Atmos. Chem. Phys. Discuss., 12, C12684–C12686, 2013
www.atmos-chem-phys-discuss.net/12/C12684/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Weak sensitivity of cloud condensation nuclei and the aerosol indirect effect to Criegee + SO_2 chemistry" *by* J. R. Pierce et al.

Anonymous Referee #1

Received and published: 14 February 2013

Recent research has indicated that stabilized Criegee Intermediates (sCI) could play an important role in atmospheric oxidation of e.g. SO_2. In this manuscript by Pierce et al., the authors implement sCI+SO_2 chemistry into the global aerosol model GEOS-Chem. The main focus of the work is on the effect of the additional SO_2 oxidation pathway on cloud condensation nuclei (CCN) concentrations. The manuscript is well written, concise and with clear conclusions. The manuscript should be published after addressing the following comments.

1) The main conclusion of the manuscript is clear, and maybe expected: additional sulfuric acid has little effect on CCN concentrations in regions where most of the particle growth is due to organics. This result should of course be somewhat sensitive to model parameters such as nucleation rate and the assumed size of emitted primary particles.

With e.g. lower activation nucleation coefficient you should have less nucleation, less competition for growing vapours, and maybe the CCN-sensitivity to sCI+SO_2 might be higher? If possible, the authors should include a sensitivity simulation with modified nucleation parameters (or primary emission diameter), or at least add some discussion on the issue.

2) The description of the sensitivity tests with additional SOA should be more clear: is the SOA mass artificially increased (with emissions correlated with CO), or are some precursor emissions increased? It seems that SOA formation is increased in a way that XSOA experiments do not provide more sCI, but only more particle growth?

3) Is the GEOS-Chem originally missing H_2SO_4 if compared against observations? As the implemented chemistry seems to induce large changes in H_2SO_4 concentrations, it might help the reader to know if this initial effect is towards a better direction for model performance.

4) The same for CN: does the inclusion of sCI+SO_2 chemistry improve for example the intra-annual variation profiles of total particle number? The additional chemistry would likely increase the magnitude of the seasonal cycle, which could already be overestimated by the model? The two chosen locations (Hyytiälä and AMAZE) might not reveal the whole picture of the effect. Unfortunately, the effect seems to be largest in areas with very little observations. Maybe at least an additional station could be included from Eastern US, where the effect can reach 15% for CN?

5) While 1-year simulations are likely enough for the annual CN/CCN calculation, the AIE is somewhat sensitive to the modeled cloud fields. The approach here uses climatological cloud fields and prescribed updraft velocities, and considers only the cloud albedo effect, which could underestimate the total aerosol indirect effect related to sCI+SO_2. With the presented changes in CCN concentration, the indirect aerosol effect is likely small, but Section 3.5 should include an uncertainty range for AIE, and discussion on the limitations of the approach.

ACPD
12, C12684–C12686, 2013

5) The aerosol indirect effect (Section 3.5) varies significantly from model to another. It would be useful for the reader to know some baseline for aerosol indirect effect in the GEOS-Chem, such as anthropogenic aerosol indirect forcing since pre-industrial, even if this can be found from the included references.

6) Could some future aspect be included in the conclusions: if assuming a significant decrease in global anthropogenic SO_2 emissions (50-90%) and a simultaneous increase in the biogenic VOC emissions (induced by climate warming), what would happen to the CN or CCN sensitivity to sCI+SO_2?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 33127, 2012.

ACPD

12, C12684–C12686, 2013

Interactive Comment