# Response to Anonymous Referee #1

We thank Anonymous Referee #1 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.

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**Comments and suggestions:**

*Both size distribution and hygroscopicity initialization of the model are oversimplified if the goal is to describe pyrocumulus clouds. It is incorrect to assign a single k-value to the entire size distribution or to assume a well-behaved size distribution so close to an active source. Clearly size resolved mixing state will be important to characterize CCN activity. Fresh emissions often contain sub-100 nm particles organic with inhomogeneous inorganic inclusions (Posfai et al., 2003, JGR) and particles larger than 150 nm size are often sooty agglomerates Chakrabarty et al. (2006, JGR). It is therefore logical that k-values should vary significantly with particle size.*

**Responses and Revisions**

We agree that the $\kappa$ values would vary with particle size. The problem is that our current model can not utilize size-resolved $\kappa$ values and we would address this with a modified model. Also as mentioned in the manuscript, Page 8650, line 26, *'For this purpose, we also suggest and intend to apply models that enable assigning different hygroscopic properties to accumulation mode and coarse mode particles, as the latter are likely to be less hygroscopic.'* Anyway, our sensitive studies suggest that $N_{CD}$ does not strongly depend on $\kappa$ under pyro-convective conditions.

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**Comments and suggestions:**

*Size distributions very close to fires are highly variable and will depend on cooling and dilution rates. Number concentrations in the plume are so large that coagulation occurs on the time-scale of updrafts.*

*The role of the particle size distribution in droplet formation is not adequately addressed. Only a typical size distribution is assumed and particle size distribution parameters are not systematically varied. It is particularly important to note the relative sensitivity between size and hygroscopicity for CCN number concentration which is well known from Kohler theory, i.e. dlnk/dlnD = -3. This sensitivity is approximately preserved for calculations of droplet number concentration (see McFiggans et al., 2006, ACP, Table 1, using sensitivity to soluble fraction as surrogate for k-values). It should be clarified in the text that a near equivalent axis with mode diameter can be added to the hygroscopicity axis in Figure 6. The role of the size distribution should be more explicitly acknowledged.*

**Responses and Revisions**

We agree that the size distribution is important, and we added sensitivity studies of particle size using the method of McFiggans et al 2006 and Feingold 2003. Equivalence of $D_g$ and $\kappa$ are also addressed in the revised manuscript, as detailed in the following text.

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., $\mathrm{N_{CN}}$, $\mathrm{D_g}$, $\sigma_g$, w or the mass fraction of ammonium sulphate $\varepsilon$ 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 $\mathrm{N_{CN}}$, $\mathrm{D_g}$, $\sigma_g$, w and $\varepsilon$ 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 $N_{CD}$ approaches zero (in case of very high $N_{CN}$ and/or $D_g$ values), $\ln N_{CD}$ 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 $\overline{S}(\mathrm{N_{CN}})$.

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 $s^{-1}$, $N_{CN}$= 8x$10^4$ $cm^{-3}$), one in aerosol-limited regime (w=15 m $s^{-1}$, $N_{CN}$= 1x$10^4$ $cm^{-3}$) and another one in the transitional regime (w=10 m $s^{-1}$, $N_{CN}$= 5x$10^4$ $cm^{-3}$). For each of the three investigated combinations of w and $N_{CN}$ we varied the size distribution and hygroscopicity parameters as follows: $\mathrm{D_g}$ = 60-200nm, $\sigma_g$ = 1.2- 2.0, and $\kappa$ = 0.005-0.6.

As shown in Tab. 1, $\overline{S}(X_i)$ is positive for $\mathrm{D_g}$ and $\kappa$ 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, $\overline{S}(\mathrm{D_g})$, 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 $\mathrm{D_g}$ and $\kappa$, 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 $\overline{S}(\mathrm{D_g})$ 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 $D_g$, $\sigma_g$ and $\kappa$).

| | 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 |
| $\sigma_g$ | -0.50 | -0.91 | -0.92 |
| $\kappa$ | 0.15 | 0.17 | 0.13 |

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

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**Comments and suggestions:**

*It is not easy to suggest a way out of this dilemma; one possible way forward is to leave the notion of size distribution k-values behind and use the more parcel-model-native input of supersaturation activation spectra, i.e. cumulative CCN concentration as a function of supersaturation (e.g. Gunthe et al., 2009, ACPD, Fig. 9). Such spectra could be generated from a range of size distributions an assumed hygroscopicity behavior. Once the spectra are generated they can be scaled to the desired aerosol number concentration and then droplet number concentration can be calculated*

