the velocity maximum and its height roughly simulate the acceleration that the air must experience and the progressive destabilization of the air column (Fig. 1b).

$$w(z,t) = \begin{cases} W \sin\left(\frac{\pi}{2}\frac{t}{T}\right) e^{-\frac{1}{2}\log^2(0.004t - 0.0008z)} & (0.2z - t) < 0 \\ 0 & otherwise \end{cases} \quad (1)$$

In Eq. 1, W represents the maximum updraft speed (with respect to both height and time) in m s^{-1} and T is the length of the simulation in s. The value of W was set to 5 m s^{-1} taking into account the measurements of the ACRIDICON-CHUVA AC09 flight, where the w oscillated between 0 m s^{-1} and 8 m s^{-1} (Cecchini et al., 2017). This flight was performed by the High Altitude and Long Range Research Aircraft (HALO) on the same date of the aforementioned sounding (Wendisch et al., 2016; Machado et al., 2014). It sampled the top of growing convective cumulus over remote regions of the Amazon, starting close to the local noon, in the dry-to-wet season transition.

2.1 Microphysics representation

For the simulations performed in this work, we have used the TAU[2] size-bin-resolved microphysics scheme that was first developed by Tzivion et al. (1987, 1989) and Feingold et al. (1988) with later applications and development documented in Stevens et al. (1996); Reisin et al. (1998); Yin et al. (2000b, a) and Rotach and Zardi (2007).

TAU differs from other bin microphysical codes because it solves for two moments of the drop size distribution in each of the bins rather than solving the equations for the explicit size distribution at each mass/size point, which allows for a more accurate transfer of mass between bins and alleviates anomalous drop growth.

In this version of the TAU microphysics[3], the cloud drop size distribution is divided into 34 mass-doubling bins with radii ranging between 1.56 μm and 3200 μm. The method of moments (Tzivion et al., 1987) is used to compute mass and number concentrations in each size bin resulting from diffusional growth (Tzivion et al., 1989), collision-coalescence and collisional breakup (Tzivion et al., 1987; Feingold et al., 1988). Sedimentation is performed with a first-order upwind scheme. Aerosols are represented by a single prognostic variable, its bulk number concentration, which is assumed to follow a log-normal size distribution. Thus, activation is calculated by applying Köhler's theory to this aerosol distribution, using a 0.25 factor that attempts to accommodate for the fact that not all CCN will grow to the size of the first bin of the droplet distribution.

One disadvantage of our approach is the lack of an explicit representation of the droplet activation mechanism, that would require the definition of bins to simulate the hygroscopic growth of aerosol particles. Another source of inaccuracies comes from considering only one aerosol mode with a single value of κ, ignoring quasi-internal or external mixing states (Rissman et al., 2004; McFiggans et al., 2006; Ervens et al., 2007). However, at first, this approach makes our results suitable to understand how changes in the aerosol properties affect the simulations of numerical models in operational or research configurations, which rarely use a detailed description of the aerosols.

[2]The acronym TAU refers to the Tel Aviv University, where it was primarily developed

[3]Version available at https://www.esrl.noaa.gov/csd/staff/graham.feingold/code/ (Accessed on: 04/11/2017)

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3 Sensitivity analysis

We employ a phase space defined by two bulk properties of the DSD (hereinafter "bulk phase space"): N_d (cm^{-3}), which coincides with the zeroth moment of the DSD, and D_{eff} (μm), which is the ratio between the third and second moments.

Sensitivity tests in the bulk phase space provide a very efficient means to evaluate how a specific parameter variability can affect the evolution of cloud-top DSDs. Here, we test the sensitivity of N_d and D_{eff} at the cloud top to variations in N_a, $\bar{r}_a$, the geometric standard deviation (σ_a) of the aerosol size distribution and κ, using ranges normally found in the Amazon atmosphere (Gunthe et al., 2009; Martin et al., 2010; Pöhlker et al., 2016) (Table 1).

Table 1. Aerosol parameters used for the sensitivity tests: intervals for values and steps between them.

Parameter	Interval	Step
N_a (cm^{-3})	$200 - 900$	100
$\bar{r}_a$ (μm)	$0.02 - 0.08$	0.01
σ_a ()	$1.1 - 1.9$	0.2
κ ()	$0.1 - 0.5$	0.1

The sensitivities were calculated as the slope of the linear fit between Y and X_i in logarithmic scale for normalization:

$$S_Y(X_i) = \frac{\partial lnY}{\partial lnX_i}\Big|_{X_k} \tag{2}$$

where Y represents either N_d or D_{eff}, and X_i is the aerosol property affecting Y. $S_Y(X_i)$ represents the relative change in Y for a relative change in X_i and places less reliance on the absolute measures of parameters (Feingold, 2003; Reutter et al., 2009; Ward et al., 2010). The subscript X_k indicates that when calculating the sensitivity to X_i, the other aerosol parameters are held constant. For each value at which X_k is fixed, we will obtain a new value of $S_Y(X_i)$, i.e. we can also calculate $S_Y(X_i)$ as a function of X_k ($S_Y(X_i, X_k)$).

The latter differentiates our approach from previous studies. Feingold (2003) included the variability of all $X \neq X_i$ when calculating the linear regression between lnY and lnX_i, only distinguishing the results for two subsets of N_a. Similarly, Reutter et al. (2009) analyzed the sensitivities to $\bar{r}_a$, σ_a and κ for three combinations of N_a and w, but all values of Y calculated at a given value of X_i were averaged prior to fitting. This analysis was then expanded by Ward et al. (2010), who calculated $S_{N_d}(\kappa)$ for different values of $\bar{r}_a$ and σ_a used to initialize the parcel model. Now, we use a more general approach that allows us to study the responses of both cloud droplet number concentration and effective diameter to changes in each aerosol characteristic, as a function of the other aerosol parameters used to initialize the model.

