

MODELING THE RELATION BETWEEN CCN AND THE VERTICAL EVOLUTION OF CLOUD DROP SIZE DISTRIBUTION IN CONVECTIVE CLOUDS

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1. INTRODUCTION

As suggested in previous works in which satellite and radar data are analyzed (e.g., Rosenfeld 1999), pollution acts severely in cloud microstructure, increasing the number of droplets per unit volume and reducing their mass, as the available water vapor condenses in a larger concentration of cloud condensation nuclei (CCN). The consequence, as suggested by many authors, is the inhibition of warm rain formation through collision-coalescence processes.

It is a common procedure to farmers at Amazon to burn forest while preparing the land for plantation of pasture during the dry season. The fires produce a large amount of small aerosols (mainly via gas-to-particle conversion) and some giant and ultragiant aerosols, mostly composed of soot particles and leached ashes. As a result of large amount of CCN, clouds with large droplet concentrations are produced. Due to its small size, those droplets are not collected by the potential precipitation embryos, which would enable the production of warm rain. As a consequence of this and also to the fact that large scale environment usually have reduced relative humidity, which leads to elevated condensation levels, rain is produced essentially through the cold phase, inside the cells with the necessary vertical development to produce ice.

Recently some of articles have addressed the issue of warm rain suppression in Amazon (Rosenfeld, 1999; Andreae et al, 2004). Nevertheless, because multiple processes interact with each other, experimental studies on warm rain initiation need be complemented with numerical models. Although simple models, as parcel model, can reveal some aspects of the system, the uses of complex model account better for the existence of particularities in such intricate phenomenon.

In this work, we present results from numerical models based on results from the LBA/EMfiN!/SMOCC-2002 experiment. We simulate the variability in cloud microphysics structure observed during the experiment and the influence of pollution on the warm rain initiation. Simulations span a wide range of situations varying from a clean marine to a very polluted environment.

2. BRIEF CAMPAIGN AND DATA DESCRIPTION

LBA/EMfiN!/SMOCC-2002 campaign was designed for studying the impact of biomass burning in climate in Amazon basin, Brazil. The cloud microphysical campaign consisted of several flights and took place from 21 September to 18 October 2002 and was especially designed to investigate different microphysical regimes observed during the dry-to-wet transition period in the Amazon basin. A detailed description of the campaign is given by Andreae et al (2004). A map showing the experimental sites is shown in Figure 1.



Figure 1. Brazil map showing the main experimental point investigated during LBA/EMfiN!/SMOCC-2002 campaign.

The measurements were made using the instrumented aircraft of Ceará State University, UECE. The data of this work came basically from two instruments: the UW83-1 CCN counter, which provides CCN concentration for a range of user-specified supersaturation and a FSSP-100, modified by Droplet Measurements Technology, which allows detection of large aerosols particles.

The different sites encompass four different types of masses:

1. "Blue Ocean" (at Northeast Brazil Coast, which receives aerosols mainly from Atlantic Ocean, due to the predominantly easterly flow.)
2. "Green Ocean" (at Westerly Amazon, over which the forest remains mostly untouched)
3. "Polluted" (over Mato Grosso and Rondônia states, where a large number of fires is responsible for a significant aerosol loading every year, usually until September).
4. "Transitional" (over Rondônia, during the month of October, after a significant rain events)

During the experiment observations have shown wide droplet spectra were found close to Fortaleza, with a fast development of raindrop size particles (with radius between $100\mu\text{m}$ e 1mm). Rainwater takes a significant fraction of the liquid

water mass in already low altitudes (slightly above 2700 m), which agrees with observations of radar echoes of rain clouds with tops below 3 km at Fortaleza.

In a polluted environment, as that found during the intense burning season, the distribution function shows typically two modes, the first composed of small droplets (modal radius close to 10 μm); and a second, of large particles (radius approaching 500 μm). It is noticeable that the small particle mode does not widen, neither there is significant mass growth in the large particle. As collection of small droplets by the large particles do not seems to occur warm rain does not develop.

During the “transition season”, the environment still presents a high concentration of aerosols, but in an amount slightly lower than that found in the middle of the burning season. Also more water becomes available due to the change in the large scale circulation over the Amazon region. As a consequence, raindrop size particles appear faster and are able to collect somewhat larger (compared to their counterparts in a polluted environment) droplet. Some warm rain may develop in this situation.

