Response to Anonymous Referee #2

We thank Anonymous Referee #2 for the review, constructive comments and suggestions for improvement of our manuscript. Detailed responses to the individual comments (including additional information and figures from the revised manuscript) are given below.

Comments and suggestions:

Details: The authors use both expressions (pyro-)convective and pyro-convective seemingly interchangeably throughout the manuscript. A clarification of exactly what is meant by (pyro-)convective clouds in the beginning would be very helpful. To my understanding the parentheses are not appropriate because the results can not be generalized to convective clouds. The initial aerosol distribution which is applied is typical for biomass burning aerosol and neither such a high number concentration nor such a monomodal size distribution is typical under non-pyro conditions. By the same token I think the title of the article is misleading and should be adjusted for clarity

Page 8644, line7ff: "... at the base of pyro-convective clouds, we have performed cloud parcel model simulations assuming a mono-modal particle size distribution characteristic for young biomass burning aerosols. The dry particle size distribution is determined by an accumulation mode with a count median or geometric mean diameter of Dg = 120 nm, a geometric standard deviation of ←g = 1.5 (Reid et al., 2005; Janhäll et al., 2009),..." Here you clearly state that the initial aerosol size distribution is characteristic for biomass burning aerosol, but later you apply your findings to convective clouds in general. How does that fit?

Responses and Revisions

We have replaced the term (pyro-) convective with either pyro-convective or convective. With regard to generalization, we have performed additional tests with different size distributions (different D_g and σ) and all tests confirmed the

existence of the three regimes. In a mega-city study and a study of pristine rainforest aerosols, Su et al 2009 and Gunthe et al (2009b) also came to the same conclusion using the measured size distribution (as shown in Fig RS1 and RS2). This information is added in the revised manuscript as detailed below. More over, we clarified that only the concept of the three regimes (shape of isolines) can be generalized while the absolute levels of isolines are variable.

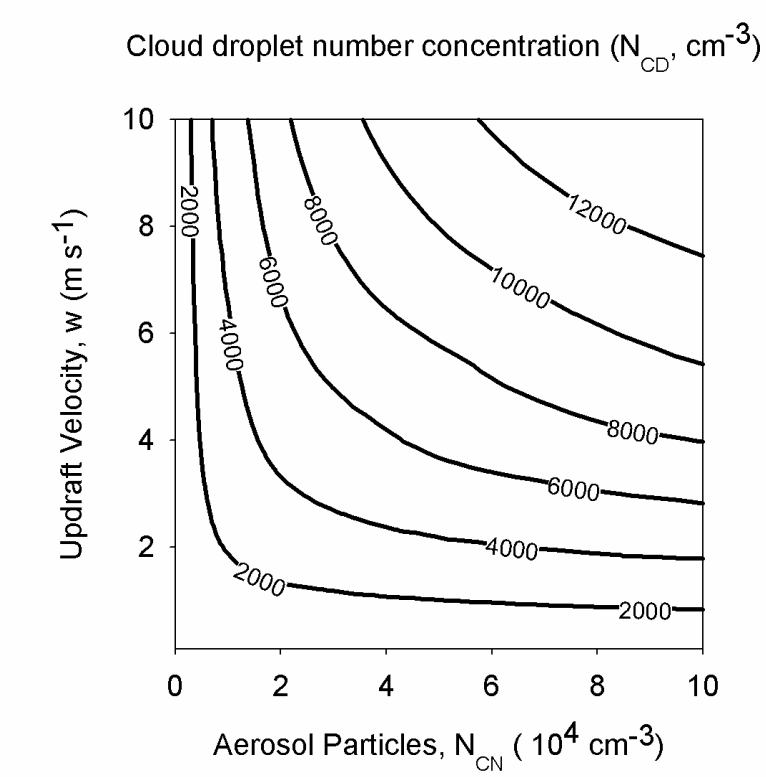


Figure RS 1 (in Response only). Cloud droplet number concentrations (N_{CD}, cm^{-3}; isolines) calculated as a function of updraft velocity (w, m s^{-1}) and initial aerosol particle number concentration (N_{CN}, cm^{-3}). The simulations are based on the averaged size distribution of Campaign CAREBEIJING2006 (Su et al, 2009)

