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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.

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Page 13 - "Whatever the reason, less water uptake at smaller sizes implies slightly less sensitivity of activation to updraught velocity, since a faster updraught that activates more particles will encounter particles that are less hygroscopic." This statement seems very complicated to me. Unless I am misinterpreting it, the intent is to say "smaller, less hygroscopic particles are less likely to activate."

Page 13 - "THE GFd of the intermediate hygroscopic background particles tend to be somewhat smaller than the dominant mode in continental polluted environments, indicating the hygroscopicity of the background particles in urban areas is reduced by the presence of less hygroscopic compounds." What does the second half of this

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sentence contribute that the first half doesn't?

Page 13 - "The readily hygroscopic background mode generally dominates continental polluted aerosols, though considerable fractions of marginally hygroscopic particles may be found in proximity to urban areas." Reference?

Page 13 - The sentence "Only a few HTDMA measurementsĚ" needs re-writing.

Page 13 - "The free tropospheic aerosol is dominated by readily hygroscopic particles." You have two measurements for relatively brief periods of time. You can not say that the free-trop aerosol is dominated by readily hygroscopic particles. You can say that the two periods of measurements indicate larger hygroscopic fractions in the free troposphere.

Page 14 - Why are the results of Aklilu and Mozurkewich (2004) and Carrico et al. (2005) not also included Tables 3 and 4?

Page 14 - "Ěquasi-internal mixture of sulphate, sea salt and organics (see ??)." ?

Page 15 - "Comparison of a measured activation spectrumĚ and can also reveal limitations of the measurements (ref,s)." I think that the term closure (or CCN closure) should be included in this discussion,. The discussion of error and closure is interesting (I believe that there is a paper by Jacobson that discusses this also). If the errors in the measurements are larger and similarly with the assumptions, then do we learn anything from "closure"?

Page 16 - "Since determination of requiresĚ" determination of what?

Page 16 - "Investigations planned for a large continuous flow instrument (ref.) should lead to a resolution of this issue." I think this statement is inappropriate for this paper. Either remove or re-write to say it "may contribute to this issue."

Page 16 - Section 3.2.1 discusses hygroscopicity factors. Now section 3.2.3 is discussing them again. I appreciate that there is a connection between CCN and GF's (the work of Brechtel and Kriedenweis should referenced w.r.t. this), but much of this stuff belongs in section 3.2.1. Only the connections between CCN and GF's should be discussed here.

Page 16 - "external mixing of DIFFERENT hygroscopic compounds results in distinct growth modes, whereas Ě This finding is consistent Ě" What is the reference for 'this finding'?

Page 16 - Numerous studies combining different methods indicate that these marginally hygroscopic particles are dominated by elemental carbon (refs.)Ě" What about OC? The discussions of the on-line measurements and section 3.2.1 suggest that OC might be responsible.

Page 16 - "The absolute value of hygroscopicity Ě will determine the critical supersaturationĚ" Some reference to the Kohler equation discussed earlier on would be useful here.

Page 17 - "Ě i) suppression of the number of solute molecules in solution in a droplet of given size," I think this is more a point of clarification. Is this truly suppression or merely a displacement that is being discussed here. In other words, are you saying that there are fewer molecules of solute because there are these other more-or-less insoluble molecules? Or is this suppression, whereby the presence of the less soluble material somehow truly reduces the fraction of the available molecules of solute going into solution? This terminology occurs in one or two other places in section 4 (e.g. near bottom of page 18 "This will reduce the surface tension and Kelvin term, but may alsoĚ), and it needs to be clarified. (Overall, I think that section 4 is very well done.)

Page 18 - "Bilde and Svenningsson (2004) postulated Ě when smaller than the fully dissolved wet size." When what is smaller? It seems obvious that any undissolved core will be smaller than the fully dissolved wet size.

Page 18 - "On balance, it may be considered that the effect of limited solubility is of

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limited relevance to real multicomponent atmospheric aerosol activation." I think that this statement may be valid when presented in the proper context, that is that limited solubility precludes that component from contributing to significant water uptake. On the other hand, the presence of such components reduces the overall growth rate of the particle. In a CCN chamber, this may make little difference, but in a updaught at cloud base the competition between the rate of cooling and the water uptake is important to the supersaturation, and the effect can be significant. Why are the references in section 4.1.3 and the summary discussion in section 4 (Comments on solubility effects alone, page 24) completely different?

