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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impacts of aerosol indirect effect on past and future changes in tropospheric composition" *by* N. Unger et al.

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Response to reviewer #1 (F. Raes)

We thank the reviewer for helpful comments that have improved the manuscript.

General comments

1. Agreed.

2. Although we use an empirical formulation to determine CDNC rather than a mechanistic approach as in Abdul-Razaak et al. (2000) or Lohmann et al. (2007), assumptions required in mechanistic approaches regarding cloud-scale vertical velocity and aerosol size (when only aerosol mass is predicted) suggest that additional uncer-

tainties may arise since cloud-scale updrafts are not resolved in GCMs and aerosol sizes are not well constrained. Menon et al. (2003, JAS) compared SCM simulations of CDNC predictions with models that include both empirical and physically based schemes (including the Abdul-Razak and Lohmann et al. scheme) with observations and found no indication that mechanistic approaches were superior to empirically-based approaches. Additionally, the schemes used in this work for CDNC predictions have been compared with satellite-based retrievals in prior work (Menon et al. 2008, JGR) and were found to be within retrieval uncertainties.

3. We did originally prepare versions of the figures in percentage change. However, very large percentage changes may occur where the examined parameters/processes are unimportant or have very low absolute values giving a distorted view. There are further complications in representing competing changes in sign using percentage change. It would probably be excessive to include both absolute and percentage changes as we already have a large number of figures. Therefore we reached a compromise. We provide the figures in absolute values, which allows other modelers to compare easily with their values and gives a true representation of where the impacts are important and we include the percentage values in the text to provide a sense of the sensitivity of the aerosol-cloud impacts relative to the emissions changes.

4. We include more explicit discussion of the processes that drive the ACI impacts on the composition variables that we examine. In Section 3.2 (Impact of ACI on J(O1D) photolysis rate) we relate the ACI impacts to changes in cloud optical depth, cloud cover and increased scattering above clouds. Please see Response to Reviewer #2, especially point (7). We now include explicit description of ACI-induced changes to meteorological parameters, most importantly precipitation, that facilitates understanding of the changes to wet deposition.

Specific Comments:

1. We agree with the reviewer and have amended the title and text throughout the

entire manuscript to use 'ACI' instead of 'AIE'.

2. We would like to keep some description of the forthcoming simulations/methodology in the Introduction section. The main reason is that the experimental setup is conceptually quite complex involving differences between various pairs of simulations. Including some methodological explanation in the Introduction section provides useful/helpful guidance to the reader in terms of what to expect.

3. We now include in the Introduction section a detailed description of the various parameters that we chose to explore (including J(O1D), CH4 lifetime and sulfate wet deposition etc.) so they no longer 'come out of the blue':

'We focus on several composition parameters that are likely to be sensitive to changes in clouds. The photolysis of O3 yields excited state oxygen atoms, O(1D), that react with water vapor (H2O) to form the hydroxyl radical (OH), the major tropospheric oxidizing agent. This reaction pathway is the most important direct source of OH and therefore key in the photochemical processing that may lead to O3 formation or destruction and controls the lifetime of methane. The rate of this photolysis process (J(O1D)) is dependent on available incoming ultra violet radiation and therefore susceptible to cloud changes. In-cloud formation of sulfate aerosol may represent 50% of the total production budget globally, although the importance may be higher in polluted regions (Koch et al., 2006). Wet deposition is a key loss process for many trace species, especially nitric acid (HNO3) that represents radical termination in O3 formation chemistry, and aerosols and therefore strongly affects the ground level distribution of these pollutants.'

4. We have changed '. . ..supplements the decrease' to 'ACI impose further decreases in J(O1D)'.

5. In the conclusion section, to the paragraph beginning 'The major limitations of the study are the large uncertainties associated with the ACI, partly driven by uncertainties in aerosol emission inventories (especially for carbonaceous aerosols),. . ..', we have added: 'partly due to the parameterizations used to link aerosols with cloud droplet

number, ...'.

6. Done. Acronyms are re-defined in captions for tables and figures.

7. We suspect there may be a misunderstanding on the reviewer's behalf here. By definition, there is no value for 'aerosol indirect effect' when ACI are not included in the model. Of course, clouds will change and therefore cloud radiative forcing due to climate changes driven by changes in greenhouse gases. However, these changes are not related to aerosols.

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