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4 Results

The cloud-top was defined here as the last model level, from surface to top, where the droplet concentration was larger than 100 per cm^3. It is represented by black lines in Fig. 2. Figure 2 shows N_d, D_{eff} and the mixing ratio of cloud droplets (q_c), for the entire simulation. Note that the upward advection causes a maximum of N_d at cloud-top for all times. As droplets ascend and mix with new droplets, they grow by diffusion of vapor and, to a lesser extent, by collision-coalescence. As a consequence, D_{eff} and q_c are larger in upper levels at the last times of the simulation.

Firstly, we represented the cloud-top information in the bulk phase space to discuss the "isolated" effect of each parameter, when fixing the values of the others. The control values of the parameters employed here are N_a=800cm^{-3}, $\bar{r}_a$=0.05μm, σ_a=1.5, and κ=0.1.

Figure 3a shows the sensitivity of cloud-top DSDs to the initial concentration of aerosols. Note that an increase of the aerosol concentration increases the number of activated drops, as expected. This nucleation enhancement induces a smaller effective diameter because of water vapor competition, despite a slightly increased liquid water content (not shown). Thus, if the water vapor amount is kept constant, the diffusional growth for each droplet is slowed. The latter manifests as a trend to the horizontal orientation (in the direction of larger values of N_d) in the bulk phase space. For the most polluted situations, a tendency to attain an almost constant effective diameter is evidenced.

Figure 3b shows the sensitivity of cloud-top DSDs to the median radius of the aerosol population. The effects of increasing $\bar{r}_a$ are similar to the consequences of increasing N_a. If we keep the aerosol size distribution shape constant (i.e. the same total concentration and standard deviation) and increase $\bar{r}_a$, then more droplets are activated because of the larger availability of aerosols with sizes above the activation threshold. Thus, nucleation increases, whereas diffusional growth decreases.

The tests in Fig. 3 evidence a type of "saturation" effect for the larger values of $\bar{r}_a$ tested, i.e. the sensitivity decreases as this parameter increases. The combination of two factors explains this behavior: the water vapor availability and the position of the size distribution curve with respect to the critical radius for droplet activation (r_c). Even if continuous water vapor supply from the surface occurs, the water vapor can be completely consumed in each time step, depending on the aerosol availability and the diffusional growth rate. If the number of activated aerosols is able to consume all the water vapor that reaches a layer in a time step, an increase of its quantity will not introduce differences in the DSD. Moreover, for certain positions of the size distribution curve with respect to r_c, an increment in $\bar{r}_a$ does not produce a significant impact on the number of activated aerosols.

Figure 3c shows the sensitivity to the standard deviation of the aerosol size distribution. In our tests, increasing this parameter also causes an increment of the droplet concentration through an enhancement of the nucleation rate. However, the effect of varying σ_a is more important at the earliest stages/lowest levels of the cloud (the extremity with the smallest values of N_d and effective diameter in each path). σ_a modifies the shape of the aerosol size distribution. Although an increase of $\bar{r}_a$ always enhances the number of activated droplets, the same does not apply to σ_a. Instead, the effects of changing σ_a depend on the position of r_c with respect to the size distribution function. For certain values of r_c, increasing σ_a can induce a reduction in the number of droplets that become activated, whereas, for others, it can cause an increase of the number of activated droplets

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(non-monotonic behavior). Given that r_c varies with height, it explains the differences in the effect of σ_a as the cloud height increases. These tests also illustrate that the sensitivity is larger for smaller values of σ_a, because the log-normal distribution shape is more sensitive to σ_a in the interval 1.1-1.5 than in 1.5-1.9.

Finally, Fig. 3d shows the effects of varying κ in the simulation. Given the control values of the other parameters, the effect of changing κ is relatively small. Nevertheless, we can see that increasing κ favors the nucleation through a decrease of r_c. As a consequence, N_d increases, whereas the effective diameter decreases.

The previously mentioned saturation effect can be identified for every spectrum of tests in Fig. 3. There is always an interval of values of the tested parameter in which the system becomes less sensitive. The latter has been discussed before in the literature; for example, it is known that the sensitivity to κ increases substantially as κ decreases (Petters and Kreidenweis, 2007). However, that effect is more or less evident depending on the values of the other parameters.

To illustrate that sensitivity variation, we calculated $S_{\bar{N}_d}(X_i)$ and $S_{\bar{D}_{eff}}(X_i)$, with X_i being N_a, $\bar{r}_a$, σ_a or κ. $\bar{N}_d$ and $\bar{D}_{eff}$ are the time averages of N_d and D_{eff} at cloud-top for each simulation, respectively. From Eq. 2, $S_{\bar{N}_d}(N_a)$, for example, is the slope of the linear fit between the values of $\bar{N}_d$ and N_a in logarithmic scale, for a given combination of $\bar{r}_a$, σ_a and κ. The sensitivity to one aerosol parameter can then be calculated a number of times equivalent to all possible combinations of the values of the other parameters in Table 1. From its definition, it follows that positive (negative) values of $S_Y(X_i)$ correspond to increasing (decreasing) Y as X_i increases. Also, $\mid S_Y(X_i) \mid = 1$ means that a given variation in X_i is accompanied by the same absolute variation in Y.

Figures 4, 5, 6 and 7 show $S_Y(X_i)$ as a function of all values of N_a, $\bar{r}_a$, σ_a and κ considered. Generally, $\bar{N}_d$ can be almost three times more sensitive to changes in the aerosol parameters than $\bar{D}_{eff}$, which stems from the mathematical definition of these physical magnitudes. Also, the results for $S_Y(N_a)$ agree with the theoretical limits referred in the literature and all sensitivity calculations include the ranges of previously reported values (Feingold, 2003; Reutter et al., 2009; Ward et al., 2010). For each value in the x-axis of figures 4, 5, 6 and 7, there are several combinations of the other two parameters; as a result, there are several points for each value of the x-axis in the figures.

