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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimating the impact of natural and anthropogenic emissions on cloud chemistry: the influence of organic compounds" *by* L. Alfonso and G. B. Raga

Anonymous Referee #2

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This paper presents model simulations of the effects on cloud microphysics and precipitation of A) two different parameterisations of the fraction of the initial aerosol that is not made up of the major inorganic ions and B) 8 different cases concerning gas phase concentration and liquid phase reactions. The initial aerosol has a low number concentration (100/cm3) and a small fraction of inorganic material (10% by mass is ammonium sulphate).

The paper deals with an important issue and I find that it can be published after some changes. The most important are the first two discussed below: the aims if the paper and how the conclusions are presented.

The first thing I would like to discuss is how the authors state the goals of the paper.

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They can be found on page 594 lines5-13 and are far to general. These questions are addressed in the paper, but only for a specific set of conditions. The statement of the authors concerning these conditions and arguments why they are interesting to study in the introduction would help the readers a lot and avoid misunderstandings. For example are emission of gases and particles from Petroleum industries in maritime zones mentioned in the abstract, but they are not discussed in the paper. The title indicates that the results are discussed in terms of natural and anthropogenic emissions and in the paper there are very few references to natural/anthropogenic.

Concerning the conclusions, I do not have any problem with the validity of the conclusions, for the conditions treated in the paper. However, the conclusions, if read alone, indicate that they are general. It has to be clarified for which conditions the conclusions are valid!

In my opinion, changes according to the two comments above have to be made before publication.

I also have some comments concerning the initial aerosol in the two cases:

The authors argue that low particle number is valid for a marine aerosol, but how does that fit with the chemical composition. It is not according to the general picture of marine submicron aerosol particles that they only contain 10% soluble inorganic ions. If you have more recent information or site-specific information, please make references.

In the IOC case, the organic fraction is modelled as oxalic acid. The authors argue that oxalic acid is one of the most abundant organic compounds in tha atmospheric aerosol particles. However, it only makes up a small fraction of the WSOC and is not typical when it comes to properties that are relevant for cloud droplet nucleation, for example molar mass. Most other WSOCs contribute a smaller number of ions/molecules per particle volume (or mass) than oxalic acid does.

I also have some problems with the parameterisation of oxalic acid in the Köhler equa-

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tion (eq. 11 page 602). As in many other recent papers the dicarboxylic acid (DA) is treated as if it is fully dissociated at activation. Based on dissociation constants, some of the larger dicarboxylic acids should be treated as if they were not at all dissociated in the Köhler theory at activation of pure DA particles (but, they still influence the pH). Oxalic acid is somewhat more acidic, but is it really fully dissociated at activation? Have you made any calculations based on the dissociation constants (table2c)?

Another problem with oxalic acid as a model compound might be its vapor pressure. There might be losses of oxalic acid to the gas phase. However, I find that this is beyond the scope of this paper and there is also still a lack of data.

I also have a question concerning the influence of soluble gases on the activation. Are they taken account of in the activation simulation? Or have you made any sensitivity calculations that indicates the relation between the number of molecules coming from the initial aerosol compared with the number of dissolved gas molecules?

Reading the paper carefully, it is clear which results refer to the high and low sulfuric acid concentrations respectively. However it would help the reader a lot, if this was also stated for example in the figure captions, in the conclusions and preferably also in the text when discussions that depend on the sulfuric acid concentration are made.

More detailed comments:

p.593 l. 7-8 If "aqueous phase" refers to cloud droplets, it might be better to just say that, since also aerosol particles are often (most of the time!) aqueous solutions also at relative humidities far below 100%.

p.593 l. 20-23 The paper by Facchini (2001) is a review of the present status of knowledge concerning organic compounds and aerosol particle/cloud drop interaction. "The remaining part of the organic carbon was considered insoluble in cloud water and the effects of organic compounds were mainly linked to the formation of organic films on the surface of droplets." is almost a direct citation from the Facchini paper, but in her

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paper it refers to the situation during the past 20 years, and not to their own work. Please check!

p.596 l. 1 The reference (Ogura and Takahashi, 1971) is missing in the reference list.

p.601 l. 4-5 It is somewhat misleading to say that in the CIC case the CCN are composed only by inorganic compounds, in particular ammonium sulfate, since in this case the ammonium sulfate contributes only 10% by mass.

p.601 l. 28 It would help the reader if you state that the percentages are by mass also in the text and not only in the table.

p.605 l.18-p.606 l. 1-2 It says "When the concentration was increased, a simulated cloud with an unrealistic long lifetime was obtained." Could this be an indication of a problem in modelling the precipitation? Since precipitation is a major issue in this paper, it would be interesting to know if the ability of the model to reproduce precipitation has been tested.

p.606 l.25 "Ridley et al. (1990)" should be "Ridley et al. (1992)" according to the reference list.

p.607 l.19-21 It says "In our simulations, we only tested the influence of the concentration in the precipitation development. We did not change any other parameters of the CCN distribution like the dispersion and the mean radius." It is not clear to me, what the meaning is. Does concentration refer to particle number concentration or (most probably) chemical composition?

p. 615 l.14 The paper by Granby and Christensen has one more author: Christian Lohse

In the text I could not find citations to the following 3 papers that are in the reference list, please check:

Pruppacher, H. R. and Klett, J. D.: Microphysics of clouds and precipitation, Kluwer

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Academic Publishers, 1997.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics, Jonh Wiley & Sons,1326, 1998.

Sempere, R. and Kawamura, K.: Comparative distributions of dicarboxylic acids and related polar compounds in snow, rain and aerosols from urban atmosphere, Atmos. Environ., 28,449Ű459, 1994. 10

Table 5: In the IOC case the maximum concentration of droplets is higher then the initial aerosol particle concentration. To help the reader understanding this, a short discussion would be god.

Figure 6 and 7: What is the reason for the decrease in the gas phase concentrations at a height of 4000m?

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 591, 2004.