In the case of a moderately polluted, as can be characterized the transition period from the dry season to the rainy season in the Amazon, the precipitating modal particle is less developed than the clean continental environment, as expected. Moreover, it reveal that the modal radius tends to have difficulties in overcome the values of $\sim 10 \mu\text{m}$. In association with the different CCN concentration found during the aforementioned cloud microphysical regimes, the cloud droplet concentration varied by a factor of about 10 from the blue ocean to the polluted environment.

Clean environments showed smaller droplet concentration, as expected. At the coast, the smallest concentration were found. Despite the possible influence of pollution coming from Fortaleza urban areas, which may account for the occasionally occurrence of concentration around 1000 cm^{-3} , the collision coalescence process may be efficient in this case. Concentration values of the order of 1000 cm^{-3} were also found during flight over the western Amazon.

In polluted environments droplet concentrations are much higher, often exceeding 2500 cm^{-3} . Such increased droplet concentration suggest a significant suppression of precipitation development in the warm phase, since the available water vapor condenses in a large number of very small particles with little chance of coagulation.

The transitional environments shows values of droplet concentration that are close to the ones found in the polluted counterpart. This suggests that the greater precipitation efficiency of clouds during such period is not only due to CCN and droplet concentration.

3. THE MODEL

To perform the simulations of the cloud warm phase we use a parcel version of the model described in Costa et al. (2000) and its complete bidimensional version.

The model was set to allow 178 CCN particles categories, regarding the specific needs on representing CCN of different sizes (including giant or ultra-giant particles) and chemical composition categories. It comprises all microphysics processes associated with liquid phase: nucleation, condensation, evaporation, collision-coalescence, collisional and spontaneous breakup. Additionally, the model comprises 100 hydrometeors bins.

The CCN distribution function is obtained for nuclei radius ranging from approximately $0.006 \mu\text{m}$ up to $6.293 \mu\text{m}$, which corresponds to supersaturation from 3% to approximately zero. The CCN bin concentration is obtained by adding different sets of relations of the form

$$f_i = A_i \left(\frac{r}{r_0} \right)^\alpha \exp(-\beta r) \quad (1).$$

The result is then constrained to follow the relation $N = cS^k$, with c and k ranging from clean to very polluted cases, according to observations.

The numerical parcel model simulations are performed with a prescribed vertical velocity. The model was initialized using conditions typically found during the rainy season in Amazon: surface pressure of 960 hPa, temperature of 303 K, and relative humidity of 80%. As a result, the simulated cloud base was about 460 m. In the bidimensional simulation the initial condition follows exactly the same describes in Costa et al. (2000).

4. WARM RAIN HEIGHT

The use of a parcel model to evaluate modification of the cloud ability to produce rain is questionable. First because it assumes that all particles and drops are confined in the ascending volume. Second, in the cases in which the droplets grow to large sizes because their fall velocities exceed the updraft of the parcel, and thus they would leave the ascending parcel. Third, there is no feedback between the growth of clouds particles and the cloud dynamics, since the model only treats the microphysics of the growth and does not the effect of released latent heat. Therefore the use of a parcel model is only appropriated for the early development stage of the cloud when the particles are still very small or when rain is just starting to develop. These conditions are followed in this section.

According to satellite and local observations (Rosenfeld and Gutman, 1994; Gerber, 1996) the appearance of precipitation is correlated to a required effective radius (r_{effe}) threshold for the drop spectrum, after which the warm rain process is able to develop. We call this value of effective radius the “warm rain threshold” ($r_{\text{effe}} = 14 \mu\text{m}$) and the altitude in which such a threshold is reached inside a parcel in a convective cloud core “warm rain height”.

Figure 2 shows the warm rain height as a function of droplet concentration for simulated condition ranging from a very clean to a very polluted situation for a ascending speed of 5.0 m/s. Simulations also point out that changes in large scale conditions, i.e., a higher water vapor supply in

the environment, can contribute significantly to the reduction of the warm rain initiation altitude. Figure 2 also shows the comparison of 3 simulations, where the initial relative humidity was modified. The height of the clouds base evidently tends to diminish with the increase of the initial relative humidity, but the effect of a higher humidity on the warm rain initiation altitude is still more significant, especially for droplet concentrations higher than 1000 cm^{-3} . For droplet concentrations of about 2000 cm^{-3} , as the ones found in the polluted and transition season, a reduction of 10% in the relative humidity causes a rise of the altitude of warm rain of more than 1200 m. This result suggests that the relative humidity in the atmosphere is the most important factor to determine the warm rain occurrence during the transition season.