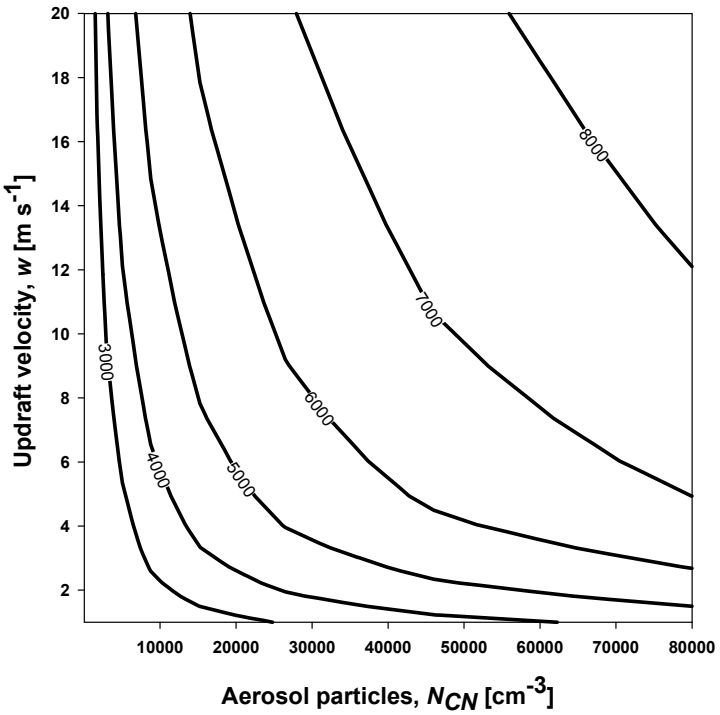


Figure RS 2 (in Response only). Cloud droplet number concentrations (N_{CD}, cm^{-3}; isolines) calculated as a function of updraft velocity (w, m s^{-1}) and initial aerosol particle number concentration (N_{CN}, cm^{-3}). The simulations are based on marine aerosols given by Seinfeld and Pandis with following fit parameters: *N1*=133 cm^{-3}, σ1=4.5394, Dp1= 8 nm, *N2*=66.6 cm^{-3}, σ2=1.6218, Dp2= 266 nm, *N3*=3.1, σ3=2.4888, Dp3= 580 nm

Page 8646 Line 4

'This was confirmed by sensitivity studies with different aerosol size distributions (D_g : 60 nm to 200 nm; σ_g : 1.2 to 2.0). Moreover, Su et al (2009) found the same type of regimes over w and N_{CN} in their simulations based on measured particle size distribution and κ values from mega-city Beijing and the same applied for pristine rainforest aerosols (Gunthe et al, 2009). Under these different conditions, the levels of isolines change but the shape stays the same. Just like in ozone chemistry, the existence of NO_x- and VOCs- limited regimes can be generalized but the quantitative relations have to be adjusted to different conditions.'

Comments and suggestions:

Page 8641, line15: "for symbols and parameter values see Sect. 2.2 and Rose et al.,2008a)." I also recommend providing explanations for symbols and parameters within the text of this paper.

Responses and Revisions

We added the following explanations.

Page 8641 line 15

'Seinfeld and Pandis, 2006; $D_{wet,c} = \frac{2A}{3\ln s_c}$ with $A = \frac{4\sigma_{sol} M_w}{RT\rho_w}$ using $s_c = s$; $D_{wet,c}$ is the critical droplet diameter, s_c is the critical water vapor supersaturation ratio, σ_{sol} is the surface tension of solution droplet, M_w is the molar mass of water and ρ_w is the density of pure water)'

Comments and suggestions:

section 2.2: I recommend giving the original citations for the Koehler theory. The same is true for the osmotic coefficient reference model.

Responses and Revisions

Original citation include.

Page 8641 line 21-26

'According to Köhler theory (Köhler 1936), the equilibrium water vapor saturation ratio s_{eq} is given by ……and an osmotic coefficient (OS) reference model (Robinson and Stokes 1959)'

Comments and suggestions:

Page 8641, line 24: ". . .we have tested two different approaches of describing the influence of aerosol chemical composition and hygroscopicity on aw. . ." Do you

mean you parametrized the chemical composition in form of kappa?

