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Interactive comment on "The effect of physical and chemical aerosol properties on warm cloud droplet activation" *by* G. McFiggans et al.

G. McFiggans et al.

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Reviewer comments are in bold

Additions/corrections are in italics

Responses to comments are in regular font.

Page numbering refers to the version of the manuscript dated 9/12/2005 in the document properties section of the PDF.

The authors have written an extensive and very interesting manuscript and address in many ways the need to have a review of aerosol effects on warm cloud formation. I would like to comment the authors on their work and effort that has gone into this manuscript!

After going through the document, I have noticed some important reference omissions and a few incorrect statements. These need to be corrected and addressed before the manuscript is publishable.

General comment: The paper does not consider numerous studies (e.g., Conant et al., 2003; Meskhidze et al., 2005) that focus on cloud-droplet closure, and the ability of cloud droplet formation parameterizations to predict cloud droplet concentrations in in-situ clouds. A relevant issue is quantifying the CCN prediction error with cloud droplet number error, and its relevance for the aerosol indirect effect (e.g., Sotiropoulou et al., 2006). Please include them.

Results from Conant et al. (2004) are summarized in Table 7, but are not cited in the bibliography. This omission is now corrected with Conant et al. (2004), plus the other studies presented in Table 7 (but not cited in the text), now in the corrected references.

The necessary additions to the references have been made:

Conant W. C., T. M. VanReken, T. A. Rissman, V. Varutbangkul, H. H. Jonsson, A. Nenes, J. L. Jimenez, A. E. Delia, R. Bahreini, G. C. Roberts, R. C. Flagan, J. H. Seinfeld: Aerosol-cloud drop concentration closure in warm cumulus, J. Geophys. Res., 109, D13204, doi:10.1029/2003JD004324, 2004

Snider, J.R., and J.-L. Brenguier: Cloud condensation nuclei and cloud droplet measurements during ACE-2, Tellus, 52B, 828-842, 2000

Yum, S.S., Hudson, J.G., and Xie, Y.: Comparisons of cloud microphysics with cloud condensation nuclei spectra over the summertime Southern Ocean, J. Geophys, Res., 103, 16625-16636, 1998

Meskhidze et al. (2005) was omitted from the discussion, as was Peng et al. (2006), since both deal with validation of a parameterized droplet activation model. In the revision we included mention of both Meskhidze et al. and Peng et al. The following is appended to last paragraph on pp. 8573:

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Excluded from Table 7 are closure exercises employing parameterized, as opposed to parcel model, droplet activation codes. As was mentioned (Section 3.2.2) such model validation exercises build on a successful comparison of measured and parcel-model-predicted droplet concentration values; examples of the former include Meskhidze et al. (2005) and Peng et al. (2005).

Meskhidze, N., A. Nenes, W.C.Conant and J.H.Seinfeld: Evaluation of a new cloud droplet activation parameterization with in situ data from CRYSTAL-FACE and CSTRIPE, J. Geophys. Res., 110, D16202, doi:10.1029/2004JD00573, 2005

Peng Y., U. Lohmann, R. Leaitch: Importance of vertical velocity variations in the cloud droplet nucleation process of marine stratus clouds, J. Geophys. Res., 110, D21213, doi:10.1029/2004JD004922, 2005

Since our manuscript was submitted (7 June 2005) before the release of Sotiropoulou et al. (2006) (6 November 2005), the latter was not cited or discussed.

Page 8521: In mentioning activation parameterizations for lognormal aerosol, the authors did not mention the Fountoukis and Nenes (2005) parameterization, which can consider an external mixture of lognormal aerosol concurrently competing for water vapor, as well as aerosol containing organic surfactants that depress surface tension and the water vapor accommodation coefficient. The authors also do not reference the Nenes and Seinfeld (2003) which can consider all the compositional complexities of the Fountoukis and Nenes (2005) formulation, but within a sectional aerosol framework. It should also be noted that Cohard and Pinty parameterization are developed for a generalized sigmoidal CCN spectrum, and not necessarily for lognormal aerosol alone.

The parameterizations of Nenes and Seinfeld (2003) and Fountoukis and Nenes (2005) are now referred to. Since we are not evaluating the parameterizations in this paper, we have not expanded.

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Theoretical and model-based analyses of activation in terms of lognormal size distributions have been reported by Cohard et al. (1998); Feingold (2001, 2003); Rissman et al. (2004); Abdul-Razzak and Ghan (2000); Nenes and Seinfeld (2003); Fountoukis and Nenes (2005); and experimentally determined (e.g. Martinsson et al., 1999).

