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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Large methane releases lead to strong aerosol forcing and reduced cloudiness" *by* T. Kurtén et al.

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We thank Dr Pierce for his thorough work and constructive and encouraging comments.

Answers to general comments.

1) We agree that it would be interesting to see the variability in the computed forcings due to various uncertainties. Unfortunately, the runs we have carried out do not really give us any useful information on this. The results are certainly sensitive to, among other things, the used nucleation parameterization. If we had used, for example, a binary nucleation parameterization only, the effect of methane on CDNC via OH would be smaller. In ECHAM-HAM, the SOA formation does not depend on the oxidant fields

(see below), but naturally the chosen SOA yield affects the results.

2)The direct effects were not computed in the original runs, but they are very likely small compared to the indirect effects. To check that this is the case, we ran a one-year simulation on ECHAM with direct effects included for the reference and 10xCH_4 scenario. The direct effect forcing was +0.024 W/m^2 for clear-sky and-0.077 W/m^2 for all-sky conditions – a small cooling as predicted by Dr. Pierce. Precise value would require longer runs, but this test shows clearly that the direct forcing is small compared to the indirect forcing. This will be mentioned in the revised manuscript.

Answers to specific comments:

1)The binary nucleation scheme of Vehkamäki et al. (2002) was used in ECHAM runs and the scheme of Kulmala et al. (1998) in GLOMAP runs. In our previous test runs with GLOMAP-bin model we have not observed any significant differences in cloud base CCN between the two schemes. The references will be added to the revised manuscript.

2)The SOA schemes used in ECHAM and GLOMAP (emissions, condensation) are described in Makkonen et al. (2009) and in Spracklen et al. (2005), respectively. In the ECHAM runs, no changes were made to organic oxidation – that is, the SOA yields are the same in all runs. In the GLOMAP runs, the rate of which the biogenic precursors are transformed to SOA depends on the oxidant fields, but the total SOA yield (e.g. averaged over full year) is virtually the same in different runs.

The following sentences will be added to the revised manuscript:

"ECHAM5-HAM and GLOMAP apply prescribed monthly emissions of monoterpenes (Guenther et al. (1995)) to estimate emissions of biogenic precursors for SOA. In ECHAM5-HAM, a fixed fraction of 0.15 of emitted BVOCs is assumed to form condensable SOA immediately after emission. The SOA production in ECHAM5-HAM is independent of oxidant fields. In GLOMAP, monoterpenes are oxidised to SOA in re-

actions with O_3, OH and NO_3 with fixed yields of 0.13 for each reaction. In GLOMAP, changes in oxidant fields affect the rate at which monoterpenes are transformed to SOA, but total SOA yields are nearly unaffected in different runs. Both models assume zero saturation vapor pressure for SOA products, so that SOA is partioned on the seven aerosol modes according to the relative condensation sinks of the modes."

3)Indicates will be changed to indicate, as suggested.

4)In ECHAM, no changes were made to liquid-phase oxidation – H_2O_2 concentrations are fixed. Thus, the ECHAM results cannot really be considered realistic for liquid-phase oxidation. In GLOMAP, the H_2O_2 sources and sinks are dynamically calculated, so the effects of OH on HO_2 and thus H_2O_2 formation and liquid-phase oxidation are correctly accounted for. The H_2O_2 concentration increases by approximately 63 percent in the 10xCH_4 scenario compared to the reference scenario, so the effect is significant. This will be pointed out in the revised manuscript. It is difficult to say how H_2O_2 changes would have affected the ECHAM results. Higher H_2O_2 (i.e. higher methane) would lead to higher sulfate production in clouds, and probably the changes in CCN in ECHAM would be somewhatsmaller than the results computed with fixed H_2O_2.

New references given here:

Kulmala, M., Laaksonen, A., and Pirjola, L.. Parameterizations for sulfuric acid/water nucleation rates, J. Geophys. Res.-Atmos., 103(D7), 8301–8307, 1998.

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., Bennartz, R., Feichter, J., and Kulmala, M.: Sensitivity of aerosol concentrations and cloud properties to nucleation and secondary organic distribution in ECHAM5-HAM global circulation model, Atmos. Chem. Phys., 9, 1747-1766, doi:10.5194/acp-9-1747-2009, 2009

Spracklen, D. V., Pringle, K. J., Carslaw, K. S., Chipperfield, M. P., and Mann, G. W. A global off-line model of size-resolved aerosol microphysics: I. Model development and

prediction of aerosol properties, Atmos. Chem. Phys., 5, 2227-2252, 2005.

Vehkamäki, H., Kulmala, M., Napari, I., Lehtinen, K. E. J., Timmreck, C., Noppel, M., and Laaksonen, A. An improved parameterization for sulfuric acid/water nucleation rates for tropospheric and stratospheric conditions, J. Geophys. Res. 107(D22), 10.1029/2002JD002184, 2002

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