Responses and Revisions

Yes, we added references clarifying the approach.

Page 8641 line 24

'we have tested two different approaches of describing the influence of aerosol chemical composition and hygroscopicity on a_w: an effective hygroscopicity parameter (κ) Köhler model (Petters and Kreidenweis, 2007, Kreidenweis et al., 2009, Pöschl et al., 2009)'

Comments and suggestions:

Page 8642, line14, "...The hygroscopicity parameters of biomass burning aerosols range from 0.01 for freshly emitted smoke containing mostly soot particles to 0.55 for aerosol from grass burning, and the average value of _ in polluted continental air is 0.3±0.1 (Andreae and Rosenfeld, 2008; Rose et al., 2008b; Pöschl et al., 2009)...." Please provide the original citations for kappa of the freshly emitted smoke containing mostly soot particles. I assume you state these numbers here to explain why you used 0.2 as average value for kappa. Please clarify your argumentation.

Responses and Revisions

In the same paragraph, we introduced the κ-model and these sentences served as an introduction of the kappa values from measurement. As specified in the referenced study of Andreae and Rosenfeld (2008), from where we have taken the values, the κ values for freshly emitted biomass burning aerosols are based on unpublished data from M. O. Andreae (2007) and S. Kreidenweis (2007). It is specified in the revised manuscript.

Comments and suggestions:

Page 8642, line 20, "...For the simulation of real atmospheric aerosols (rural and biomass burning) we have used _= 0.2 and s = 1300 kg m-3...." Did you do the

simulations for rural and biomass aerosol or did you "add" biomass to the rural aerosol? If yes, what kind of initial size distribution did you use. Why did you use kappa 0.2 for rural aerosol and for biomass burning? Later you vary kappa, you should probably state this here.

Responses and Revisions

We removed the word, 'rural' and added a reference to clarify why kappa = 0.2 was chosen. It was taken from Rose et al. (2008b), who obtained this value from atmospheric CCN measurements of smoke during an intense biomass burning event.

Page 8642 line 20

'For the simulation of real atmospheric aerosols from biomass burning we have used $\kappa = 0.2$ (Rose et al., 2008b)'

Comments and suggestions:

Section 2.3: I appreciate the inclusion of the test of the kappa approach in a cloud parcel model, but still I have some questions: If I understand you correctly, the first part of your validation is the test of the kappa approach against the OS model in your cloud parcel model. Did you use kappa 1.28? You do not specify this in the text. Could you also provide this test for species other than sodium chloride, e.g., mixed particles? Why did you limit yourselves to the size distribution specified by Segal and Kain, rather than testing for a wider range of size distributions? If you show some more tests here, this could justify your conclusion that kappa is suitable to describe atmospheric aerosol particles in a cloud parcel model.

Responses and Revisions

We show the use of kappa 1.28 in the text explicitly. To further test the model, we also compared a typical particle size distribution of urban aerosols. Both studies showed similar results. We added corresponding text in the revised manuscript.

Page 8643 line 15

'Additional tests with typical particle size distributions of urban aerosols (Su et al, 2009) showed similar deviations in S_{max} and N_{CD} for constant $\kappa = 1.28$ and no

deviations with $\kappa = f(\mu_s)$.'

Comments and suggestions:

What is the difference in Smax for the different model runs? You attribute the differences to the "simplifying assumptions" of the kappa approach. What are these? Could the differences also be caused by the application of the surface tension of water at 25 C.I am wondering what causes the shape of Ncd in Figure 1b for the kappa approach. Do you have an explanation for the clear deviation at low levels? Did you also check your results against the alternative cloud parcel model for the kappa and the OS approach? It could be interesting to show/discuss these results as well.

