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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 #2

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The paper presents a study on the initial cloud number concentration formed in pyro-convective clouds. A cloud parcel model with detailed spectral cloud microphysics is used for the prediction of the initial cloud number concentration. A sensitivity study concerning the cloud droplet concentration dependence on the initial aerosol number concentration and the updraft velocities is presented. Based on these two parameters, three different regimes of cloud droplet formation are defined: aerosol-limited, updraft-limited and aerosol & updraft sensitive regime.

In general the paper is well written and presents interesting results that are valid for

pyro-convective clouds, and an attempt is made to apply these results to convective clouds. I am not, however, fully convinced by the argumentation presented. The authors overgeneralize their results based on calculations done for what are clearly pyro-convective conditions (monomodal size distribution with a mean diameter of 120nm, high updraft velocities, etc.). Some sections of the paper use circular argumentation which results in confusion. The interpretation of the influence of hygroscopicity on cloud number concentration should be done more carefully. I, therefore, suggest a revision of the manuscript before publication.

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 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.

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

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?

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 κ 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.

Page 8642, line 20, “...For the simulation of real atmospheric aerosols (rural and biomass burning) we have used κ= 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.

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. 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.

Page 8644, line7ff: “... at the base of pyro-convective clouds, we have performed cloud parcel model simulations assuming a mono-modal particle size distribution characteris-

tic 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 σg = 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?

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.

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?

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.

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.

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%).

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.

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%).

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 8635, 2009.