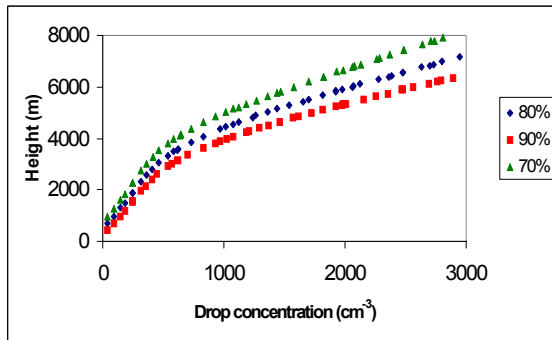


Figure 2. Warm rain height as a function of drop concentration for three different humidity at the ground.

Figure 3 shows the modification on the altitude of warm rain formation when different vertical velocities are imposed to the model. Velocities are set to 0.20, 0.30, 0.50, 1.00, 2.00, 3.00, and 5.00 m/s. As we can see, at low vertical ascents (up to 2.0 m/s) drop concentration does not increase beyond 2000 cm^{-3} . It means that, at low vertical velocities, the maximum supersaturation reached in the parcel air permits only a small fraction of the CCN to be activated. Certainly, this fraction represents the largest nuclei in the spectrum. When higher vertical velocity is imposed, a higher fraction is activated, producing higher concentration, which, in its turn, leads to the suppression of warm rain formation.

5. MODELED EFFECTIVE RADIUS AND MODAL RADIUS WITHIN A PARCEL MODEL

As we can see in Figures 5 the relation between the modeled liquid water content (LWC) and r_{effe} in the lower part of the graph follows an approximately 3-power law (with correlation up to 0.9998), although an exponential profile can also be applied (with lower correlation, in this case ~ 0.9766). As drop concentration increase the spectrum becomes more peaked. As a consequence, the exponent power law deviates from the 3 values, growing with the increasing concentration of finest portion of the CCN and according to the nucleation of new small droplets.

Above certain point, the relation between r_{effe} and liquid water content deviates quickly from the 3-power law trend due to non-linear nature of warm rain formation process. From the Figures we can also see that, according to the drop concentration, the values of $r_{\text{effe}} = 14 \mu\text{m}$ is attained only for relatively high values of liquid water content in the case of polluted clouds.

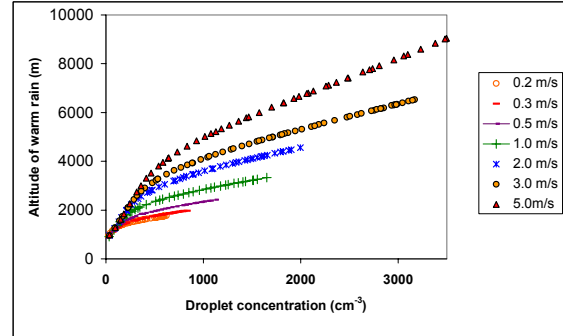


Figure 3. Altitude of warm rain formation as a function of cloud droplet concentration for different vertical velocities.

The behavior of the modal radius does not have the same depiction in the parcel model. In Figure 4, for example, the values of effective radius and modal radius are extremely well correlated up to $14 \mu\text{m}$ in the blue ocean case, despite the scattering of data due to the bin nature of the radius representation. After this interval the values of effective radius increase while the modal radius saturate. The phenomenon is reproduced in other simulations cases, except that, as drop concentration increase, the fast growth in the effective radius occurs with lower values of modal radius. This results are supported by the work of Freud et al (2005) in which modal radius do not shown an undefined growth tendency as that demonstrated by the effective radius.

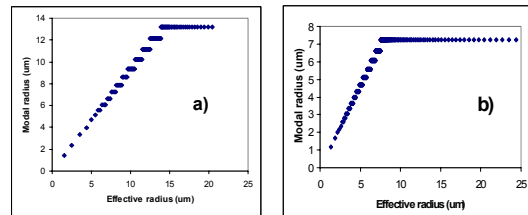


Figure 4. Comparison between the modal radius and r_{effe} for four two situation simulated by the parcel model (a) Blue ocean; and (b) Polluted.