**Responses and Revisions**

The cumulative CCN concentration as a function of supersaturation ($N_{CCN}=f(S)$) is equivalent to the size-resolved $\kappa$ method. However, the $N_{CCN}=f(S)$ method depends on the function *f*, and the goodness-of-fit. As shown in Table 4 of Gunthe et al., 2009, ACPD, the deviations between the measured CCN and the $N_{CCN}=f(S)$ method (using power law approaches, $N_{CCN,S}=N_{CCN,1}*S^k$) are much larger than the deviations with the constant $\kappa$ methods.

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**Comments and suggestions:**

*The other part of the manuscript is a sensitivity study of droplet number concentration to various input parameters. A lot of prior work is available in the literature, and the basic sensitivities of model to input parameters are well understood. Although some of this work is cited in the manuscript it is not properly discussed.*

*The sensitivities that are found in this manuscript should explicitly be compared to values reported previously (especially McFiggans et al., 2006, Table 1). It is important to note whether increasing aerosol number concentrations to those expected in pyrocumulus clouds truly changes the known sensitivities or not. Also there should be explicit comparisons to Rissman et al. (2004, JAS) who define regions in the sensitivity space where composition (chemical effects, i.e. k-values and surface tension) trump updraft velocity (dynamical effects).*

*For the more general discussion in the manuscript about aerosol and updraft limited regimes the authors should use published data to back up their claims. For example empirical correlations between droplet number concentration and aerosol number concentrations have been obtained in field campaigns and fitted to power law expressions based on Twomey theory (see Ramanathan et al. 2001, Science, Figure 5 for a not so recent review).*

**Responses and Revisions**

We followed the referee's suggestion adding comparisons and discussions about the previous studies, i.e. McFiggans et al 2006 (Page 8648, line 7, see responses above) and Ramanathan et al 2001a and 2001b (shown below). For the work of Rissman et

al., 2004, we have referred to it in the discussion about the chemical effects (Page 8649, line 27).

Page 8646, 23

'In comparison to our model results, the observed aerosol-cloud droplet relations reported by Ramanathan et al. (2001a) would lie at the border between updraft-limited and transitional regimes of CCN activation (Fig. 7), corresponding to updraft velocities around ~0.5 m s-1. Note, however, that the reported relations are characteristic for marine rather than pyro-convective conditions, which may limit the comparability. Model investigations using typical marine aerosol properties are under way and will be presented and discussed elsewhere (Gunthe et al., in preparation). Nevertheless, first results indicate that the regimes are not very different.'