Section 3.1.3: The work of Rissman et al., (2004) should also be referenced here, as it provides sensitivity ratios (calculated analytically) using a modified aerosol activation parameterization. The title of the paper itself indicates that the Twomey effect may actually decrease droplet formation, and this is shown clearly. Furthermore, Rissman et al. explore the effect of the organic fraction (solubility, surface tension depression), which is not done in Feingold (2003), and provide conditions where sensitivity of droplet number to fluctuations in organic variability can compete with dynamic variability in clouds.

Section 3.1.3: Discussion of Rissman et al. (2004) is now included. At first sight there is an apparent disparity in the conclusions drawn from this paper and from those of Feingold (2003) and Ervens et al. (2005). In Rissman et al., it is stated that the importance of composition is highest at high updraft velocities whereas Ervens et al suggest that at high velocities, composition is relatively unimportant. The disparity is resolved when one realizes that the Rissman et al. measure of sensitivity to composition is normalized by the sensitivity to updraft velocity. Thus a high value of $\phi(\epsilon_0) = \epsilon_0/w\partial N/\partial\epsilon_0\partial N/\partial w$ (where ϵ_0 is the composition parameter) doesn't necessarily mean that $\partial N/\partial\epsilon_0$ is large but that it is large relative to $\partial N/\partial w$. Thus, the conclusion that $\phi(\epsilon_0)$ in Rissman et al. is more important at high updraft doesn't tell the whole story since we don't know about the absolute values of $\partial N/\partial\epsilon_0$ and $\partial N/\partial w$. At high w, supersaturation tends to be high and therefore an increase in w does not add many more particles ($\partial N/\partial w$ is small); most are activated anyhow. The appearance of $\partial N/\partial w$ in the denominator means that $\phi(\epsilon_0)$ is large. But at high S, activated fractions are already high so the composition effects are not important in an absolute sense. Both studies do however agree that N is more sensitive to the size parameter r_g, and that N is more sensitive to r_g under

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polluted conditions.

Rissman et al (2004) performed a more detailed analysis of the effect of various composition factors such as solubility and surface tension, as well as size distribution parameters. Their results were derived from analytical solutions, and presented in terms of a sensitivity relative to the sensitivity of drop number concentration to updraft velocity $\phi(\chi) = (\chi/w)(\partial N_d/\partial\chi)/(\partial N_d/\partial w)$*, where* χ *is a composition factor such as organic mass fraction* ϵ_o*. The authors concluded that when defined this way, sensitivity to composition factors* $\phi(\chi)$ *is highest for aerosol typical of marine condition, and increases with increasing updraught velocity. However, these are conditions under which supersaturation and activated fractions are high, and an increase in* w *does not add many new drops (*$\partial N_d/\partial w$ *is small). The appearance of* $\partial N_d/\partial w$ *in the denominator tends to increase* $\phi(\chi)$*. Thus at high* S*, even though* $\phi(\chi)$ *is large, composition effects may not be important in an absolute sense,. Because the individual sensitivities (*$\partial N_d/\partial\chi$ *and* $\partial N_d/\partial w$ *or their logarithmic equivalents) were not reported, it is difficult to compare their results to those of Feingold (2003) for overlapping parameter space. Both studies do however agree that* N_d *is more sensitive to the size parameter* r_g*, and that* N_d *is more sensitive to* r_g *under polluted conditions. The greater sensitivity of cloud droplet number to size compared to composition illustrates that the aerosol size must be captured as a primary pre-requisite. The sensitivity to the compositional complexities should only be investigated in the knowledge that the size and number information is likely to be equally important (or moreso). It should be noted that the treatment of composition does not address the sensitivity to composition changes with size and to varied composition at any one size; evidence for the prevalence of both being provided in the forthcoming sections. The sensitivity of activation and cloud droplet number to more detailed aspects of aerosol composition is discussed in Section 4.2*

Page 8538: It should be noted that a major difference between some CCN instruments is that some * count * droplets that form, while others * infer * CCN spectra.

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The methodologies of counting versus that of Hudson (1989) are contrasted on p. 8537 of the draft manuscript. This is expanded in the following way:

..With the exception of the technique developed by Hudson (1989) CCN activation spectra are obtained from step-wise scans of the imposed supersaturation and presented as cumulative distributions. In the former technique the size distribution of activated droplets is measured and used to derive the differential CCN activation spectrum.

Page 8539: A representative value is 10 s and is consistent with growth times in many CCN instruments. Although experimentally. This statement is not exactly true. The groups that use continuous flow chambers typically examine the effect of exposure time on CCN activation, and examine whether or not the exposure time biases the CCN concentrations.

