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Atmospheric Chemistry and Physics Discussions

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

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Anonymous Referee #2 The work is an interesting contribution to up-to-date research on aerosol / cloud interactions, although the number of cases that are examined is limited to two. These two cases are airborne measurements performed on two different days during which different aerosol and cloud properties were observed. While on one day the aerosol was composed of 75% sulfate (by mass), it was a mixture consisting of approx. 50% sulfate and organic compounds on the second day. The total number concentration was higher on the second day, and also the cloud droplet number concentration (CDNC) and other parameters, e.g. the updraft velocity. The measurements were used to initialize model calculations which helped to give information on the sensitivity of the CDNC on updraft velocity and chemical composition. The main result presented is, that the carbonaceous components of the below-cloud aerosol contributed to the increase in the CDNC. When in the model the aerosol was treated as an

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external mixture and the carbonaceous component was treated as hydrophobic or only slightly soluble, then the model results were outside the range of the measurement observations. Therefore, this scenario could be excluded. All other scenarios (differing in mixing state and hygroscopicity of the carbonaceous components) yielded results that were within the observations, making it difficult to derive further information on e.g. the aerosol mixing state or the hygroscopicity of the carbonaceous components. From looking at the results presented in Table 6, I would even suggest that assuming the whole aerosol to be sulfuric acid would yield CDNC in reasonable agreement with the observations (the lower super-saturation (as shown in Figure S-7) is countered by the higher hygroscopicity of the particles). It follows as a whole, that the role of anthropogenic carbonaceous components was as large as was observed because these components were so much more abundant in one case than in the other, while the amount of sulfate was similar in both cases. I.e., it indeed can be said that the case study shown here shows an example for atmospheric aerosol, for which the effect of anthropogenic carbonaceous components is larger than that of anthropogenic sulfate.

Response: Thank you for the summary and the thorough review. A couple of comments concerning the above remarks: - As with Reviewer 1, you point out that these are only 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. As you suggest, the present case study is an example that previous global representations of the effect of the carbonaceous aerosol can be reasonable. There is nothing in the present observations or analysis to suggest that these processes are unique to the measurement location. - We agree that the result for the whole aerosol as sulphuric acid is in reasonable agreement with the observations, but of course the aerosol chemistry measurements do not suggest that to be a reasonable assumption. We have included it to

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demonstrate the range of possibilities, to provide some context for past aerosol-cloud studies that have made this assumption for the aerosol and to demonstrate the effect on control of supersaturation.

Reviewer 2: The whole work would improve, if it was a bit clearer structured. There were a number of typos and errors in the text and figures, hinting towards figures that didn't exist (e.g. Fig. 1c) or giving wrong units (e.g. heights in Fig. S-4), etc. A list of these errors is given in the specific comments. Also, reading the abstract, one could think that measured data are reported, only. However, the work includes modeling that helps to interpret the measurements. The result concerning the role of anthropogenic carbonaceous components could only be drawn from the modeling results. Therefore, some words about the modeling have to be added in the abstract. The same applies for the "Summary and Conclusion" section. This section needs to be more explicit about what was done and what was concluded from that. Also, it would be nice if it was described in the "Modelling"-section, which updraft speeds were used with which initial set-ups, instead of just adding more and more calculations that were done in the "Results and discussion"-section. Also, it seems the mixing state of the aerosol was changed together with the surface tension, so it is not obvious, which of these changes influenced the results in which way.

Response: We have carefully edited the manuscript for errors. We agree that the modeling must be (should have been) mentioned in the abstract and 'S&C', and it has been appropriately revised. A discussion of the application of the updraft speeds is now included in the "Modelling' section, and we have tried to clarify possible confusion of surface tension and mixing state considerations.

Reviewer 2: It is also not clear to me why this work needs a supplement. I would prefer to have all figures at hand in the main body of the text, but I would leave this up to the authors.

Response: We agree and the supplement has been incorporated into the main text.

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Specific comments:

Reviewer 2: abstract, line 1ff: The first sentence reads somewhat strange – please try to reword it.

Response: The first few sentences have been revised and also include a model reference.