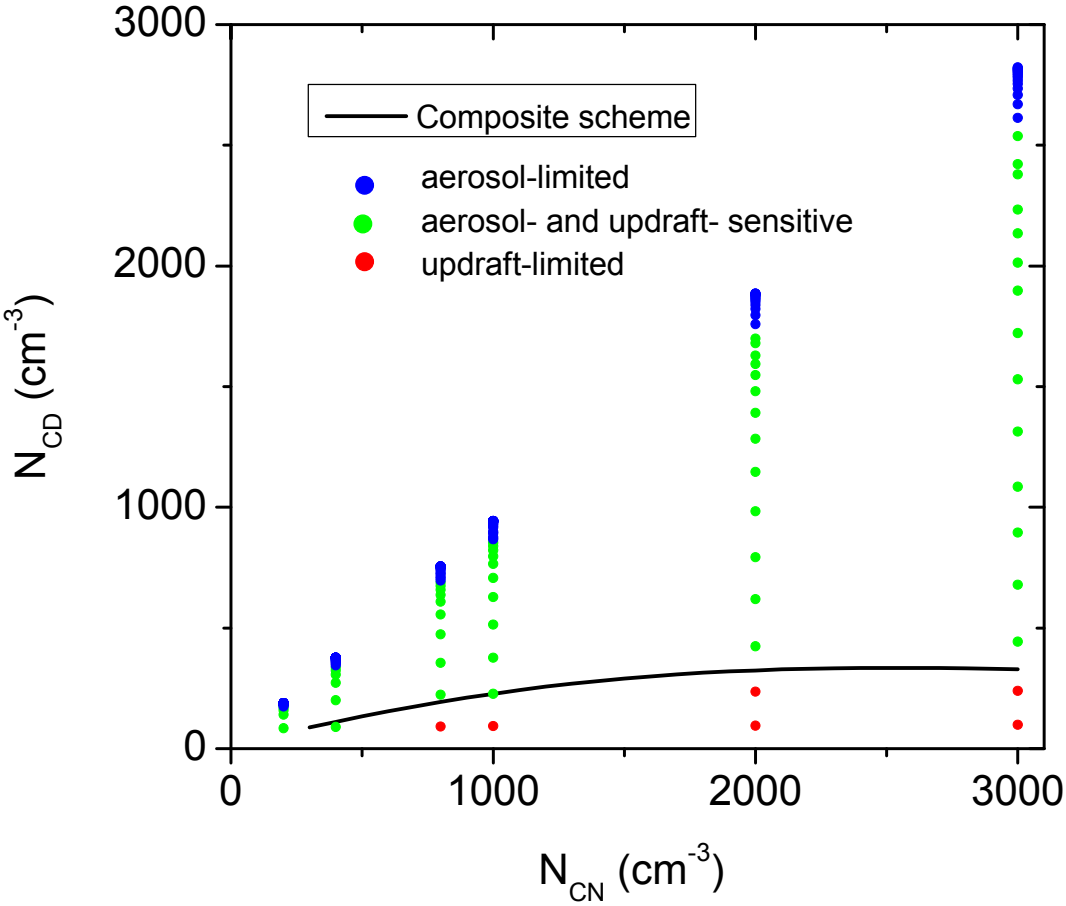


Figure 7. The thick line labeled 'Composite scheme' is obtained from a composite theoretical parameterization that fits the INDOEX aircraft data for the Arabian Sea

(Ramanathan et al 2001a, and equations for the line can be found in the Appendix A of Ramanathan et al 2001b). The colored points are obtained from the same cloud parcel model simulations as shown in Fig. 4. The colors indicated the regimes determined by the $w/N_{CN}$ ratio.

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**Comments and suggestions:**

*The relative roles of updraft and aerosol number concentration have been addressed by Twomey in his 1977 book Atmospheric Aerosols. Based on his analytical approximation he derived that Ncd –$C^{2/(a+2)}w^{3a/(2a+4)}$, where C is the cumulative CCN concentration active at 1% supersaturation and a is the slope of CCN supersaturation spectrum in log-log space. Note that was originally denoted k but that k is conflicting with hygroscopicity here. The parameter C can loosely be interpreted as accumulation mode number concentration. Twomey stated that empirical values are a- 0.5 and thus Ncd / $C^{0.8}w^{0.3}$ for typical ambient conditions. It is interesting to note that in this description the basic sensitivity is independent of aerosol number concentration, contrary to what is claimed in the manuscript, but only a function of , which is determined by the particle size distribution and to some extent hygroscopicity.*

*Regardless the functional form of the analytical estimate suggest that Figures 2 and 3 should be presented in log-log space; if the basic sensitivity of the system does not change, the isopleths should be linear. Thus non-linear isopleths will reveal if the relative sensitivity of the system changes or not and help to better define where this change occurs. It should be noted in the figure caption and the text that the sensitivities presented do specifically exclude size effects, which will have an effect on a, and thus the slope of the isopleths.*

**Responses and Revisions**

According to equation $N_{CD} \propto N_{CCN,1}^{2/(\alpha+2)} w^{3\alpha/(2\alpha+4)}$, $N_{CD}$ strongly depends on $N_{CCN,1}$, with exponent of 0.8, which was also shown in the book of Twomey (1977).

So we are not sure what the referee meant here.

To test the applicability of the Twomey approach, we added a figure with log-log scale and the following discussion.

Page 8646 line 4

'In an earlier study, Twomey (1977) described the roles of updraft and aerosol number concentration by an analytical approximation of where $N_{CCN,1}$ is the cumulative CCN concentration at 1% supersaturation, w is the updraft velocity and is the slope of CCN supersaturation spectrum in the log-log scale. For a fixed size distribution, is proportional to $N_{CN}$ used in this study. According to the analytical approximation, the $N_{CD}$ isolines (isopleths) should be linear in a log-log plot. Thus we also plotted the $N_{CD}$ isolines on a log-log scale as shown in Fig. 4 b. For either aerosol-limited or updraft-limited regimes, the isolines can be approximated by linear lines with different slope. However, over the whole range of $N_{CN}$ and w, the isoline is obviously non-linear. This means the Twomey approximation with fixed is not applicable over the whole investigated range of conditions.'