A proposition for a curve to represent the relation between the effective radius and the liquid water content obtained with the parcel model is also proposed and show in Figure 5. It was represented according to the relation

$$LWC = \frac{N_1}{2} \left[1 + \tanh^{-1} \left(r_{\text{effe}}^{\frac{2}{3}} - c_1 \right) \right] \quad (2)$$

which fits well the behavior of r_{effe} from 3 to $20 \mu\text{m}$. In (2), $N_1(g/m^3) = 4,7761 \ln(N) - 22,241$, and $c_1(m^{2/3}) = 10,186 N^{-0.1205}$, where N is the mean drop concentration in cm^{-3} .

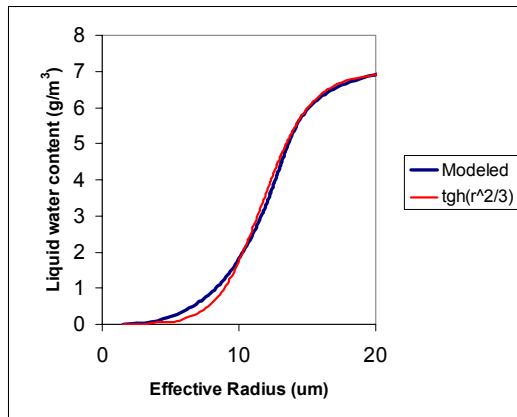


Figure 5. Parameterization of LWC as a function of r_{effe} (see text for more details).

This result can be applied to evaluate the depth a cloud from the four different types of air masses found during the SMOCC/EmfiN!/LBA need to extend for generating the first precipitation embryo necessary to produce warm rain.

Figure 6 depicts the values of observed effective radius according to the cloud depth for four regimes observed during the SMOCC/EmfiN!/LBA experiment. Points represents observations, while lines represents the values obtained from the parcel model.

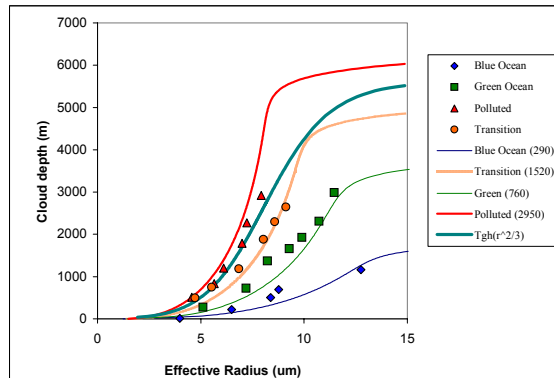


Figure 6) Comparison of modeled and observed effective radius as a function of the cloud depth. Points represent observations, while lines represent modeled values and the parameterization proposed in equation (1). Also is indicated the modeled mean droplet concentration. The $tgh(r^{2/3})$ line was obtained for a droplet concentration of 2000 cm^{-3} , which is close to the mean observed values for the polluted environment.

The warm rain height value found for the blue ocean again agrees with radar observations in Fortaleza, which shows plenty of rain development in clouds with top at or below 3000 km. The value found for the Green Ocean is well below the freezing level, which again allows warm rain to develop. Clouds in the transition regime are apparently in a borderline situation, as the warm rain height is located close to the 0°C isotherm. Because super cooled water is a common feature in

convective clouds, one might suppose that rain may develop in the warm phase already during the dry to wet transition due to the increase ability of the population of large particles to collect the small particles. Finally, in the polluted environment, at the freezing level the droplets grew only to about two third the threshold for warm rain initiation. Nevertheless, due to the nature of the relation between r_{effe} and LWC, it is probable that, in some special cases, the ice-phase processes still have not started. Namely, special condition means low vertical velocities, which implies small fraction of activated CCN, and high relative humidities in the environment

6. BIDIMENSIONAL MODEL RESULTS

In order to check this hypotheses mentioned above we apply a bidimensional model described in Costa et al. (2000). The model is axis-symmetric, vorticity based, and anelastic, and it permits the microphysics to interact directly with the cloud dynamics. As the skill of this model in reproducing aircraft data was proved before, with particular attention to the fine structure of the clouds, we decide to use the same thermodynamics situation applied in Costa et al. (2000) to the different microphysical environments investigated here. This is done not only because it avoids the necessity of validating the model but also because this situation is suitable for the investigation we are interested in carry out.

