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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol activation and cloud processing in the global aerosol-climate model ECHAM5-HAM" *by* G. J. Roelofs et al.

Anonymous Referee #1

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General comments:

The paper discusses a new parameterization of liquid aerosol activation to cloud droplets and chemical processing of activated aerosols within clouds. The parameterization is applied within a global climate model to study aerosol interactions with warm clouds at the global scale.

The paper covers a very important subject since the aerosol-cloud interactions are highly relevant with regard to the effects of aerosols on global climate. Since the parameterization considers the cloud processing of aerosols separately for different aerosol size modes, it is highly innovative compared to previous approaches which

only consider chemical changes of the total aerosol (sulfate) mass.

The methods applied appear to be sound. Most of the results are presented clearly. However, the paper needs to discuss several aspects in more detail. I would suggest publication of the manuscript after the following comments are considered by the authors:

Specific comments:

Major comments:

1) Abstract, section 3.1, and conclusions: The CDNC due to activation should be different from the CDNC in aged clouds due to self conversion of cloud droplets. Hence the CDNC simulated by the parameterization should not necessarily match the observations. Nevertheless, for the above reasons it could be expected that the observed CDNC should be lower than the CDNC of activated droplets. Since this is not the case in the present study, the model/parameterization might have problems in some cases. The authors should discuss the comparison in this manner, rather than aiming at resemblance of activation CDNC and CDNC observations.

2) Many assumptions of the study are just mentioned but not discussed with respect to their consequences. The authors should discuss how these assumptions potentially affect their results and they should provide reasons why these assumptions are necessary. The authors should discuss especially the following assumptions in more detail:

i. Sulfate is assumed to be generally present as ammonium bisulfate. (Page 523, lines 28-29)

ii. The CDNC supplied by the parameterization does not feed back to the cloud evolution and precipitation formation in the model. (Page 524, lines 13-14)

iii. In-cloud scavenging of interstitial particles is not considered (Page 524, lines 18-19)

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iv. The effect of HNO3 on activation is not considered (page 525, line 13)

v. Offline oxidant fields are considered (Page 525, 2nd paragraph). In this case the authors should mention that a sensitivity study (B2) discussed later in the paper may give some insight in the sensitivity to the oxidant fields.

vi. It is assumed that the simulated LWC is relatively accurate (Page 526, line 11)

3) The performance of the in-cloud chemistry parameterization was evaluated for a specific splitting of LWC to the diluted/concentrated (75%/25%) modes. It should be discussed how the model performs in case of other splittings. Or is the considered splitting assumed as constant? This should be explained in more detail.

4) The authors attributed the composition of the cloud droplets, and therefore the sulfate production efficiency of the respective droplets to the aerosol mode activated to form the droplets. Since cloud droplets may agglomerate due to autoconversion, the composition and, therefore, the sulfate production efficiency may change, which seems to be neglected in the present study. The authors should discuss this point and provide an estimate of the resulting uncertainty in sulfate production and aerosol processing.

Minor comments:

1) Please avoid the term undiluted. The 'undiluted' droplets are also diluted since they contain a large amount of water. They are just less diluted than the 'diluted' droplets.

2) Page 520, line 12: Specify the formulation, e.g. by writing 'CDNC is dominated by cloud droplets formed by activation of accumulation mode aerosols ...' (The formulation 'CDNC is dominated by particles from the accumulation mode' can be misleading since aerosols are not cloud droplets.)

3) Page 522, line 25: It should be described which kind of size distribution is assumed for the modes (monodispers, logmnormal,...?).

4) Page 523, line 3: It should be explained to the reader why only these modes are

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relevant.

5) Section 3.2: Specify which emission data you use for SO2 and whether these data are consistent with Roelofs et al. (1999) and Stier et al. (2005).

6) Page 531, line 23: The compensating mechanisms have to be specified.

7) Page 536, line 2: n(i,j) and r(i,j) should be explained earlier (e.g. on page 535 around line 10) since they are already used in the equation for B on page 535.

8) Page 536, lines 4-10: The authors should discuss how critical these assumptions are. How do these assumptions affect the results?

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 519, 2006.