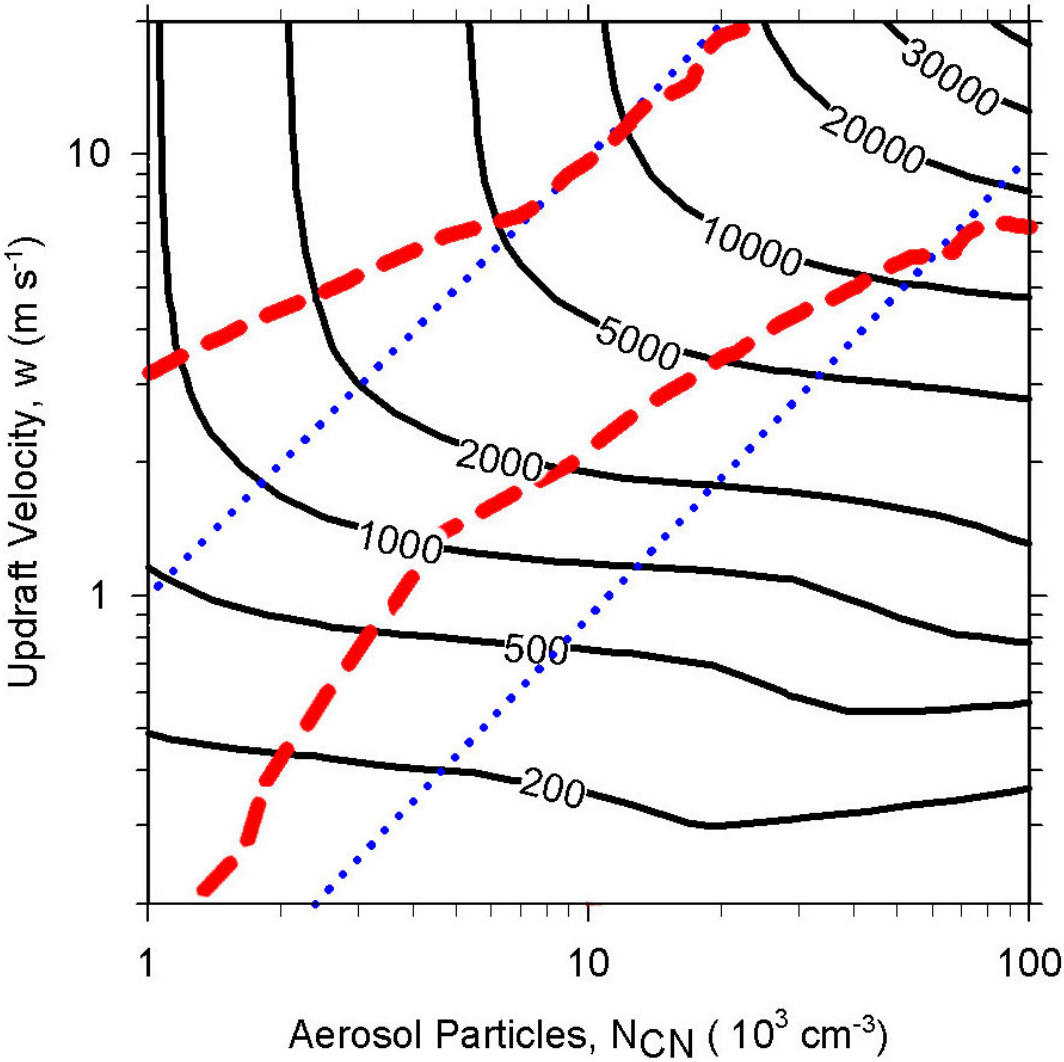


Figure 4. Cloud droplet number concentrations ($N_{CD}$, $cm^{-3}$; isolines) calculated as a function of updraft velocity (w, m $s^{-1}$) and initial aerosol particle number concentration ($N_{CN}$, $cm^{-3}$). (a) normal scale; (b) log-log scale. Red dashed lines indicate the exact borders determined by $S(N_{CN})/S(w) = 4$ or 1/4, between different regimes. Blue dotted lines indicate approximate borders defined by $w/N_{CN}$ ratio between different regimes.