Figures 7 to 10 depict the modeled values of cloud water content and rain water content after 18 minutes simulations for two cases studied here. As we can see, gross disparities occur in the values of cloud water and rain water content. The simulations also suggest that both dynamical and microphysical processes are altered at cloud edges.

In both situations high cloud water mixing ratio ($> 3 \text{ g/m}^3$) were found, particularly between 2 and 2.5 km levels. In the upper portion of the cloud of the green ocean case, the cloud water content was significantly depleted, mainly due to the conversion to rainwater. Despite the prevision for the polluted case, although scarce rain water is still present.

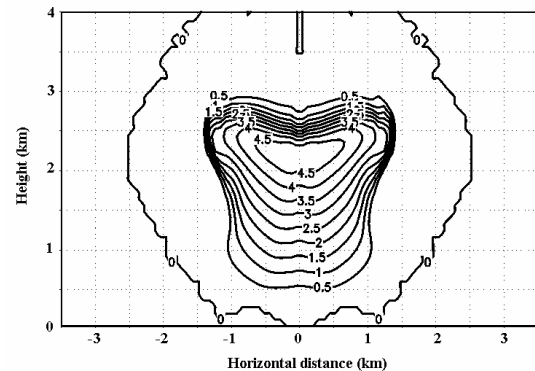


Figure 7. Cloud water content for a green ocean after 18 minutes simulation. Values in g/m^3 .

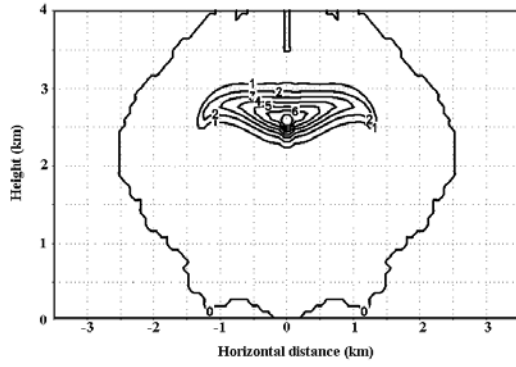


Figure 8. Rain water content for a green ocean after 18 minutes simulation. Values in g/m^3 .

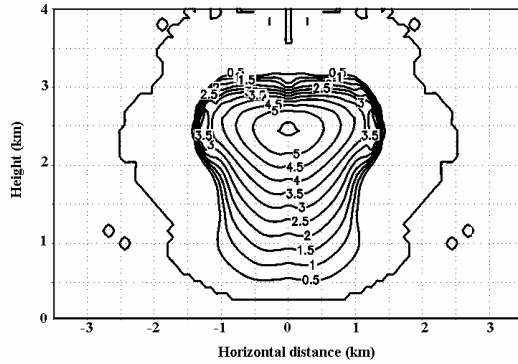


Figure 9. Cloud water content for a polluted environment after 18 minutes simulation. Values in g/m^3 .

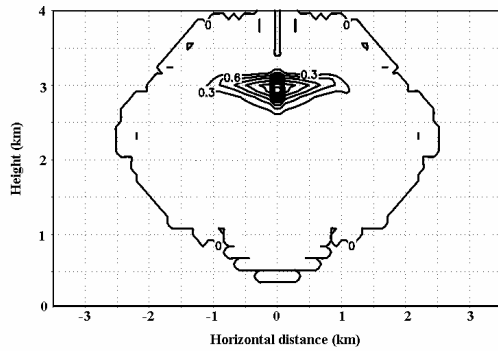


Figure 10. Rain water content for a polluted environment after 18 minutes simulation. Values in g/m^3 .

In fact, observations of droplet spectra show that low amount of drizzle is produced even on very polluted environment. In Figure 11 we can observe some drops with sizes around $200 \mu\text{m}$ present at 2500 m above cloud base.

Pollution also affects the r_{effe} and aerosol load effect can separate from the cloud depth effect. Figures 12 and 13 depict two different simulated situations (a clean marine and a polluted continental). It is clearly see that for a given cloud depth, the cleaner the environment the larger the effective radius. It can also be seen that the values of r_{effe} are generally increasing with increasing height for all regimes. In the Figures the majority of

the points that deviates from the main cloud points shown are found in the lateral or cloud's top. These points represent values of reduced liquid water content and low droplet concentrations, most of them generated by detrainment.