The impact of N_a on cloud droplets depends on the values of $\bar{r}_a$, σ_a and κ, as can be seen in Fig. 4. However, this dependency is stronger for the parameters that define the aerosol size distribution, $\bar{r}_a$ and σ_a, than for κ. Note that in Fig. 4c, varying κ has a small effect on the distribution of the points, compared to the effects of varying $\bar{r}_a$ and σ_a in Figs. 4a and 4b, respectively. The points are more dispersed for smaller $\bar{r}_a$ and σ_a and tend to be concentrated around a maximum sensitivity value as $\bar{r}_a$ increases. Generally, for smaller values of $\bar{r}_a$, σ_a and κ, $S_Y(N_a)$ can be almost null, i.e. no more or less droplets are being formed, nor its size distribution is being modified, regardless of the quantity of aerosol in the environment. In the vicinity of this state, the activation of droplets is being determined by the characteristics of the aerosol, instead of its number concentration. Hence, for smaller aerosols, the relative importance of the aerosol properties can be opposite to that at larger sizes. To complement the previous analysis, Figures 6b and 7c evidence that the sensitivity to σ_a and κ, respectively, can be significantly increased for smaller values of $\bar{r}_a$.

Figure 5a shows that the sensitivity to the median radius of the aerosol population increases for higher values of N_a –which agrees with Feingold (2003) and Rissman et al. (2004)– and for lower values of σ_a and κ. Interestingly, the lower variability

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in $S_Y(\bar{r}_a)$ corresponds to the values of N_a, σ_a and κ where the absolute value of the mean sensitivity is minimum, which is opposite to the behavior of $S_Y(N_a)$.

The same applies to the sensitivity to the geometric standard deviation of the aerosol size distribution (Fig. 6), substituting σ_a by $\bar{r}_a$ as the independent variable in Fig. 6b. However, it is remarkable that the absolute values of $S_Y(\sigma_a)$ are the highest between those analyzed here. Thus, the width of the aerosol spectrum can be more important for droplet activation than the aerosol median radius, total concentration and composition. The reason for the small sensitivity evidenced in fig. 3c is the value of $\bar{r}_a$ taken as a reference there. Nevertheless, it is important to remember that, even having a high potential impact (determined by the value of $S_Y(\sigma_a)$ in this case), the effect of varying a parameter will be determined by its range of possible values. For example, assuming that the maximum and minimum values specified in Table 1 determine the entire variation of the parameters in a given situation, it follows that the modification of the DSD induced by a variation of 0.8 in σ_a (an increase ratio of 1.72) would be smaller than the changes in droplet size and number concentration due to a 0.06-μm variation in $\bar{r}_a$ (an increase ratio of 4).

Note that $S_Y(\sigma_a)$ changes its sign, which is related to the previously commented variations in the effect of σ_a depending on the position of r_c with respect to the size distribution function. The positive values obtained by Feingold (2003) for the sensitivity of droplet size on σ_a, as well as the negative values reported by Reutter et al. (2009) for the sensitivity of droplet number concentration on σ_a should be due to the inclusion of larger aerosol sizes, where those signs are predominant (see Fig. 6b).

The effects of κ on the sensitivity to size parameters is higher compared to its effect on $S_Y(N_a)$, i.e. the composition of the aerosol can significantly affect the way droplets respond to changes in the aerosol size distribution (Figs. 5c and 6c).

Finally, the sensitivity to the aerosol hygroscopicity is the lowest between those analyzed here (Fig. 7). The absolute value of $S_Y(\kappa)$ is larger for higher N_a and smaller $\bar{r}_a$ and σ_a, and has a small dependency on N_a, for most of the values considered here. Note that, for the intervals of N_a, $\bar{r}_a$ and σ_a where the absolute value of $S_Y(\kappa)$ is maximum, it is still smaller than the sensitivity to those parameters in the same interval. However, its influence can be more than 50% of the sensitivity to N_a, which confirms that neglecting the effects of the aerosol composition is non trivial. Moreover, the effects of the aerosol composition can be significantly increased in conditions of weak updrafts (Ervens et al., 2005; Anttila and Kerminen, 2007; Reutter et al., 2009). That, in combination with the values of N_a, $\bar{r}_a$ and σ_a can determine an even more important role for κ. As Ward et al. (2010) concluded, the influence of variations in the shape parameter on $S_Y(\kappa)$ are more important for small $\bar{r}_a$. This is evidenced by the dispersion of the points for $\bar{r}_a < 0.05\mu m$ in Fig. 7b. Of course, that dispersion includes the responses to changes in N_a too, it is not entirely caused by the σ_a variability. The information contained in Fig. 7b agrees with the sensitivity of droplets to changes in κ, computed by Ward et al. (2010) for the aerosol-limited regime, as a function of $\bar{r}_a$.

From our analysis it turns out that $\bar{r}_a$ is the most important parameter, from those analyzed, that influences the sensitivity to aerosols. This is particularly interesting because of the importance that has been conventionally attributed to the aerosol number concentration. Considering this sensitivity limitation, for certain conditions, other variables, such as the aerosol median size and size distribution shape, can be more influential in determining the evolution of an air parcel.

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We calculated the time-averaged values of N_d and D_{eff} for the cloud-top DSDs at each simulation. Figure 8 shows the mean and standard deviation of these averages for each value of N_a tested. The length of the standard deviation bars is determined by the ranges of $\bar{r}_a$, σ_a and κ. Figure 8a represents the generally accepted knowledge: given a certain temperature and water vapor availability, the bulk properties of a cloud are mostly determined by N_a. However, this behavior seems to be valid only for the largest values of $\bar{r}_a$. If the aerosol size distribution is displaced to a smaller radius (Figure 8b), then more aerosol characteristics must be specified.