Responses and Revisions

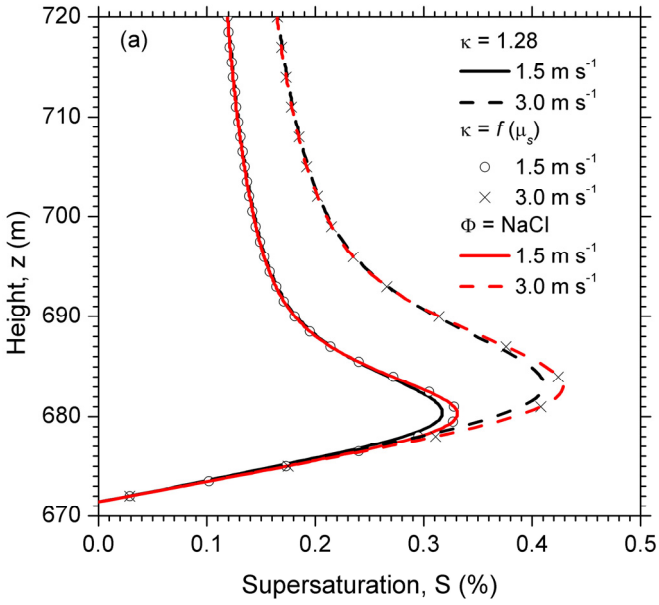
In the revised version, detailed explanations were added and simulations with molality-dependent kappa values were added (as shown below). The deviation at low levels is caused by numerical oscillations in the non-activated part of the particle size spectrum. The oscillations have been addressed in the model development paper of Simmel and Wurzler 2006. It is caused by an overestimation of the concentration gradient within the bins. The numerical oscillations can be removed by increasing a weighting coefficient for the redistribution of mass between the size bins up to 1, but this would lead to an underestimation of the droplet numbers. To give sufficient numerical stability and avoid the underestimation of droplet numbers, a coefficient of 0.6 was used in our simulation and specified in the manuscript.

Page 8643 Line 24

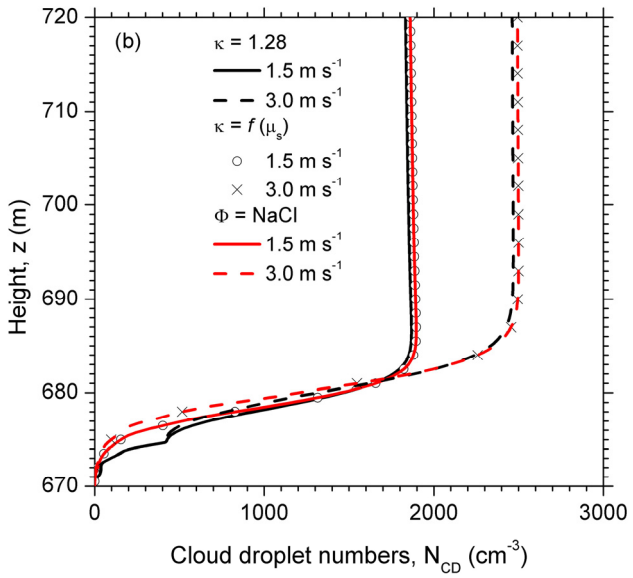
'The differences were caused by the different approaches of a_w calculations. To make a_w the same, i.e. $a_w(OS) = a_w(\kappa)$, we get the expression of κ as a function of μ_s, eq (6). Fig. 2 showed the dependence of κ on μ_s for sodium chloride.

$$\kappa = f(\mu_s) = (\exp(\nu_s \Phi_s \mu_s M_w) - 1)\frac{V_w}{V_s} \qquad (6)$$

After implementing $\kappa = f(\mu_s)$, eq (6), the κ-Köhler model produced the exact same results as the OS reference model (Fig. 1), demonstrating the equivalence of the two model formulations.'



(a)



(b)

Figure 1. Exemplary vertical profiles of **(a)** water vapor supersaturation (S, %) and **(b)** cloud droplet number concentration (N_{CD}, cm^{-3}) simulated with different Köhler model approaches: osmotic coefficient model (red lines), κ-Köhler model with constant κ (black lines) and κ-Köhler model with μ_s dependent κ (open circle and cross). The updraft velocity was set to w = 1.5 m s^{-1} (solid lines or open circle) or w = 3.0 m s^{-1} (dashed lines or cross), and the initial aerosol particle number concentration was set to N_{CN} = 3000 cm^{-3} with particle properties as specified by Segal and Khain (2006).