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**Comments and suggestions:**

*Pg 8643: The model shows a difference between the k-value approach and the OS1 model for sodium chloride. The authors attribute this difference to simplifying assumption in the k-Kohler model. Although this is possibly true, I don't think that this is supported by the text. The value k = 1.28 (sigma = 0.072 J/m2 and T = 298.15)*

*for sodium chloride is tuned to reproduce the critical supersaturation predicted from AIM water activity at sc = 0.3% (see Table 3 in Kreidenweis et al., 2005; Dd = 49.3 nm and sc = 0.3% from AIM). A -1.3% relative deviation from the predicted supersaturation occurs when this value is applied at 1% critical supersaturation (Dd = 22.3 nm and sc = 1% from AIM). This is one of the limitations of the k-model since only one parameter is available to fit the data. However, for a fair comparison between the k-model and OS1 model, the predicted critical superaturations by the two Kohler models need to be compared first. Since the OS1 model is allowed to vary surface tension with composition, and the k-model does not, and since the OS1 model and AIM may not agree on the water activity, it is a bit unfair to blame the k-model itself for the discrepancy in the parcel model. Thus the authors should use a k-value that is consistent with the OS model (i.e. run the OS model offline and fit a k-value to the output) and then repeat the calculations shown in Figure 1 with that particular k-value. Only then can the remaining discrepancy be attributed to the simplifying assumptions in the k-model rather than simply differences between the Kohler models.*

**Responses and Revisions**

In the revised version, simulations with molality-dependent kappa values and detailed explanations were added (as shown below).

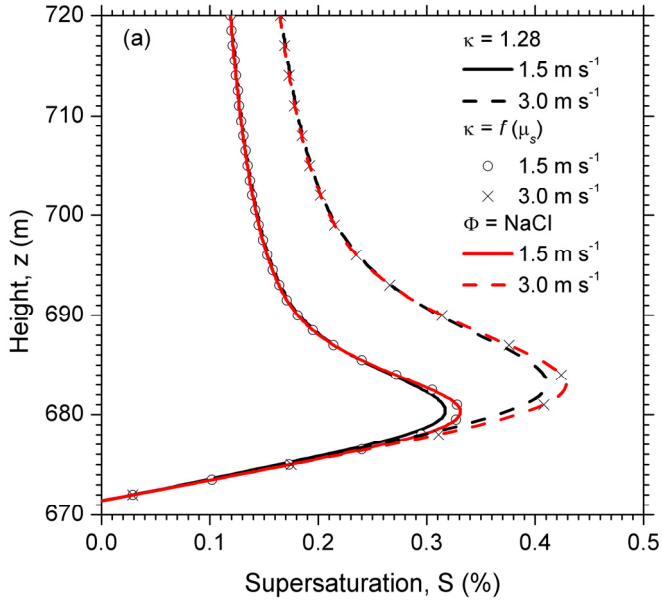
Page 8643 line 13

'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 $\kappa$ as a function of $\mu_s$, eq (6). Fig. 2 showed the dependence of $\kappa$ on $\mu_s$ for sodium chloride.

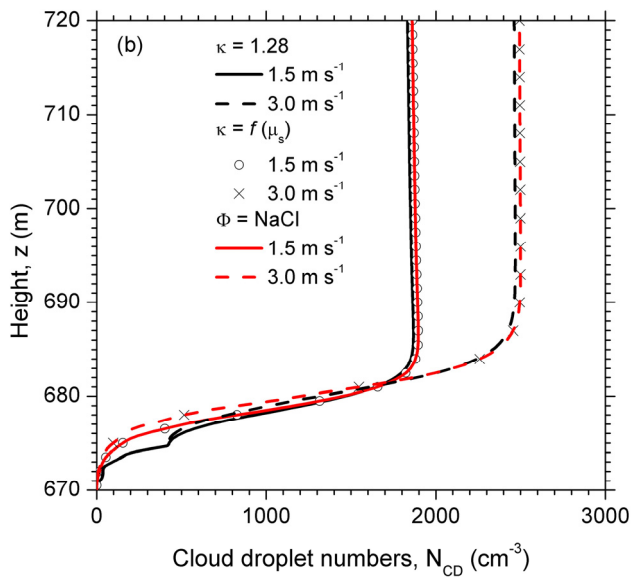
$$\kappa = f(\mu_s) = (\exp(\nu_s \Phi_s \mu_s M_w) - 1)\frac{V_w}{V_s} \qquad (6)$$

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 $\kappa$ (black lines) and κ-Köhler model with $\mu_s$ dependent $\kappa$ (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).