Our results show that the study of the aerosol-cloud interaction must include the parameters describing aerosol properties, such as the size and hygroscopicity, at least for $\bar{r}_a \leq 0.05$ μm. These parameters can produce changes in the DSD as large as those caused by changes in the aerosol concentration. The error associated with its misrepresentation increases with aerosol loading and can be as large as 23% in N_d and 15% in D_{eff} for $N_a = 800$ cm^{-3}, which is larger than the error introduced by a 25% variation in N_a, according to Fig. 8b. Note that aerosol loads larger than 800 cm^{-3} are common in the Amazon, due to biomass burning. These findings are also relevant given the current discussion about the importance of ultrafine aerosol particles in the development of deep convective clouds over the Amazon (Wang et al., 2016; Fan et al., 2018).

5 Summary and conclusions

We illustrated the influence of the aerosol number concentration, the median radius and geometric standard deviation of the aerosol size distribution, and the hygroscopicity of the aerosols on the number concentration and effective diameter of droplets at the top of warm-phase clouds, for initial conditions typical of the Amazon. Given the tested variations in the aerosol properties, the cloud DSDs were found to behave as expected. Overall, when the nucleation is favored, an increase in the droplet number concentration is accompanied by a decrease in the droplet effective diameter. The effects of the investigated parameters were similar in all stages of the cloud-top evolution, except for the geometric standard deviation of the aerosol size distribution. Changes in the aerosol size distribution shape were more important in the earliest stages of the cloud –lower cloud-top heights– due to its dependence on the position of r_c with respect to the size distribution function.

We showed that the sensitivity to each aerosol characteristic varies as a function of the tested parameter and its value depends on the base value of the other parameters. The median radius of the aerosols is the most important parameter, from those analyzed, that influences the sensitivity to the others. Based on its value, it is possible to define, inside Reutter et al. (2009) aerosol limited-regime, a concentration-limited regime, when other aerosol properties can be neglected, and a regime where all size distribution characteristics, total number concentration and hygroscopicity significantly influences the droplet number concentration and effective diameter. This expands the result of Ward et al. (2010) and states that w/N_a, or supersaturation-based regimes (Reutter et al., 2009), cannot fully predict the dependence of CCN activity, not only on the aerosol composition, but on all aerosol characteristics.

Despite using a simpler modeling approach, our results agree with previous studies, which assures the validity of our calculations. Thus, the application of these conclusions is not limited to models that use a similar representation of microphysics processes, but also to theoretical or experimental studies.

Author contributions. LHP performed the model simulations, the model–data analysis and prepared the manuscript. LATM and MAC provided guidance with the definition of the model initial conditions. LATM, MAC and MSG provided guidance with the choice of the variables and its interval of values and the model–data analysis. All authors contributed to the design of the study and the preparation of the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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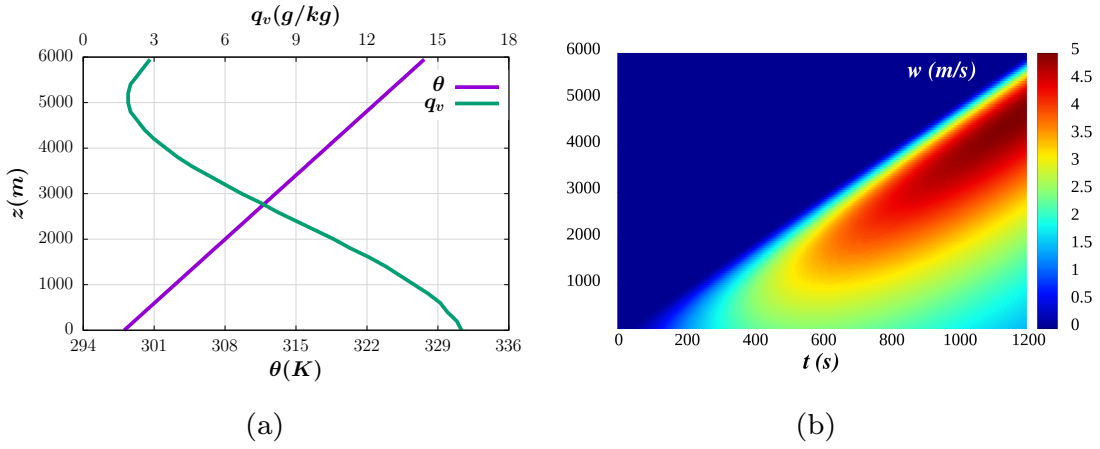

Figure 1. Model configuration: (a) initial conditions and (b) prescribed field of vertical velocity

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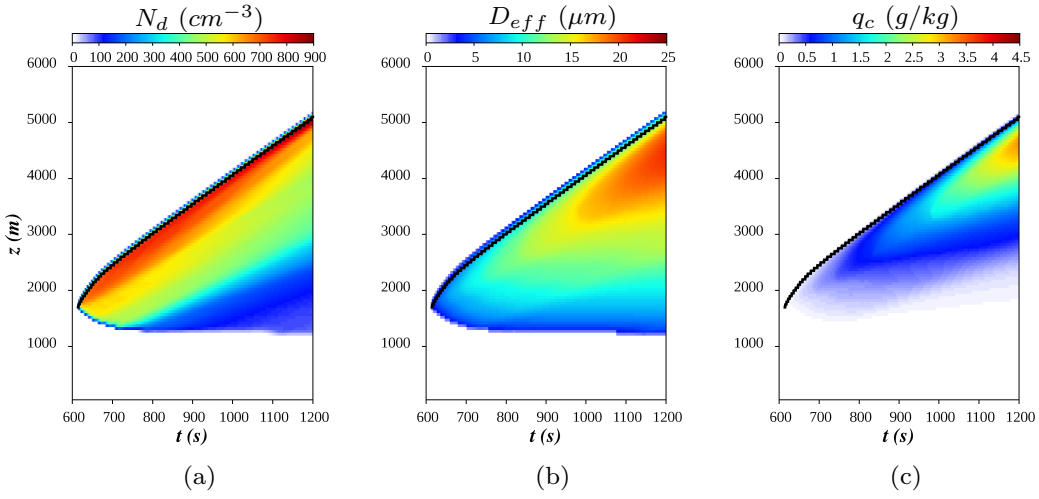

Figure 2. Evolution of N_d (cm^{-3}), D_{eff} (μm) and q_c (g/kg) in the simulation. The black lines represent cloud-top.