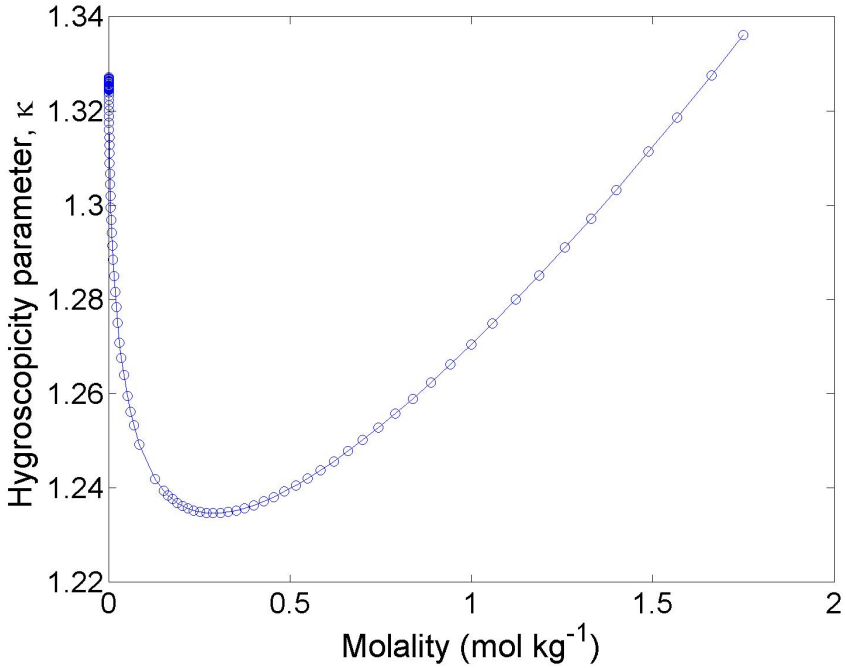


Figure 2. Dependence of hygroscopicity parameter κ as a function of solute (NaCl) molality. The expression $\kappa = f(\mu_s)$ can be found in Sect. 2.3, eq (6).

Comments and suggestions:

Page 8646, line 6:". . .This is due to the fairly similar CCN properties of aerosols in most regions of the world (Andreae and Rosenfeld, 2008; Rose et al., 2008a; Gunthe et al., 2009) and confirmed by sensitivity studies with different aerosol size distributions (not shown) and effective hygroscopicities (Sect. 3.2). . ." What do you mean by fairly similar CCN properties? How do you define CCN properties? If you mean hygroscopicity, which you vary between 0.001 and 0.6 in your simulations, the

CCN properties are definitely not "fairly similar". I would like to see the missing sensitivity studies on the aerosol size distribution or at least some numbers on the influence. If you want to generalize your modeling results to convective clouds, this would be a helpful tool.

Responses and Revisions

We deleted the statement '*This is due to the fairly similar CCN properties of aerosols in most regions of the world (Andreae and Rosenfeld, 2008; Rose et al., 2008a; Gunthe et al., 2009) and confirmed by sensitivity studies with different aerosol size distributions (not shown) and effective hygroscopicities (Sect. 3.2).due to the fairly similar CCN properties of aerosols in most regions of the world (Andreae and Rosenfeld, 2008; Rose et al., 2008a; Gunthe et al., 2009)*', and added sensitivity studies with different aerosol size distributions in the revised manuscript as detailed below.

Page 8648, line 7

'Based on cloud parcel model simulations, Feingold (2003) proposed a linear regression method to calculate the relative sensitivity of one parameter (i.e. cloud-top effective radius) against the other parameters (variables). McFiggans et al (2006) have also used this method to calculate the sensitivities of cloud droplet concentrations on other parameters, defining sensitivity $S(X_i) = \partial \ln \mathrm{N_{CD}} / \partial \ln X_i$ where X_i is the investigated parameter affecting $\mathrm{N_{CD}}$, i.e., $\mathrm{N_{CN}}$, $\mathrm{D_g}$, σ_g, w or the mass fraction of ammonium sulfate ε as a proxy of particle hygroscopicity. To calculate, for example, $S(\mathrm{N_{CN}})$ they plotted all values of $\mathrm{N_{CD}}(\mathrm{N_{CN}}, \mathrm{D_g}, \sigma_g, w, \varepsilon)$, i.e., $\mathrm{N_{CD}}$ calculated as a function of variable values of $\mathrm{N_{CN}}$, $\mathrm{D_g}$, σ_g, w and ε against $\mathrm{N_{CN}}$ on a log-log scale. Then a linear regression was applied and the slope of the fit line was taken as $S(\mathrm{N_{CN}})$.

