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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.

R. Leaitch (Referee)

Richard.Leaitch@ec.gc.ca

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Review of McFiggans et al. for Atmos. Chem. Phys.

General Comments This review confines itself to how the physical and chemical properties of the atmospheric aerosol impact the nucleation of cloud droplets.

The review appears to be a connection of pieces written by the large number of authors. The writing styles of segments differ in spots and some of the writing is occasionally a little verbose, but overall it is good and, with few exceptions, the messages are clear.

Connections are drawn among the various sections of the paper where appropriate, but there is some repetition. I find that some of the sections are more compelling than

other sections, e.g. section 4 is the most interesting to me.

This is not a comprehensive review, in so much as it deals with more recent advances, but it is authoritative. The most recent work included tends to be a little parochial, and I think it is important that the authors draw a line for “published” work to be included in this paper, e.g. revise the paper to include anything that has been published or accepted for publication before an appropriate date in 2005. Most importantly, this should not be a forum for unpublished work (further comments about this are below).

There are also a few technical issues (see my specific comments). My overall impression is positive, and with some revision this could be an excellent summary.

Specific Comments Page 1 - intro - the cloud lifetime effect is almost always attributed to Albrecht. However, Ou and Liou published a detailed modelling study in JGR dealing with precisely this issue about the same time as Albrecht.

Page 2 - “It is not the intent of this article to investigate the broad question of dynamical cloud systems, Ě The main purpose of this review is to identify one particular and important aspect of the aerosol indirect effect, namely, the properties of the aerosol which dominate their activation in warm clouds. This aspect Ě For example, the relationships between droplet number and aerosol number and between effective droplet radius and Na may be both directly probed.” These statements, first ignoring the dynamics and then saying that you can use the Na-Nd relationship to study the effect of aerosol properties on clouds are contradictory. If I am new to this field, then I might interpret this to mean that dynamics have little impact on Nd, when really the most important aspect of this is the interplay between the updraught and the droplet growth rates. This is properly discussed later in this review, but the intent should be properly explained in this particular.

Page 2 - “An increasing body of evidence suggests that the complexity of atmospheric aerosol may preclude realistic treatment of cloud formation using the levels of simplification incorporated intoĚ” I might argue that its less to do with the complexity of the

ACPD

5, S3664–S3669, 2005

aerosol and more to do with the complexity of the cloud dynamical systems that preclude this. Indeed, this review suggests the aerosol complexity may be insignificant (page 24 - “Taken together Ĕ is unlikely to be significant.”). But regardless of how you view this, I had to read this statement a few times to be clear about what it was trying to say. A re-write would help.

Page 2 - “Long-range transport Ĕ are not always free of polluted plumes Ĕ”. Is this the only way that pollution influences the remote areas of the NH?

Page 2 - “In addition, the rich variety of natural Ĕ can also lead to a level of complexity, not broadly appreciatedĔ” What is a level of complexity? “rich varietyĔ”?

Page 2 - “..to establish i) the domionant characteristics of THE atmospheric aerosolĔ”

Page 3 - “where i is the van’t Hoff factor, representing the number of dissociated ions per solute moleculeĔ”. Following the classic cloud physics tests (Mason, and Pruppacher and Klett; Seinfeld and Pandis avoid the van’t Hoff factor), this definition is incorrect. The van’t Hoff factor includes the departure from ideality. In your equation 3, either the “i” should be replaced by , or the s removed. This mistake is becoming common.

Page 3 - Remove “It is clear that” and simply start with “The atmospheric aerosol does not solely comprise Ĕ” Immediately afterward, why is it necessary to say “A relatively minor and straightforward modificationĔ” rather than “A modification to the Raoult termĔ”? I see no value to these adjectives.

Page 3 - In equation 6, the “S” should be “B”.

Page 4 - “Ĕ supersaturation (which is directly proportional to updraught velocity)Ĕ” This is only true until the droplets have grown large enough to have a significant impact on the rate of water uptake. W.r.t. the next sentence, it is the critical radius and not the critical supersaturation that is important. There is a time factor and merely achieving the supersaturation does not guarantee activation. The critical radius is mentioned

ACPD

5, S3664–S3669, 2005

further down in this paragraph. Also, you should explain why some large particles may not activate but are still considered as cloud droplets. This paragraph can be confusing and needs revision.