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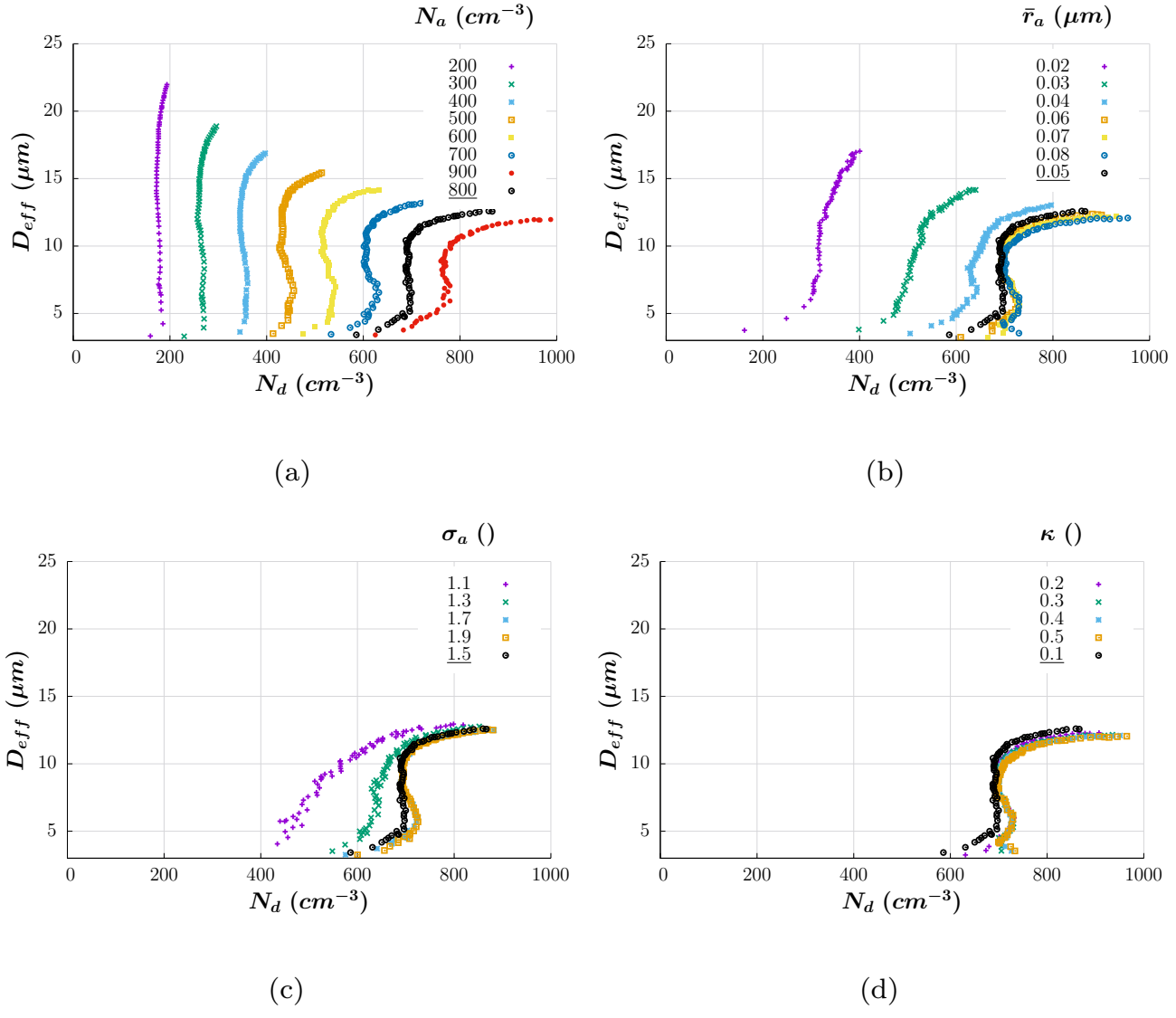

Figure 3. Illustration of the sensitivity of cloud-top bulk properties to (a) the aerosol number concentration (cm^{-3}), (b) the median radius of the aerosol size distribution (μm), (c) the geometric standard deviation of the aerosol size distribution (dimensionless), and (d) the aerosol hygroscopicity (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.