Reviewer 2: abstract, line 13: "were similar on both days“ – “similar” is too strong, here, especially since you just said that the number concentration differed by a factor of 2. (Looking at Figure 2, they are also not really similar in shape, : : :)

Response: We have revised this statement, but we disagree about “similar”. We have defined a range now over which we believe the shapes of the distributions are ‘similar’.

Reviewer 2: p 2133, line 20: What do you mean by "some sulphate“? You might want to restructure the whole sentence.

Response: We have re-written it to read “In the absence of significant water activity of the carbonaceous aerosol, a relatively small fraction of sulphate in a particle can influence the ability of the mixed sulphate-carbonaceous particle to act as a CCN”.

Reviewer 2: p 2135, line 8ff: The global cloud albedo effect of -1.85 Wm-2 is smaller than the added up values you cite in the following (-0.3, -1.16, and -0.52 Wm-2) – please check this!

Response: The authors of that study note that the total forcing does not equal the sum of the individuals because of the nonlinear relationship between CDNC and aerosol number concentrations. We have added this statement.

Reviewer 2: p 2138, line 18ff: Did you account for the fact that the size distribution measured by SMPS and APS were those for dried aerosol, while those measured by FSSP300 were for ambient conditions? How? Also: how did you convert aerodynamic (APS) to geometric diameter?

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Response: We did not attempt to account for the potential contribution from water to the FSSP-300 particles. The measurements used for the size distributions were collected below cloud at RH values of 83% for Flt 1 and 85% for Flt 2. We mention this in the revised text. The aerodynamic diameter of the APS was converted to a geometric diameter assuming spherical particles and a density of 1.5 g cm-3. The latter was mentioned in a figure caption but overlooked in the text. It has been added to the revised text also.

Reviewer 2: p 2141, line 10-11: The text here says “total mass distributions”, while the caption of Fig. 6b says “volume distribution”. Which density did you use to convert between mass and volume? (In Table 4 you give 1.7 g cm-3 and 1.5 g cm-3 for Flight 1 and Flight 2, respectively. Are these the values you used here, too? Explain how you obtained them and why they differ.)

Response: Thank you. We have revised the text accordingly and the figure was correct; it compares volume distributions, not mass. The volumes are calculated from the AMS using densities of 1.78 g cm-3 for sulphate and 1.2 g cm-3 for the total organic components. The values in table 4 are estimates for approximate reference based on mean densities arising from the above.

Reviewer 2: p 2141, line 12-13: Specify what you mean by “detectable sodium and chloride”.

Response: This has been revised and the supplemental figure has been moved to Figure 6c in the revised manuscript.

Reviewer 2: p 2142, line 18ff: Please say straight away, that you made calculations for (almost) all four updraft speeds for both flights (Table 6 only misses values for an updraft speed of 20 cm/s for Flight 2). You need this for your comparisons, and the reader is just confused if you reveal what you did only bit by bit.

Response: First, we have added the gust velocity figure into the main text as Figure

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7 (the previous Fig 7 is now Fig 8), and we have revised this text accordingly. This remark is a little confusing. In this paragraph we do say what updraft speeds we use. Later we do apply the updraft of flight 2 to the aerosol of flight 1; but there is a specific purpose to that as we discuss beginning on line 2144, line 26. To bring that discussion in at this point might be more confusing, but we have added a sentence at the end of the paragraph. At this point, we are only defining the updraft speeds we use.

Reviewer 2: p 2143, line 13-15: ": : : updraft speeds used by Fountoukis et al. (2007) are relatively lower than based on our approach and are mostly for cumulus." This sentence seems to be incomplete (and misleading: didn't you get your values from measurements? Or what do you mean by approach?) Would one not expect higher updraft speeds in cumulus clouds?

Response: We agree that this is unclear, and we eliminated much of that text.

Reviewer 2: p 2143, line 18-20: ": : : and the updraft speed" appears twice, here – specify, which updraft speed you mean, in both cases

Response: We have removed this.

Reviewer 2: p 2144, line 15ff: Please add for the third scenario, that also assuming all of the particles to be made up of sulfuric acid would have produced decent results, too. As you show this in Table 6, it should also be mentioned in the text. This does, to my understanding, show, that the increase in particles is one of the main factors governing the increase in CDNC, no matter what these particles really are (and as long as they are not completely hydrophobic). Please comment on this, in the text here or further below (see comment on p 2146, line 5ff).

