Atmos. Chem. Phys. Discuss., 12, C11257–C11259, 2013
www.atmos-chem-phys-discuss.net/12/C11257/2013/

Atmospheric Chemistry and Physics Discussions

ACPD

12, C11257–C11259, 2013

Interactive Comment

Interactive comment on "Oxidation of SO_2 by stabilized Criegee Intermediate (sCI) radicals as a crucial source for atmospheric sulphuric acid concentrations" *by* M. Boy et al.

Anonymous Referee #2

Received and published: 4 January 2013

The paper uses ambient observations and modelling analysis in an attempt to quantify the contribution of stabilised Creigee Intermediates (sCI) as an oxidant of sulfur dioxide and a source of sulfuric acid. The paper concludes that sulfuric acid concentrations are underpredicted by the model unless the reaction between sCI and SO2 is accounted for. The paper presents a useful analysis on an important topic that will be of interest to the community. I recommend publication after the following comments have been accounted for.

Major comments

1) It would be useful to include an analysis to demonstrate that the simulated H2SO4

with the standard model is statistically outside the observations given the uncertainty in the observations.

2) The rate of reaction between sCI and H20 needs to be discussed and accounted for. A wide variety of rate constants are available in the literature and the sensitivity of the results to this uncertainty should be explored. See also the open comments on this issue by G. Sarwar.

Minor comments

P27695, L22. Include citations to earlier papers that also demonstrated this (e.g., Spracklen et al., 2008; Makkonen et al., 2009, Pierce and Adams, 2009, Wang and Penner, 2009).

P27700, L13. No need to spell out acronym for MALTE again here.

P27700, L1. Here it is not clear whether you use the models to simulate both Hyytiala and Hohenpeissenberg or just Hyytiala. Please clarify.

P27700, L24. What about data from Hohenpeissenberg?

P22701, L7. Same comment as above.

P27701, L20. Would be useful to say how thick the lower model layers are and how many model layers are typically within the forest canopy.

P27702, L2. What do you assume for forest environment at Hohenpeissenberg?

P27703, L26. Only total monoterpenes have been measured at Hyytiala. How do you know that beta-pinene concentrations are not high here?

P27707, L16. Behaviour might be a better word than trend here.

P27707, L18-19. Replace the word behaviour with variability.

P27708, L22. Clarify what these percentages refer to.

ACPD

12, C11257–C11259, 2013

P27708, L5. Please include the equation for the calculation of CV(RMSE).

P27714, L24. Inclusion of these mechanisms in a global model has now been implemented by Pierce et al. (2012). References

Makkonen, R., Asmi, a., Korhonen, H., Kokkola, H., Järvenoja, S., Räisänen, P., Lehtinen, K. E. J., Laaksonen, a., Kerminen, V.-M., Järvinen, H., Lohmann, U., et al.: Sensitivity of aerosol concentrations and cloud properties to nucleation and secondary organic distribution in ECHAM5-HAM global circulation model, Atmospheric Chemistry and Physics, 9(5), 1747–1766, 2009.

Pierce, J. R. and Adams, P. J.: Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates, Atmospheric Chemistry and Physics, 9(4), 1339–1356, doi:10.5194/acp-9-1339-2009, 2009.

Pierce, J. R., Evans, M. J., Scott, C. E., D'Andrea, S. D., Farmer, D. K., Swietlicki, E., and Spracklen, D. V.: Weak sensitivity of cloud condensation nuclei and the aerosol indirect effect to Criegee + SO2 chemistry, Atmos. Chem. Phys. Discuss., 12, 33127-33163, doi:10.5194/acpd-12-33127-2012, 2012.

Spracklen, D. V, Carslaw, K. S., Kulmala, M., Kerminen, V. M., Sihto, S. L., Riipinen, I., Merikanto, J., Mann, G. W., Chipperfield, M. P., Wiedensohler, A., Birmili, W., et al.: Contribution of particle formation to global cloud condensation nuclei concentrations, Geophysical Research Letters, 35(6), -06808, 2008.

Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, Atmospheric Chemistry and Physics, 9(1), 239–260, doi:10.5194/acp-9-239-2009, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 27693, 2012.

ACPD

12, C11257–C11259, 2013

Interactive Comment