Figure 4 - although it may be obvious, you should say “the WATER saturation ratio.”

Page 5 - The Twomey parameterization was built to work for small k values, and evaluated in cumulus in the SH. It does not work as well for high k values.

Page 5 - I don’t understand the reference to Jones et al (1994) or Boucher and Lohmann (1995) in the context of “an increase in available CCN leads to a monotonic increase in cloud droplet concentrationĚ” Jones et al use an empirical result that demonstrates the often observed “saturation” at higher values of Na. Boucher and Lohmann also used an empirical result that exhibited a monotonic increase on a log-log plot, but not on a linear plot.

Page 7 - “recent paper from Arctic from Kulmala’s group”?

Page 7 - Table 1 is calculated here, not in Feingold (2003). You need to include the ranges that the results in Table 1 were calculated from. Also, what model was used - Feingold’s?

Table 1 - This result is very interesting. It indicates that the soluble fraction of the aerosol contributes only about 10% to the variation in Nd. Although there are some disclaimers in the remainder of section 3.1.3 about variation of composition etc, this result does say that VARIATION in composition is of much less significance. It would be very valuable if the authors told us why the size distribution and updraught are so much more significant than composition.

Page 7 - The statement “It must be remembered that droplet activation is dependent on the number distribution of particles of a given type and not the mass distribution” may be a little misleading. I think what is intended here is that droplet numbers are more proportional to particle numbers than particle mass. However, the activation of any

ACPD

5, S3664–S3669, 2005

particular particle is very dependent on the particle mass (soluble mass in particular). This distinction should be clear.

Page 8 - “The assumption that BC belongS to the insoluble fraction of the aerosol has been questioned by recent experimentsĚ” Table 1 indicates that the soluble composition is of relative less importance, and now it is being suggested that BC may be important. A rational “Ě the organic partitioning between water droplets and insoluble material is crucial.” Crucial to what?

Page 9 - re-write “Likewise, model mixtures of WSOC for many different types of aerosol in a range of locations are available their definition is in progress.”

Page 9 - First paragraph of section 3.1.4.2.1. Add Rupakheti et al. (AS&T, 2005) to this discussion of submicron particles and HC connections.

Page 10 - “The high organic content at sizes around the droplet activation threshold suggest that the effect of organics on warm cloud activation may be larger than their contribution to the overall mass.” This is an important statement that I only partly agree with, but an explanation is certainly needed. And I think that the explanation should be in the context of Table 1, e.g. the organics play an important role in shaping the size distribution.

Page 10 - discussion of nitrate. Very significant nighttime nitrate is observed in rural regions in eastern North America (Rupakheti et al., AS&T 2005).

Page 10 - “Unpublished results from a mountain station Ě” As in one of my opening comments, I do not believe that unpublished data should be part of a review. Included and accepted, it then becomes “published.” The data should go through the proper publication process. It is unfair otherwise.

Page 10 - “The mass spectral fingerprints Ě are very similar from site to site.” What is significant about them? A high degree of oxygenation does not refer to the whole organic spectrum.

ACPD

5, S3664–S3669, 2005

Page 11 - Single particle techniques have certain important advantages, as described, but they do not represent the total mass of the particle in any way close to the AMS. This distinction should be made.

Page 11 - “a little larger than the sizes where particle composition is most important for activation.” Refer to section 3.1.1.

Page 11 - “Indeed it is extremely difficult to prepare pure sulphuric acid particles in the lab without organic contamination.” Is it then fair to say the reverse, given that so many studies are looking at CCN activity of organics in lab settings?

Page 11 - “Consequently, the widespread oxidation of SO2 to H2SO4 in the atmosphere Ĕ ensures that any particle that has spent much time in the atmosphere has acquired at least a small percentage of sulphate.” Reference?

Figure 8 - Reference?

I am still in the process of transcribing my comments on the balance of the paper, and will submit them in another day.

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