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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud albedo increase from carbonaceous aerosol" *by* W. R. Leaitch et al.

Anonymous Referee #1

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The results of this study can be summarized as follows: 1) An aerosol-CDNC closure study is conducted using data from two flights through marine stratocumulus clouds in the NW Atlantic. An aerosol-CDNC closure study (see Conant et al., 2004) is where CDNC observations are used to validate a parcel model that predicts cloud droplet number concentration (CDNC) from observed aerosol size distribution, composition, and cloud updraft velocity. Closure was achieved within experimental uncertainty. 2) By varying the assumed chemical properties of organic matter (OM) in the parcel model, the study finds scenarios when reductions in OM solubility can either enhance or reduce the CDNC relative to the assumption that the OM has chemical properties equivalent to that of sulfate. These variations, however, are not large enough to be constrained by the closure study, except to say the OM is not both externally mixed and insoluble.

These findings are not entirely earthshaking, given that similar results have been found

in prior aerosol-CDNC closure studies of Conant et al., 2004; Meskhidze et al., 2005; Fountoukis et al., 2007, as well as the aerosol-CCN closures and sensitivity studies of VanReken et al., 2003; Dusek et al., 2007; and Rissman et al., 2004, to name a few. The results are valuable since the observations were conducted in the NW Atlantic adding a new climatological regime to those in which the prior closure studies were conducted. Furthermore, the sensitivity study explores a potentially important and counter-intuitive chemical effect in which lowering the solubility of OM in the accumulation mode can catalyze the activation of sulfate at smaller sizes.

A main goal and unique value of this paper is to discourage the practice in some GCM modeling studies of treating OM as being less efficient (per unit mass) than sulfate in nucleating cloud droplets. The results of this closure study together with those cited above place the focus more upon the size distribution of aerosol than its chemical makeup. To defend the idea that OM is as (or more) efficient in affecting cloud microphysics than sulfate, then, one has to focus how an emission perturbation affects aerosol size distribution and number concentration (and only to a lesser extent, composition). Making this argument convincing would require either 1) a globally representative data set, or 2) a detailed large-scale simulation. The latter would require a realistic treatment of the evolution of the aerosol size distribution with time, accounting for processes such as condensation, coagulation, cloud processing, etc. Such an effort would have been beyond the scope of the present study.

Since there are only two cases being presented, it is not clear that this result will compel modeling groups to change how OM is treated in GCM cloud parameterizations. However, it does present one fairly straightforward counter-example to the conventional wisdom that the lower solubility of OM makes it less efficient at producing CDNC than sulfate. It is noteworthy that Meskhidze et al., 2005 also observed that aerosol number concentrations were higher (per unit mass aerosol) in their high OM cases than in the low OM cases, lending supporting evidence to this argument.

The study does not mention two of the prior aerosol-CDNC studies cited above in which the role of OM was explicitly discussed (references below). Please also consider the following minor comments:

Minor Comments:

p2132 L16-17: "principally due to the increase in the carbonaceous components" overstates the generality of the comparison between the two cases being studied. The causality in this wording implies either a general physical relationship, or a controlled experiment. The two flights don't represent a general relationship between mass concentrations and CDNC, since one can envision a mass concentration from Flight 1 having a different size distribution that might yield the CDNC concentrations found on Flight 2. Nor is this a controlled experiment in which the properties of a sulfate aerosol population are tested before and after a carbonaceous perturbation is introduced. It would be safer to say "principally due to the increase in the CCN concentration" which would be more defensible.

p2135 L25: "Increase in size distribution is explained by" is vague and inconsistent with other uses of the term. (Does the term "size distribution" refer to shape or both shape and concentration together?) I suggest replacing this phrase with "Increase in number concentration is associated with" to be more consistent with the representativeness of the data being presented.

p2140 L23-25: The influence of aerosol on LWP is not supported by the data shown in this manuscript due to unconstrained differences in meteorological regimes between the two flights. This suggestive association should be removed. Mention of potential aerosol influences on LWP would be more appropriate in the introduction. The discussion in the introduction is presently very limited to the Twomey effect.

p2141 L6-7: The differences in the shape of the size distribution below 100 nm is crucial to activation behavior differences. Thus it is not accurate to imply number concentration is the only principal difference between the size distributions.

p2147 L2: Insert "mass" between "aerosol" and "is" to clarify that number (and CCN) concentration changes are not uniquely attributable to either OM or sulfate changes. There are surely other factors that cause these size/concentrations to be different.

Fig 2: Legend should read "Flight 1 - SMPS+FSSP-300" instead of "Flight 1 with FSSP-300".

Fig 2 caption: Replace "Fig 2a" with "Table 5" or "Fig S-5"

Figs 2-3: Are PCASP data used in this study? If not, they don't need to be discussed in the text. If so, they should be plotted in Figure 2 (and cited in Figure 3, if PCASP is used there).

References:

Conant WC, VanReken TM, Rissman TA, Varutbangkul V, Jonsson HH, Nenes A, Jimenez JL, Delia AE, Bahreini R, Roberts GC, Flagan RC, Seinfeld JH, Aerosol-cloud drop concentration closure in warm cumulus, J. Geophys. Res., 109, DOI: 10.1029/2003JD004324, 2004.

Meskhidze N, Nenes A, Conant WC, Seinfeld JH, Evaluation of a new cloud droplet activation parameterization with in situ data from CRYSTAL-FACE and CSTRIPE, J. Geophys. Res., 110, DOI: 10.1029/2004JD005703, 2005.

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