Atmos. Chem. Phys. Discuss., 10, C2539–C2544, 2010
www.atmos-chem-phys-discuss.net/10/C2539/2010/

Atmospheric Chemistry and Physics Discussions

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

W. R. Leaitch et al.

Richard.Leaitch@ec.gc.ca

Received and published: 11 May 2010

Anonymous Referee #1 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.

Response: The title of the paper is "Cloud albedo change from carbonaceous aerosol". The focus on "closure" misses the point that this work actually demonstrates a change in the light scattering properties between the two clouds attributable to the carbonaceous particles. The application of the model (or 'closure') is only a vehicle to delineate the processes leading to the differences in cloud albedo and enable attribution. We refer to the CCN papers as appropriate, but to say that CCN closure is relevant is again missing the point. As the reviewer is undoubtedly aware, CCN measurements do not account for growth rates and hence the feedback to cloud base supersaturation. CCN chambers are significantly different from atmospheric clouds.

Reviewer 1: 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.

Response: Thank you for the comment.

Reviewer 1: 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 rep-

resentative 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.

Response: This comment is a little confusing. We refer to two published global simulations in this paper that already suggested the importance of OM. The present results support those modelling studies. The notion that this work places focus on the size distribution rather than the chemical composition is overstated. What we show is that the CDNC result from a combination of effects. The term “efficient” can mask the understanding of the processes. OM may be more or less efficient relative to water uptake or it may be more or less efficient relative to shaping CCN number concentrations. We do not suggest that OM is more “efficient” than sulphate at nucleating cloud droplets.

Reviewer 1: 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.

Response: The other reviewer also brings up the point that this is only a comparison of two cases. Seldom, if at all, is a paper criticised for having too much data, yet papers using relatively large datasets can not contrast the observations with sufficient detail to provide much interpretation of the processes unless the number of significant processes impacting the subject is relatively few. In the case of the indirect effect, and even just considering the cloud albedo effect, the number of significant processes is considerable; this is a major reason that it is so difficult to constrain. Contrasting only a few cases, as done here, can be informative. Further, there is nothing in the observations or the analysis to suggest that these processes are unique to the measurement

location.

Reviewer 1: 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).

Response: We have added reference to these two other studies.

Please also consider the following minor comments: Reviewer 1: 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.

Response: There are two reasons that the original statement is justifiable: 1) The similarity of the number size distributions over the relevant size range; the conclusion is based on the observations rather than hypothetical; and 2) the sulphate mass in the second case is not preferentially distributed among the smaller particles (if different at all, it is more distributed in larger particles relative to flt 1). We carefully consider the necessary aspects using the model and the conclusion that the difference is “principally due to the increase in the carbonaceous components” is correct. In response to the reviewer’s suggested statement, we did not measure CCN.

Reviewer 1: 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.

Response: Thank you, we have revised the statement accordingly.

Reviewer 1: 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.

Response: We mention this for completeness only. We have modified the statement to be clear on this point. It is inappropriate to venture into this in the introduction. The introduction is intentionally very limited to the Twomey effect.

Reviewer 1: 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.

Response: In figure 2, we show that the size distributions are consistent (shape-wise) down to about 60 nm, which is well below any possible activation diameter for these types of clouds. This is also evident from the parcel model calculations.

Reviewer 1: 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.

Response: We have changed the sentence to read “The increase in the fine particle cloud base aerosol is associated with an increase in the mass concentration of the carbonaceous aerosol.” The differences in the size distributions are primarily in number (above 60 nm). Since the sulphate mass concentrations are close and the sulphate of flight 2 resides more in larger particles, the difference in number concentrations (at

least in the 100-400 nm range; figure 6a) is due mainly to the carbonaceous components. We can not describe the processes by which this happened. Such a discussion is important, but it is outside the scope of this study. We presume that one situation the reviewer is considering is that nucleation of sulphate spawned the increase in number concentrations and that the organic just condensed. Another reasonable possibility is that the aerosol picked up primary anthropogenic carbonaceous aerosol during its passage over the east coast of Canada and that some sulphate (either from anthropogenic or marine sources) condensed or mixed with it. In either case, it is the carbonaceous components that are increasing the numbers of particles; there was clearly insufficient sulphate to do this.

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

Response: Corrected, thank you.

Reviewer 1: Fig 2 caption: Replace “Fig 2a” with “Table 5” or “Fig S-5”

Response: Thank you. In response to this and comments of reviewer 2, we have integrated the supplemental figures with the main figures. Figure S-5 has been added to Figure 2.

Reviewer 1: 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).

Response: The PCASP (APNC140) are used for the profile data in Figure 4 as well as Figure 5 because of the greater time resolution. The PCASP data are not used for the size distributions because they are limited in particle size; however, the relevant number concentrations of the PCASP and SMPS compare well.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 2131, 2010.