When this method was applied in the supersaturation-quenched regime where $\mathrm{N_{CD}}$ approaches zero (in case of very high $\mathrm{N_{CN}}$ and/or $\mathrm{D_g}$ values), $\ln \mathrm{N_{CD}}$ values

approaching $-\infty$ can strongly influence and distort the slope of the linear fit. To avoid this problem, we used modified method in which all values of N_{CD} calculated at a given value of X_i were averaged prior to fitting. Then the averaged values of N_{CD} were plotted against X_i on a log-log scale and linearly fitted. Since this method gives averaged values of relative sensitivities, we denote it with $\overline{S}(N_{CN})$.

In the sensitivity studies, we used the same model setup and input parameters as detailed above (Sect. 3.1) to investigate three cases in different regimes, i.e. one in updraft –limited regime (w=5 m s^{-1}, N_{CN}= 8x10^4 cm^{-3}), one in aerosol-limited regime (w=15 m s^{-1}, N_{CN}= 1x10^4 cm^{-3}) and another one in the transitional regime (w=10 m s^{-1}, N_{CN}= 5x10^4 cm^{-3}). For each of the three investigated combinations of w and N_{CN} we varied the size distribution and hygroscopicity parameters as follows: D_g = 60-200nm, σ_g = 1.2- 2.0, and κ = 0.005-0.6.

As shown in Tab. 1, $\overline{S}(X_i)$ is positive for D_g and κ in all the regimes. This is because larger particles or more hygroscopic particles have a lower critical supersaturation. Across all regimes of CCN activation, the sensitivity of N_{CD} against particle size, $\overline{S}(D_g)$, is two to three times higher than the sensitivity against chemical composition, $\overline{S}(\kappa)$. This is consistent with the relative sensitivity of s_c on D_g and κ, in which $(\partial \ln s_c / \partial D_g)/(\partial \ln s_c / \partial \kappa) = 3$. The sign of $\overline{S}(\sigma_g)$ is negative because the tail of the distribution at large sizes results in activation of larger droplets, reducing supersaturation and N_{CD} values. For both $\overline{S}(D_g)$ and $\overline{S}(\kappa)$ the deviations between the aerosol-limited, transitional and updraft limited scenarios agreed to within +/- 15%. In contrast, $\overline{S}(\sigma_g)$ in the aerosol-limited scenario was by a factor of 1.8 lower than in the transitional and updraft-limited scenarios. The sensitivities determined in our case study for the aerosol limited regime are very similar to the values reported by McFiggans et al 2006 for their polluted case (3000 $cm^{-3} > N_{CN} > 1000$ cm^{-3}). ’

Table 1: Relative sensitivities of N_{CD} on X_i, $\overline{S}(X_i) = \partial \ln \overline{N_{CD}} / \partial \ln X_i$ (where X_i is one of D_g, σ_g and κ).

	Aerosol-limited regime (S_{max} > 0.5%)	Transitional regime (S_{max} = 0.2-0.5%)	Updraft-limited regime (S_{max} < 0.2%)
D_g	0.39	0.45	0.32
σ_g	-0.50	-0.91	-0.92
κ	0.15	0.17	0.13

The three regimes are **(a)** aerosol-limited regime (w = 15 m s^{-1} and N_{CN}=1x10^4 cm^{-3}); **(b)** transitional regime (w = 10 m s^{-1} and N_{CN}=5x10^4 cm^{-3}); **(c)** updraft-limited regime (w=5 m s^{-1} and N_{CN}=8x10^4 cm^{-3}). The ranges of X_i are D_g : 60nm to 200nm, σ_g : 1.2 to 2.0, hygroscopicity κ : 0.005 to 0.6.

Comments and suggestions:

Page 8646, line 15-17: Here you connect low updraft velocities to biomass burning. What does low mean in this context? line 22-23: Isn't this circular argumentation? And how high are "very high updraft" velocities in this regime?

Responses and Revisions

In line 17, we referred to biomass burning under conditions of shallow convection, 'in shallow convection of polluted air over locations or regions with strong sources of aerosols such as biomass burning'. We deleted the word 'very' for consistency with other parts of the manuscript.

