**Atmospheric
Chemistry
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Discussions**

Interactive
Comment

Interactive comment on "A global modeling study on carbonaceous aerosol microphysical characteristics and radiative forcing" *by* S. E. Bauer et al.

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We would like to thank both our reviewers for their productive comments. We addressed most questions in the revised version of the paper and will give some additional clarifications here in the answer to the reviewer's comments:

Rev #1

Comment Nr 1: Modes versus Moments and other model questions

The model description is improved, better describing the external forcings and model setup. However the reviewer raised two mayor questions:

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How do we obtain aerosol size distribution information, as needed to calculate micro-physical and optical calculations, from a 1-point quadrature scheme and only carrying the information of two moments, aerosol mass and number, per population?

The MATRIX model is still under development. The version used here and described in Bauer et al 2008, was developed with only two moments in order to have a framework to develop the microphysical parameterizations. This development is now concluded and as a next step we are working towards using higher moments to describe the evolve-ment and the mixing state of aerosol populations. Eventually we expect to carry more quadrature points and moment information but for less populations, or mixing state def-initions as used now. However at this stage of our model development we have to make assumption about the shape of the aerosol size distributions. We assume a lognormal shape with constant width. Those specifications are given in the model paper Bauer et al 2009 and the current paper is improved to better explain those assumptions.

Condensation versus coagulation?

Table 3 in Bauer et al 2008, presented the budget per microphysical process for each aerosol population and we see that taking into account all aerosol populations, coarse and fine, coagulation is the dominating mixing process. However I should be careful with general statements like you criticize, as I can only make such a statement in light of the microphysical processes I take into account. In MATRIX right now, only con-densation of H2SO4 onto aerosol surfaces is counted as condensation, in reality many more gases can condense which possibly could lead to domination of condensation over coagulation as dominating mixing process. I have corrected that section in the paper.

Comment Nr 2: Size versus number concentration change

The second comment basically included two questions. 1) What is more important a change in the size of a certain emission, however this would consequently also lead to a change in number concentration, or the change simply in number concentration with

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constant particle sizes?

I agree this is an interesting question, but in this paper we only investigated the first part of this question and therefore I can't comment on the effect of simply changing the emitted number concentration. Furthermore I believe every aerosol microphysical model might react very differently to these kind of sensitivity experiment, as here the model design especially the representation of size distributions, whether it is a modal, sectional or moments scheme might play an important role. It would be very interesting to repeat these simple experiments with different model types. I improved the manuscript to better explain the experimental setup.

2) The second part of question deals with the question why CDNC concentrations are changing with different OC/BC initial emission sizes.

Please be aware that by changing the size of the emitted BC/OC particle, by constant mass the number concentration is changed. This consequently will lead to changes in coagulation between OC, BC and other particles, as well as impacting the condensation of H_2SO_4 on the particles surface and the aerosols water uptake capability. The reviewer argued that the coagulation between a hygroscopic e.g. sulfate aerosol with an non-hygroscopic particle would not change the CDNC number concentration, as here only the hygroscopic particles would be counted. Well unfortunately there is never an easy answer in the world of aerosol microphysics. We have several effects here that are overlapping with each other. First of all we change BC and OC emission sizes. BC itself without any coatings can't serve as a CDNC particle, however when coated with sulfate or organic material it can become hygroscopic and OC is hygroscopic. Secondly, in our model the hygroscopic populations, ACC and OCC, change in number concentration due to several effects:

1) ACC concentrations depend on the availability of its precursor species, e.g. SO2, NH3, H_2SO_4 and further oxidants. H_2SO_4 has the possibility to form a hygroscopic ACC particle, or condense on the surface of a pre-existing particle, e.g. ACC, OCC,

one of the several BC classes etc. Therefore the number concentration of hygroscopic particles is already influenced by the competition for precursor species like H2SO4 between pre-existing particles and new particles, before eventually coagulation between hygroscopic and non-hygroscopic particles will take place and change the number concentration.

2) Secondly in the discussed experiments we changed as well the emission sizes of OC particles, therefore directly influencing the number concentrations of the directly emitted OCC and BOC particle number concentrations. Table 1 gives CCN concentrations [cm-3] per aerosol population for the discussed experiments.