Response: We don't disagree with the premise, but the observations clearly show that the particles of flight 2 were not completely or even mostly sulphuric acid, so to suggest that this is a viable scenario is incorrect.

Reviewer 2: p 2146, line 5ff: Here you comment on the model scenario for which

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all particles were composed of sulfate. As said above, this would also yield results comparable to the observation. In this case, the lower super-saturation is balanced by the larger hygroscopicity. Please comment on that fact in the text here or above (p 2144, line 15ff).

Response: We have changed the last sentence of the paragraph as follows: "However, if we assume that the particles are composed completely of sulphate a slight reduction in the CDNC results because the large increase in droplet growth rates reduces the cloud base supersaturation (Figure 9); despite the slightly lower value, the CDNC resulting from the all sulphate scenario falls within the range of the observed CDNC." We are not in favour of trying to promote number or water uptake characteristics as more or less important in the discussion of OM vs sulphate. It is a point of this paper that we must consider both sides of that coin.

Reviewer 2: p 2148, line 21 and p 2153, line 32: I am not sure what the rules about citing a webpage are, but personally I would give this in a footnote, not within the citations.

Response: We are happy to leave it or place it where the ACP editors prefer.

Reviewer 2: Caption of Table 3: "profiles of Fig. 1c" – there is no Fig. 1c! Please, also explain in the caption why some values are given in parenthesis.

Response: It has been corrected to Figure 5 and the values in parentheses are defined (the PILS values).

Reviewer 2: Table 6: Maybe I missed this, but you might want to add a sentence to the text explaining that the derived CDNC for Flight 1 does not depend on the assumption of the solubility of the organic fraction, because the sulfate is determining the aerosol particles activation to droplets by far.

Response: The following is the second sentence of revised Section 5 (Results and Discussion): "There is little or no change in the CDNC with change in the hygroscopicity

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of the OM due to the relatively large sulphate fraction that controls water uptake."

Reviewer 2: Table 6 again: Shouldn't there be one more yellow box, for Flight 2, updraft of 50 cm/s, at the organic solubility of 200g/l? This case is similar to the other two that are marked in yellow.

Response: Yes.

Reviewer 2: Caption of Figure 2: ": : : model calculations (Fig. 2a) : : :" – what do you mean by Fig. 2a. There is no such Figure.

Response: Corrected.

Reviewer 2: Figure 3: It is hard to distinguish the circles and the squares in A and B – please find a different way to display the model results!

Response: The model values are displayed differently.

Reviewer 2: Caption of Figure 3: Delete the ">" in the index of "APNC".

Response: Done.

Reviewer 2: Figure 5: Please change the cloud marker for Flight 1. If you used a shaded area here, both cloud altitudes could be seen at the same time. Also, the values at cloud base differ from those given in Table 3. Why?

Response: If you look closely at Figure 3, you will notice that the clouds do not significantly overlap in altitude; we believe that the shaded areas in Fig. 5 are appropriate. The values differ as discussed in the caption of Table 3; the Table 3 values are for level flight.

Reviewer 2: Caption of Figure 6 A: It says, that "sulphate to sulphate plus organics" is shown, while "Organics / (Sulfate + Organics)" is displayed at the axis. Correct this!

Response: Thank you. It is corrected.

Reviewer 2: All figures of the supplement: Purple and black are hard to distinguish!

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You need to use a different color code for Flight 2 in all cases! Figure S-3: In both panels, the data for Altitude and APNC are often overlain with the FSSP-data. Either put the former two datasets on top (they should not bury the FSSP-data, as they scatter much less), or make separate panles. Figure S-4: The “Altitude (m-MSL)” is given in the wrong numbers (*1000 seems to be missing). Figure S-5: The diameter should be given in micrometer, not nanometer. Interactive comment on Atmos. Chem. Phys. Discuss., 10, 2131, 2010.

Response: As recommended by the reviewer, the supplemental information has been incorporated into the main text and the above issues are dealt with.

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