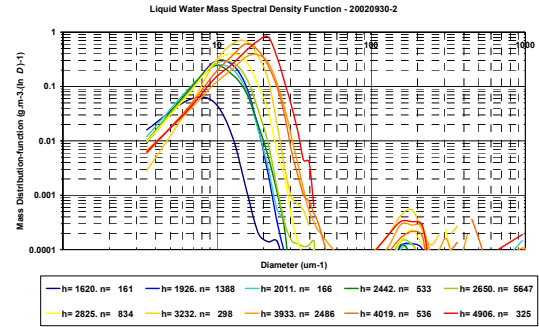


Figure 11. Droplet spectra observed on 30th September 2002. Heights and number of observations are marked in different colors.

In the simulations, values of r_{effe} are correlated to rain water appearance. In the clean case, there are lots of points of r_{effe} above $14 \mu\text{m}$, while in the polluted case just a few of them.

Table 1 shows some physical characteristics of the simulated clouds, for different microphysical environments. The values of maximum droplet concentration obtained in simulations for the four cases are close to the values found for the CCN concentration at 0.5% (which corresponds to values of 230 cm^{-3} , 254 cm^{-3} , 990 cm^{-3} , and 2010 cm^{-3}). When we compare the concentrations values obtained with the bidimensional model and those obtained with the parcel model we can verify that, although the ascending speed can be of about the same values during the maximum updraft in the bidimensional model, the values of concentration are higher in the parcel model.

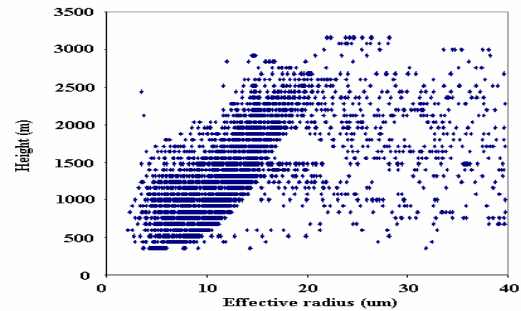


Figure 11. Simulated values of r_{effe} for a clean marine environment.

Andreae et al. (2004) used the dependence of the droplet diameter of the modal liquid water (D_L) on the height to estimate the warm rain height formation. There they defined the height where the modal radius attains a value of $12 \mu\text{m}$ as the warm rain height formation. Applying this methodology in our model to estimate the depth necessary to produce warm rain height for the four situations simulated here we found 1252.6 m for the blue ocean case; 1441.9 m for the green ocean case; 2470.3 m for the transition situation; and

3620.7 m for the polluted one. The values are in very close agreement with observation for the blue ocean (1200m), the green ocean (1600m), and the transition one (2200m); but disagree with that one of the polluted case (5000m).

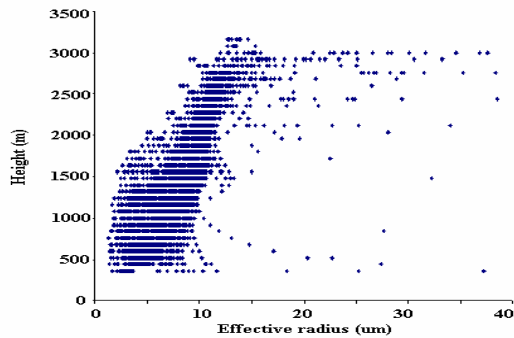


Figure 12. Simulated values of r_{eff} for a clean marine environment.

The model result is inconsistent in the polluted case. Although the cloud has produced some amount of rain, the value found is higher than the maximum cloud depth. Moreover, the use of a linear profile to represent the dependence of modal radius on height is questionable. When applied to the transition case, for instance, a linear profile produces a correlation of 0.73 while a second order polynomial representation gives 0.78 (Figure 13). In the polluted situation, the values tend to diminish, but the polynomial representation is still better than the linear one (0.75 against 0.70). This suggests that, as pollution increase, increasing also the cloud droplet concentration, as worse is the representation of the modal radius behavior on height with a linear profile.