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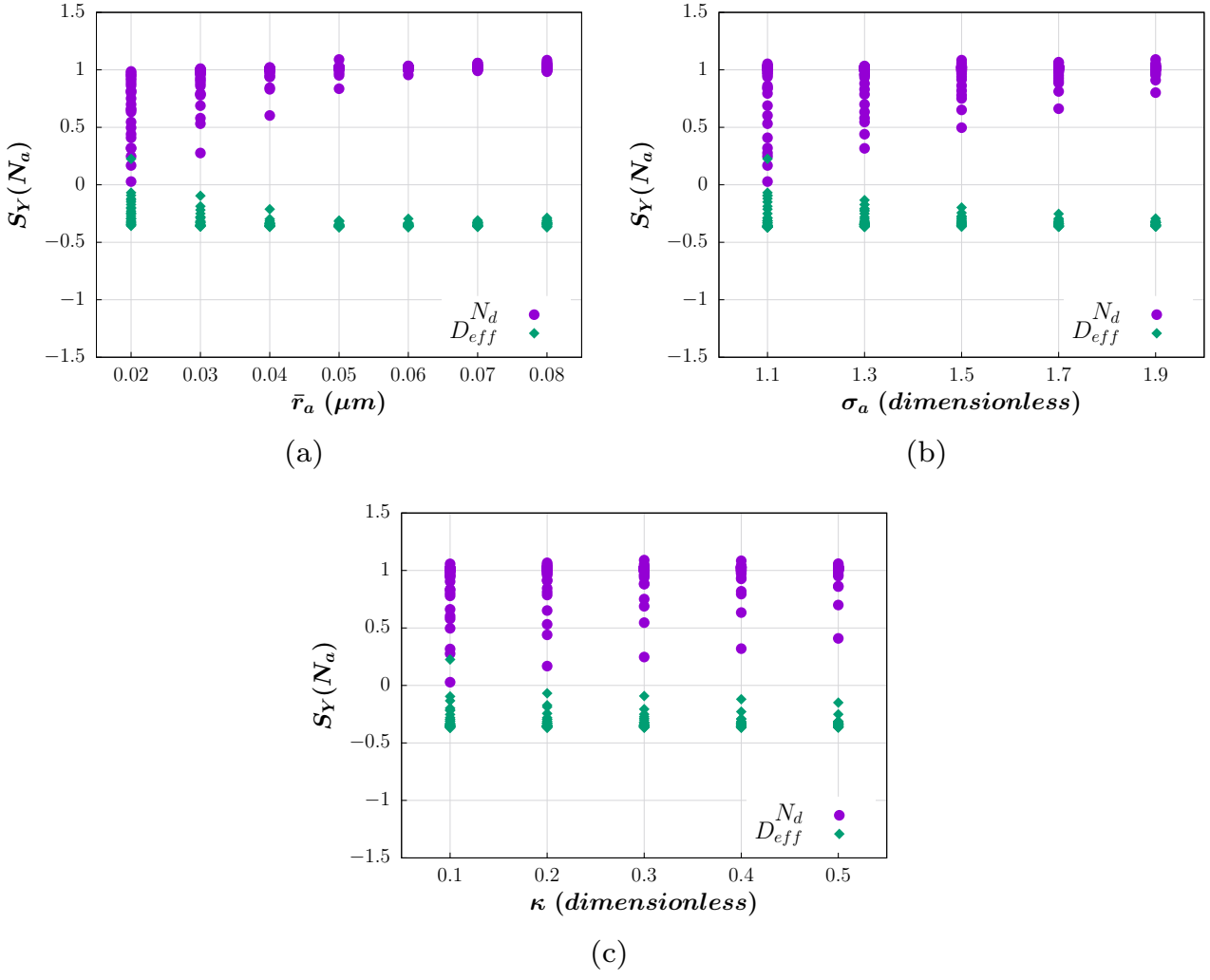

Figure 4. Sensitivities of the droplet number concentration and effective diameter to the aerosol number concentration ($S_Y(N_a)$) as a function of (a) the median radius of the aerosol size distribution (μm), (b) the geometric standard deviation of the aerosol size distribution (dimensionless) and (c) the aerosol hygroscopicity (dimensionless).

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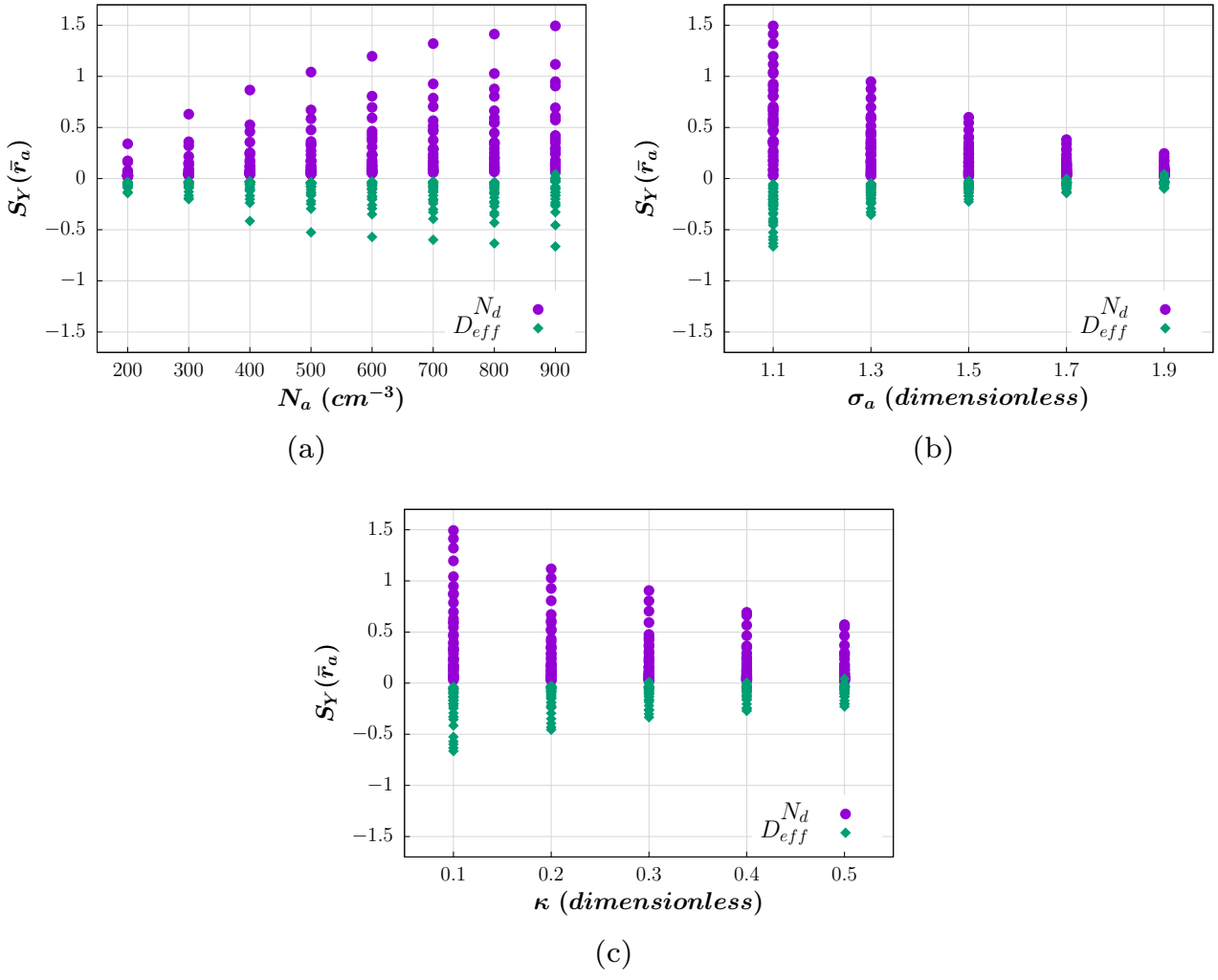