Comments and suggestions:

Page 8647, line 1: "on the other hand" should be replaced by e.g., however or in contrast. Line 6, "...high concentrations of small cloud droplets.." Has not been

shown that the droplets are small. Second paragraph: I think this whole paragraph is very speculative, and no data are presented to back this claim. The argumentation should be strengthened or omitted. Line 19-20: The activated aerosol is only scavenged from the atmosphere when it precipitates.

Responses and Revisions

We replaced 'on the other hand' by 'however' and removed the second paragraph. We corrected the statement about the scavenging as shown below,

Page 8647 Line 19

'When an aerosol particle is activated to a cloud droplet, the remaining fraction of the aerosol particles is transported as interstitial aerosol in the pyro-cloud. Unless they are scavenged by impaction with hydrometeors, they will be released into the atmosphere in the outflow region of the pyro-cloud, which can be as high as the upper troposphere or the lower stratosphere (e.g., Fromm et al., 2005).'

Comments and suggestions:

Page 8648, line 9: A citation is needed for the tested kappa range from 0.001 to 0.6.(compare also to page 8636 line 27). Line 11: provide citation for kappa = 0.3 line14- 16: The list of citations seems rather biased; further in the text the citations should be noted behind the numbers to which they belong.

Responses and Revisions

According to the journal guidelines for manuscript preparation, references are not permitted in the abstract (http://www.atmospheric-chemistry-and-physics.net/submission/manuscript_preparation.html).

In the revised manuscript, we have added citations as requested and shortened the text as detailed below.

Page 8648 line 9

'κ was varied from 0.005 to 0.6, covering the full range of effective

hygroscopicity parameters reported for CCN in continental air (Sect. 2.2, Andreae and Rosenfeld, 2008 and references therein).'

Comments and suggestions:

Page 8649, line 18: Here I disagree from the authors' opinion that NCD depends only weakly on kappa, e.g., in Fig. 6c an increase in droplet number from 11000 to 15000 over the range of kappa from 0.1 to 0.6 is found, and a 30% increase can hardly be called weak (analog Fig6b with an increase of 10%).

Responses and Revisions

The statement was mainly based on the facts that 6-fold increase in κ (from 0.1 to 0.6) causes not more than a 10-30% increase in N_{CD}. However, in the revised manuscript, we change 'weakly' into 'not strongly' .

Comments and suggestions:

Page 8650, line 3-5 and 17-18: Is a 10-nm variation in the geometric mean diameter a "realistic change"? If you want to generalize your results to convective clouds, a 10-nm variation is too small.

Responses and Revisions

In the revised manuscript, we added sensitive studies in the D_g range of 60 –200 nm (as detailed above) and included information from and references to related studies.

Page 8646 line 4

'This was confirmed by sensitivity studies with different aerosol size distributions (D_g = 60-200 nm; σ_g =1.2-2.0). Moreover, Su et al (2009) found the same type of regimes over w and N_{CN} in their simulations based on measured particle size distribution and κ values from mega-city Beijing and the same applies for pristine rainforest aerosols (Gunthe et al, 2009a,b).

Comments and suggestions:

Page 8651, line 15: ". . .particle composition and hygroscopicty. . .." Isn't the hygroscopicity of the particles based on their composition? You could replace the "and" with "expressed as". Line 18-22: You treat kappa 0.3 as proven, but this number is based on a limited number of field campaigns and should be looked at with caution. Further a deviation of 50% is quite a lot for the atmospheric relevant regime (S<0.1%).

Responses and Revisions

We clarified that the relation between composition and the hygroscopicity parameter κ.

Page 8651 line 15

'Coarse mode particles and the variability of particle composition expressed through the hygroscopicity parameter κ appear to play important roles only at very low supersaturation in the updraft-limited regime of CCN activation (in particular at S $\leq$ 0.1%).'

In the revised manuscript, we removed the value of 0.3. We agree that deviations of 50% and more are substantial, which is why we concluded : 'Thus, we suggest that further experimental and theoretical studies of CCN activation and cloud droplet formation should be focused primarily on the updraft-limited regime, low water vapor supersaturations and potential kinetic limitations of CCN activation.'(Page 8651 line 24)