Page 20 - "Atmospheric organics compounds forming expanded film coatings are not expected to affect the equilibrium timescales nor the equilibrium composition of the aqueous core." What about the condensation of products of terpene oxidation onto sulphate particles? Could this not be an exception to this statement? There is evidence for this from the Calspan chamber experiments a few years ago (Shantz et al., 2003).

Page 21 - "Ě when the droplets are smaller than about 20 m in diameter." micrometres?

Page 21 - In equation 13, you use  rather than i, as in equation 2. I think you should be consistent. Also see my comment above about equation 2.

Page 22 - "Figure McFiggans 4 showsĚ" Presumably this should be "Figure 11 from McFiggans showsĚ"

Page 24 - W.r.t. the suppression of "a" or  by organic films, I think that the paper of Medina, J. and Nenes, A. (Effects of Film Forming Compounds on the growth of Giant CCN: Implications for cloud microphysics and the aerosol indirect effect.J.Geoph.Res.,109, D20207, doi:10.1029/2004JD004666) is worthy of reference here.

Page 24-25 - The result of Johnson et al. (2005) is very interesting, and the relatively low temperature of volatilization (60oC) is especially curious. Perhaps this is a coating effect, contrary to the discussion in section 4.1.6?

Page 25 - “Only if instruments which probe the response of a particle to saturation ratio in the same way that the particle will be exposed to a changing water vapour field below and in cloud may be able to capture the real atmospheric activation behaviour. If HTDMA or CCN Ĕ fail to allow sufficient time for particle equilibrium, models based on rediction of equilibrium Ĕ will not be able to reconcileĔ” This may be true for HTDMA measurements and modelling, but I think it is not so clear for cloud modelling. According to the parcel models, particles in the base of clouds prior to the maximum supersaturation are experiencing a continuous increase in the supersaturation value. CCN measurements don’t tell us about what will happen in clouds. They do give us information on the rate that particles take up water, and they do allow us to validate or at least test our models so that we can better predict particle activation in clouds.

Page 25 - “A reduction in the surface tension alone will lead to a reduction in critical supersaturation and hence an increase in the number of activated droplets, all else being equal.” This may be true, but not in a linear sense. The reduction in surface tension will also increase the growth rates of the same particles, which will control down the maximum supersaturation. Thus, increases forecasted using equilibrium models will overestimate the increase in activated PARTICLES.

Page 25 - “Solubility was shown to reduce drop concentrations significantly for values less than about 25 g l-1Ĕ” I assume that this is still referring to the Ervens et al. (2005) paper. Shantz et al. (2003) demonstrated exactly that.

Page 26 - “Ĕwhen high van’t Hoff factors (i=3or5)Ĕ” Make sure the use of i vs  is correct.

Page 26 - “Clouds act as chemical reactors Ĕ” Ok, but the only discussion here is about the uptake of HNO3 by growing solution droplets, not about either aqueous-phase chemistry or the potential role of SO2 oxidation (e.g.) in affectng Nd. I suggest to remove this first sentence of 4.3 only because it adds nothing and confuses the message.

Page 27 - “Thus, at lowER temperatures, nitric acid Ě”

Page 27 - The point about equilibration and the effect on HNO3 uptake is an excellent one, as is the final point in the same paragraph about the air mass history. Page 28 - “Ěwater content of glutaric acid has been shown to take several hours.” Reference?

Page 30 - There is a lot of recent work on CCN closure, especially from groups at Georgia Tech and Caltech. The authors should be sure that they have up-to-date references.

Table 7 - It would be helpful to have the number of cases included in the closure. It is nice to see the mass accommodation coefficient identified here. What does examined mean here. My reading of these papers is that organics were not explicitly considered in the identified works.

Page 31 - “The marine aerosol-cloud system has frequently been assumed TO BE one OF the simplestĚ”

Page 32 - “Ěas expressed in influential papersĚ” What is the point of this statement? Also, the Charlson paper dealt with DMS and sulphate. I don’t ever recall it being brought up when sea salt alone is discussed.

Figure 16 - More unpublished data. This and its discussion must be removed or properly referenced.

Page 38 - “Cloud models Ě cannot be explained by Kohler theory aloneĚ” I disagree with this statement in so much as you can couple a Kohler theory-based parcel model with a mass transfer model (for the HNO3), add the HNO3 solute to the solubility term, and it works fine.

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