Figure 5. Sensitivities of the droplet number concentration and effective diameter to the median radius of the aerosol size distribution ($S_Y(\bar{r}_a)$) as a function of (a) the aerosol number concentration (cm^{-3}), (b) the geometric standard deviation of the aerosol size distribution (dimensionless) and (c) the aerosol hygroscopicity (dimensionless).

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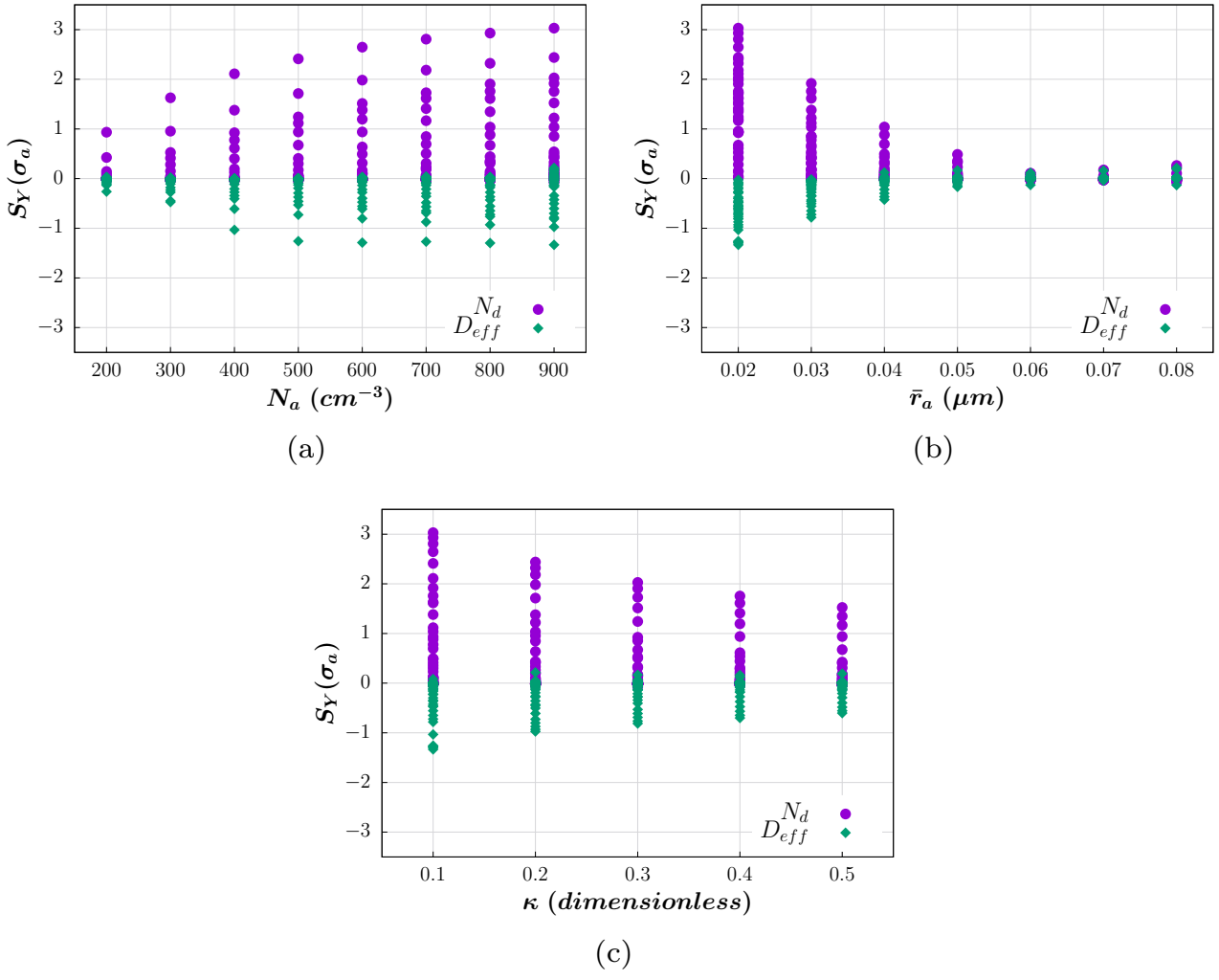

Figure 6. Sensitivities of the droplet number concentration and effective diameter to the geometric standard deviation of the aerosol size distribution ($S_Y(\sigma_a)$) as a function of (a) the aerosol number concentration (cm^{-3}), (b) the median radius of the aerosol size distribution (μm) and (c) the aerosol hygroscopicity (dimensionless).