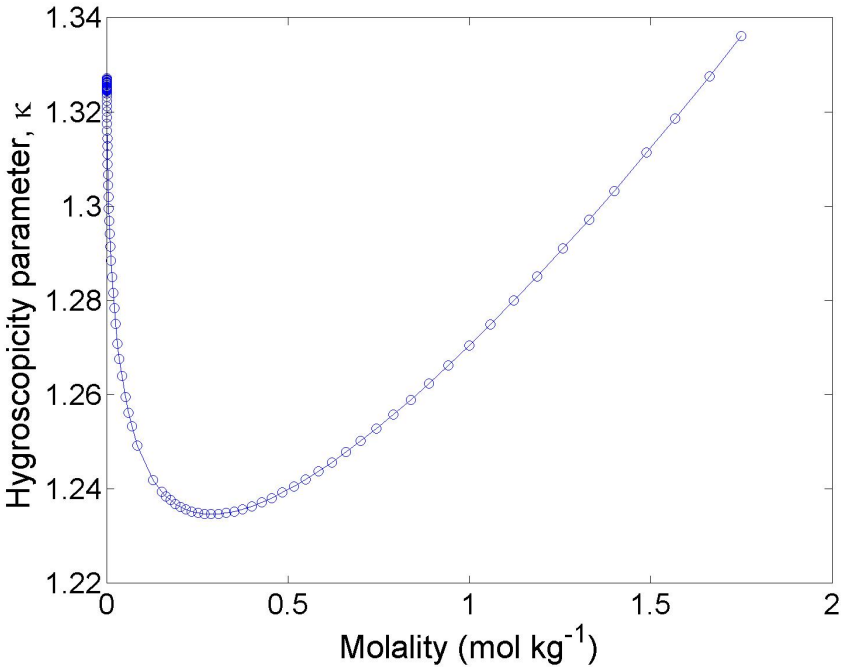



Figure 2. Dependence of hygroscopicity parameter $\kappa$ as a function of solute (NaCl) molality. The expressioin $\kappa = f(\mu_s)$ can be found in Sect. 2.3, eq (6).

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**Comments and suggestions:**

*Pg. 8648: 'The global average value of k in continental air is 0.3' – I object to call this a global average value since it is based on a few field campaigns only. I do not believe that we have all the information yet to make such a claim. It certainly is a reasonable value for large-scale grid-averaged hygroscopicity over the continents, but I am not sure that it is applicable near sources where heterogeneity and mixing state becomes more important. Further, recent studies from some of the authors indicate that values in the Amazon are lower than this 0.3 value. Even in this study this value is not really used, as stated in pg. 8642 : 'For the simulation of real atmospheric aerosols (rural and biomass burning) we have used k ↑ 0.2'. Over the continents the range 0.1 < k < 0.5 seems more realistic than promoting a single value*

*for all landmasses. I suggest to state the assumed k-value once and then focus on the model results rather than trying to tie it the assumed value too much to real world aerosols which is inevitably subject to criticism.*

**Responses and Revisions**

We deleted the statement about the global continental average of $\kappa$ in the revised manuscript.

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**Comments and suggestions:**

*I am surprised to see that water supersaturations are achieved even when aerosol number concentration become large. When too many particles are present, and these particles are allowed to take up water hygroscopically, the supersaturation can be quenched by hygroscopic growth alone before reaching saturation. It would be worthwhile to show where this limit lies or why the model used in this paper does not have such a limit. The reason to explore this limit is because the reported sensitivities will break down near that limit, which states that Ncd = 0, even in convective clouds. If such model runs occurred but were removed it will also influence the explored sensitivities reported here.*

**Responses and Revisions**

In our simulations, the lowest value of w was set to 0.25 m $s^{-1}$. Under this condition, the supersaturation was not quenched even for $N_{CN}=10^5$ $cm^{-3}$, but the depression of $N_{CD}$ have already been seen from a log-log scale plot (Fig. 4b). We also tested lower updraft velocities to achieve full quenching of supersaturation and added this information in the revised manuscript.

Page 8645, line 15

'Under extremely low w/$N_{CN}$ conditions, water would be taken up by aerosol particles, the supersaturation would be quenched and $N_{CD}$ would drop to zero. E.g. for w = 0.1 m s-1 , $N_{CD}$ would still be increased by adding more particles up to $N_{CN} \approx 1 \times 10^5$ $cm^{-3}$, but would drop to zero at $N_{CN} \approx 2 \times 10^5$ $cm^{-3}$. This phenomenon can be found in the lower right corner of Fig 4 b. '