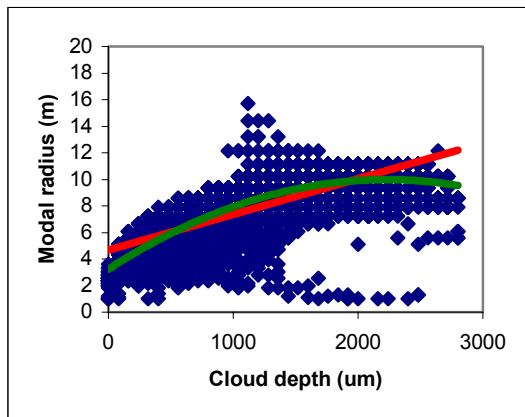


Figure 13. Modal radius as a function of height for a transition situation. The red line is a linear profile ($r_{mod} = 0.00267z + 4.68415$) while the green line represents a Polynomial profile ($r_{mod} = -1E-06x^2 + 0.0061x + 3.2351$). See text for more explanation.

7. CONCLUSIONS

This work presented results from numerical models with detailed treatment of liquid phase microphysical process, focusing on changes in the precipitation initiation over the Amazon due to

the presence of aerosols generated by forest burning.

It was shown that the parcel version of the model is able to reproduce the observed values of effective radius.

Comparisons of results from 3 sets of simulations have shown the environment humidity play a significant role in changing the process of rain initiation. It was shown that a mere 10 % change in the relative humidity may change the warm rain height by as much as 1 km, for concentration greater than 1000 cm^{-3} . This can be crucial factor to determine whether a population of clouds (namely, the ones with tops not too above the 0° C isotherm) will be able to produce precipitation or not.

When different velocities are imposed to the vertical ascent in the parcel model, the altitude on the warm rain formation is significantly modified even when relatively low concentration of CCN are imposed. It means that, at low vertical velocities, the moisture available on the air permits only a small fraction of the CCN to be activated. In this case, the fraction represents the largest nuclei in the spectrum. As higher vertical velocity are allowed to the parcel, as higher the activated fraction. This leads to higher droplet concentration, which, in fact, leads to the suppression of warm rain formation.

Environment	Blue ocean	Green ocean	Transition	Polluted
Maximum updraft (m/s)	5.86	5.87	5.88	5.84
Maximum downdraft (m/s)	2.55	2.58	2.63	2.65
Maximum cloud water mixing ratio (g/m^3)	4.82	5.37	6.38	6.59
Maximum rain water mixing ratio (g/m^3)	9.62	8.09	3.62	2.75
Maximum droplet number concentration (cm^{-3})	287	390	1226	2158
Maximum cloud top height (mb)	701.8 (3160 m)	701.8 (3160 m)	695.1 (3240m)	695.1 (3240 m)
Maximum cloud base height (mb)	966.5 (440 m)	966.5 (440m)	966.5 (440 m)	966.5 (440 m)
Maximum precipitation rate (mm/h)	31.23	23.09	7.38	3.71

Table 1. Some characteristic found during bidimensional simulations of the four microphysical regimes found during the LBA/EMfin!/SMOCC-2002 experiment

Bidimensional simulation, nevertheless, contrast with parcel model and points out that, even in very polluted environments clouds with relatively

low vertical extension (~3 km cloud depth) can still produce some amount of rain. As the dynamics of this model is more complex and permits rain water to be depleted, it is certainly more close to reality.

Model results also point out that the modal radius can be used to evaluate warm rain height formation only when clean microphysical regimes are observed, namely, that ones in which cloud droplet concentration do not exceed 1500 cm^{-3} . The failure in determining the warm rain formation is due to the fact that modal radius doesn't show an undefined tendency to grow with higher altitude and, actually, tends to saturate inside the cloud core in high polluted environments. Although in some occasion simulation produces values that reach about $12 \text{ }\mu\text{m}$, these points of high values of modal radius in polluted situation are found in the cloud border. There, values of rain water content and drop concentration are generally low, which also evidences detrainment and strong evaporation. As evaporation is more pronounced in small droplets, the appearance of modal radius with values of $12 \text{ }\mu\text{m}$ are more probable due to evaporation instead to rain water formation.

The way representing the mechanism through which modal radius undergoes a modification from a linear profile with height to saturation is probably the explanation to the higher

correlation with a second order polynomial representation.

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