To rehash, our assertion is that a mismatch between growth times (CCN instrument) versus characteristic times for peak supersaturation (cloud updraft) has not yet been thoroughly investigated as a possible reason for CCN measurement error. The reviewer counters that continuous flow CCN instruments are being used to probe this issue; no references are provided. Along these lines we note that results are available from studies performed using a static thermal gradient diffusion chamber (Oliveria and Vali, 1995). In spite of this we feel that more work is needed and we return to this recommendation on p. 8589.

Page 8585: Abdul-Razzak and Ghan (2000) developed a parameterisation based on Kohler theory that can describe cloud droplet formation for a multi-modal aerosol. This approach has been extended by Nenes et al. (2001b) to include kinetic effects, such that the largest aerosols do not have time to grow to their equilibrium size. This statement is incorrect. First of all, the appropriate reference is Nenes and Seinfeld (2003) (and not Nenes et al., 2001!). Second, Nenes and Seinfeld (2003) did not extend the Abdul-Razzak approach, which is based

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on determining maximum supersaturation by fitting parcel model simulations to non-dimensional groups through non-linear regression. Nenes and Seinfeld completely revisited the droplet growth problem and came up with a largely analytical treatment of the problem from scratch. Anyone reading the relevant papers would see the difference. Also, the Nenes and Seinfeld (2003) paper is for sectional, externally mixed aerosol with any kind of chemical composition. Surfactants are treated (figure 10 of Nenes and Seinfeld, 2003), and with the modifications of Fountoukis and Nenes (2005), film-forming compounds and slowly-growing CCN can be explicitly treated in both sectional (Nenes and Seinfeld, 2003) and lognormal (Fountoukis and Nenes, 2005) formulations of the parameterization. Page 8585: While the effect of surface-active organics and slightly soluble organics has recently been included in the parameterisation of cloud droplet formation by Abdul-Razzak and Ghan (2004, 2005), other effects of organics, such as their filmforming ability are not considered yet. Nenes and Seinfeld (2003), Rissman et al (2004) and Fountoukis and Nenes (2005) consider surface tension depression. Also, Fountoukis and Nenes (2005), hence, Nenes and Seinfeld (2003), can consider film-forming compounds (i.e., changes in accommodation coefficient). Please correct accordingly.

The text is now modified to reflect the contributions of Nenes and Seinfeld (2003) and Fountoukis and Nenes (2005) regarding organics, surface-active compounds, and film-forming compounds.

Activation of aerosol particles to form cloud droplets is one of the weakest links in estimates of the indirect aerosol effects. In order to treat cloud droplet formation accurately, the aerosol number concentration, its chemical composition and the vertical velocity on the cloud scale need to be known. Abdul-Razzak and Ghan (2000) developed a parameterisation based on Köhler theory that can describe cloud droplet formation for a multi-modal aerosol. Also, the competition between different aerosol species, such as sulphate and sea salt, (Ghan et al., 1998; O'Dowd et al., 1999) has recently been

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considered (Ghan et al., 2001). Nenes and Seinfeld (2003) derived a largely analytical solution to the equations for droplet activation. Their sectional treatment treats externally mixed aerosol of variable composition and includes kinetic growth limitations, and surface active species. As discussed in Section 4, chemical effects of the same order as the indirect effect were pointed out by Nenes et al. (2002) and as large as unresolved cloud dynamics by Lance et al. (2004). Thus, they need to be included in GCMs as well. The effect of surface-active organics and slightly soluble organics has recently been included in the parameterisation of cloud droplet formation by Abdul-Razzak and Ghan (2004, 2005). Other effects of organics, such as their film-forming ability are treated by Nenes and Seinfeld (2003) and Fountoukis and Nenes (2005).

General comment: Compositional effects on the size of Giant CCN (GCCN) can have some interesting impacts on cloud microphysics. The authors point out the work of Medina and Nenes, where film-forming compounds can potentially reduce the size of GCCN. To this effect, black carbon inclusions can also act in a similar manner (Nenes et al., 2002b), if important, a warming mechanism can decrease cloud drizzle and potentially enhance SW cooling.

Regarding giant CCN, we do not expand on the possible effect of black carbon inclusions in modifying drizzle formation since the abundance of giant CCN is so poorly known.

The following reference (p. 8538) is missing and has been added:

Sinnarwalla, A. M. and Alofs, D. J.: A cloud nucleus counter with long available growth time. J. Appl. Meteor. 12, 831-835, 1972

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 8507, 2005.