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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol- and updraft-limited regimes of cloud droplet formation: influence of particle number, size and hygroscopicity on the activation of cloud condensation nuclei (CCN)" *by* P. Reutter et al.

Anonymous Referee #1

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This is an interesting study that is of interest to the readers of ACP. The manuscript consist of two main parts, modeling of pyrocumulus and exploring sensitivities of drop number concentration to parcel model inputs. It seems to me that the pyrocumulus part of the study is a bit too oversimplified. I believe that the authors need to pay more attention to the size resolved mixing state of the aerosol since they are modeling a near source phenomenon. The sensitivity study is solid but it needs to be better placed in the context of the available literature. I have outlined my criticisms in detail below. If the authors can address these comments I recommend this manuscript for publication

in ACP.

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. 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. Thus the assumption of a clean size distribution with a single k-value that does not evolve during adiabatic ascent is not a realistic one. 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.

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 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 $Ncd \propto C^{2/(\alpha+2)} w^{3\alpha/(2\alpha+4)}$, where C is the cumulative CCN concentration active at 1% supersaturation and α is the slope of CCN supersaturation spectrum in log-log

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space. Note that α 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 $\alpha \sim 0.5$ and thus $Ncd \propto C^{0.8}w^{0.3}$ 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 α, which is determined by the particle size distribution and to some extent hygroscopicity. There have been many good objections put forth against this analysis over the years, in particular the problem that it fails in aerosol limited regimes because the power law assumed in the CCN activation spectrum has no upper limit (e.g. Cohard et al., JAS 1998). 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 α, and thus the slope of the isopleths.

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.

The sensitivities that are found in this manuscript should explicitly be compared to val-

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ues 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).

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

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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.

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 $k \sim 0.2$'. 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.

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.

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