BASE S1 S3

ACC 35 6 66

OCC 77 19 42

BOC 34 120 5

BCS 2 1 1

SS 14 6 17

MXX 1 6 2

Total 161 157 134

We improved the text in section 3 to better explain the involved processes and included the reviewer's concern as discussion point.

Comment Nr 3: Model mixing state setup.

Homogeneous internally mixed or core – shell treatment only relates to the treatment of the aerosols in the radiation scheme. The two different treatments are described in section 2.2 Aerosol radiation coupling. Therefore this does not affect the mixing state simulation in the model, which is strictly determined by the microphysical processes

of nucleation, condensation and coagulation. The mixing state options only relate to the choices possible in the aerosol – radiation coupling. Indirectly of course the mixing state is affected by the differences in climate feedbacks but those impacts are rather small. I corrected the labeling of Table 1 to address this concern.

Comment Nr 4: CDNC change in experiment BCOCBF

Experiment BCOCBF, with reduced biofuel sources, is the only experiment where we saw in the global mean decreasing CDNC concentrations compared to the BASE experiment. I kept that discussion in the original paper brief, as this is the expected result; decrease primary emissions leading to decreased CDNC concentrations. Reducing biofuel sources (BCOCBF) is the only scenario where we see decreased CDNC concentrations. The strong reduction in BC (1.5 Tg/a) and especially OC (6.4 Tg/a) particles finally resulted in a reduced number concentration of activated particles, which was not the case in the prior discussed scenarios, were the total primary emission reduction (OC and BC) was only ranging between 1.7 and 3 Tg/a. In this experiment the decrease in primary emission concentration now dominates over the mixing state effects that lead to localized CDNC increases in regions with moderate BC reductions. Eventually this leads in the BCOCBF experiment to a positive AIE +0.2 W/m2 and a net radiative change of +0.13 W/m2. However as it is contrary to the results of the three other sensitivity experiments I have extended the discussion of this effect now in the paper.

Minor corrections

We changed the manuscript according to all minor comments the reviewer pointed out, except the comments addressed here:

REV #1: p. 4545, line 15: "Most absorbing, . . .", please rewrite this sentence. Line 27: "1.4- and 13 fold", seem to me a typo here?

Khalizov et al 2009 indeed found a 1.4- to 13- fold increase in absorption and scatter-

ing for soot particles coated with H2SO4 under high humidity conditions. This finding is mentioned several times in their paper therefore we believe this is no typo. Quotation form the conclusion of the cited paper: 'For soot particles with an initial mobility diameter of 320 nm and a 40% H2SO4 coating mass fraction, absorption and scattering are enhanced 1.4- and 13-fold at 80% RH, resulting in a single scattering albedo increase from 0.1 to 0.5.'

Rev #1: p. 4559, paragraphs before section 5: it seems to be the case that in the MATRIX model, BC and OC are always mixed with sulfate, nitrate and perhaps others. Is this the reason that BC is always in a small fraction in any of those BC-containing modes?

MATRIX tracks 6 different populations, defined by mixing state that can include BC. Five of those populations, BC1, BC2, BC3, BOC and BCS must contain BC in order to be populated. However it is possible that BC is the only chemical specie in a population, in that case it would be classified as BC1, which can have a coating of sulfate, nitrate ammonium and water but it doesn't have to. The reason that we get fairly small BC fractions in a particle is caused by the fact that initially the particle contained some BC, but than the other species grow more quickly and no more BC is added to the aerosol. However this is a problem I have to further investigate, as eventually I have to reclassify a particle when not sufficient material of one chemical specie is longer present.

Rev #1: p. 4562, second paragraph: was AAOT of OC and mixtures par t of the result?

The absorption optical thickness is calculated for all aerosols in the atmosphere, therefore OC in its various mixing states will contribute to AAOT.

Rev #2

The reviewer basically asked for a more detailed model description. The model used here is described in a previous paper, and here just shortly summarized. I extended the model description to make it easier for readers to understand the model without know-

ing the previous paper. Furthermore we are preparing a separate paper on the indirect effect, however as the paper is still under preparation we included a new paragraph, section 2.2 to describe the aerosol cloud coupling. We corrected the other minor comments as suggested and carefully checked and corrected the citations and reference list.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 4543, 2010.