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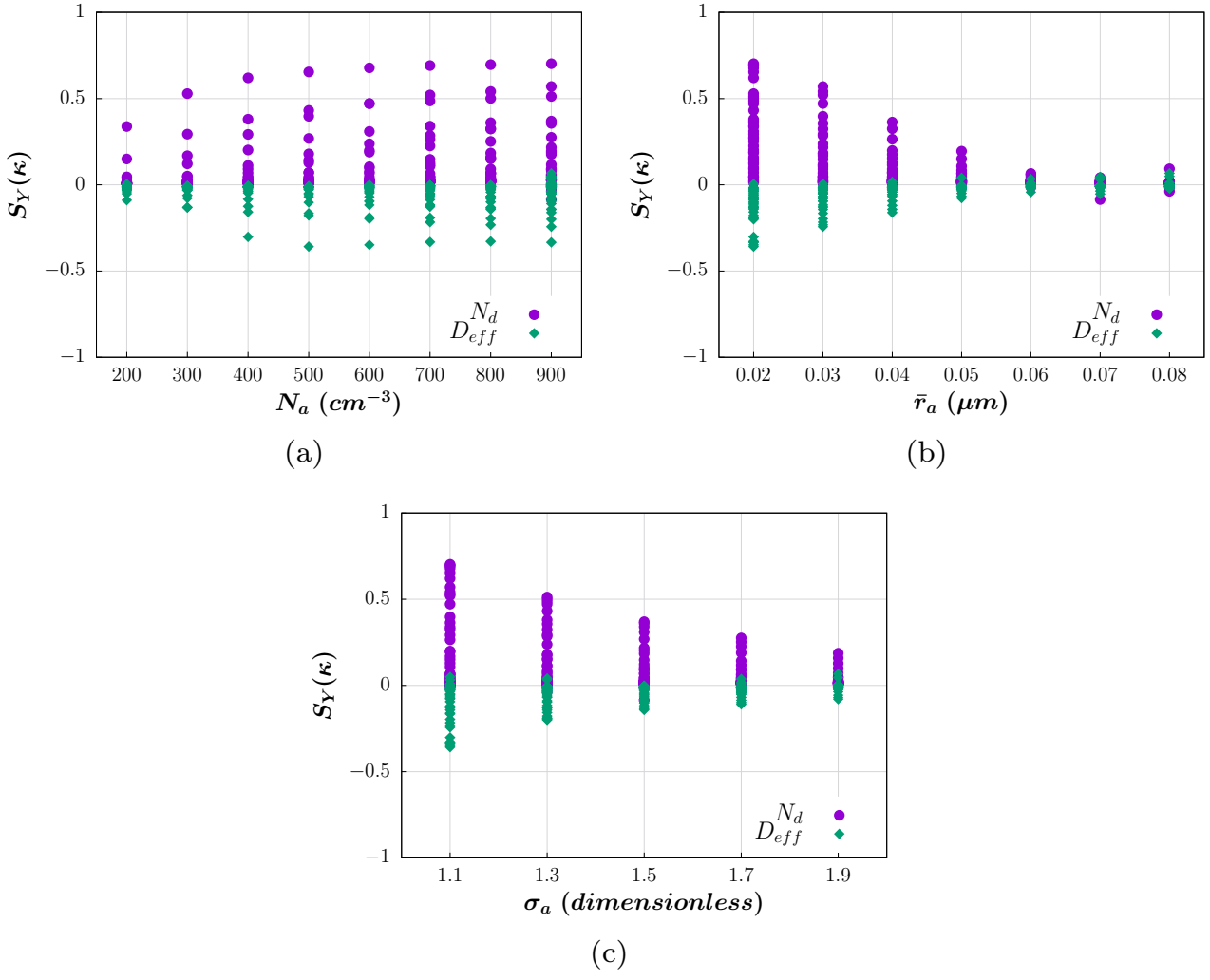

Figure 7. Sensitivities of the droplet number concentration and effective diameter to the aerosol hygroscopicity ($S_Y(\kappa)$) as a function of (a) the aerosol number concentration (cm^{-3}), (b) the median radius of the aerosol size distribution (μm) and (c) the geometric standard deviation of the aerosol size distribution (dimensionless).

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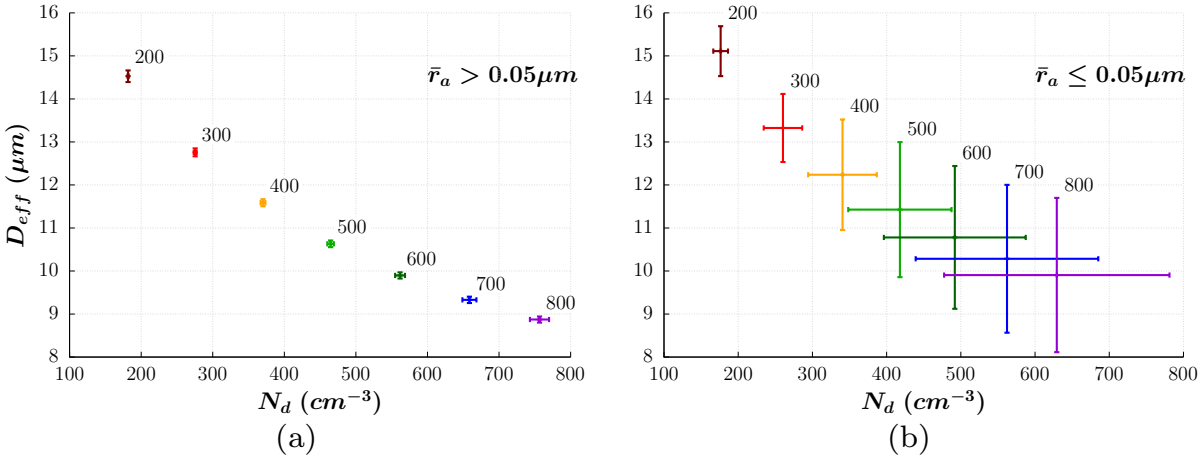

Figure 8. Mean and standard deviation of the time-averaged values of N_d and D_{eff} at the cloud top for each simulation.