Atmos. Chem. Phys. Discuss., 8, S6668–S6670, 2008
www.atmos-chem-phys-discuss.net/8/S6668/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates" *by* J. R. Pierce and P. J. Adams

Anonymous Referee #1

Received and published: 4 September 2008

General:

The manuscript investigates the relative roles of aerosol nucleation and primary aerosol emission in determining CCN concentrations in the global atmosphere. Very few studies of this kind have been made so far, so this work is definitely original enough to warrant its publication. Due to the large uncertainties in both nucleation rates and emissions, the authors approach the problem via a sensitivity analysis. The investigation is scientifically sound and the paper is very well written. However, I feel that the authors may not have fully covered all the aspects of this problem. As a result, I suggest that the authors consider carefully the few issues outlined below and either make

a few more sensitivity simulations or at least discuss whether this issues are relevant or not in this context.

Major comments:

The authors find that both CN and CCN concentrations are surprisingly insensitive to the atmospheric nucleation rate. I wonder whether this is a real feature of the system or whether it results from the chosen nucleation mechanism combined with the numerical treatment of the problem. My arguments are the following:

Apparently, the insensitivity of CN and CCN concentrations results from the extremely low Number Utilization Efficiency (NUE) for ternary nucleation, as compared with binary nucleation. The authors state that the low values of NUE for ternary nucleation are caused by very high nucleation rates which lead to large condensation and coagulation sinks, as well as underestimation of the contribution of self-coagulation to nuclei growth when using the parameterization scheme. I fully agree with the authors that this is the likely explanation. In the real atmosphere, however, nucleation mode particles rarely give a significant contribution to condensation and coagulation sinks (this can be easily checked out from measured particle number size distributions in different environments during new-particle formation events). Also, nucleation mode particle number concentrations are not usually high enough (except maybe in certain urban centers, power plant plumes) to make nuclei self-coagulation a very important contributor to nuclei growth. Thus, the very low NUE may be an artifact resulting from too high nucleation rates rather than a real feature of atmospheric aerosol dynamics.

I strongly recommend that the authors investigate this issue carefully. The simplest solution would be to artificially reduce the ternary nucleation rate by a fixed amount (for example by factors 10, 100 and 1000). Another option might be use a third nucleation mechanism such as the simple activation-type nucleation that depends linearly on sulfuric acid concentration.

Minor comments:

ACPD

8, S6668–S6670, 2008

Interactive Comment

The chosen ternary nucleation mechanism is sensitive to gaseous ammonia, especially at low ammonia concentrations. Is this consistent with how gaseous ammonia concentrations have been calculated in the model? The model predicts gaseous ammonia only when it is present in excess of neutralizing the sulfate aerosol. In atmosphere one would expect to see some gaseous ammonia also in acidic aerosol systems.

Is it realistic to have any fraction of SO2 to be emitted as primary particles? The model has already primary carbonaceous particle emissions and a large fraction of SO2 is anyway co-emitted with these particles.

How sensitive the system is to the assumed mean size of primarily-emitted particles?

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16291, 2008.

ACPD

8, S6668–S6670, 2008

Interactive Comment