Geophysical Research Abstracts
Vol. 13, EGU2011-3811-3, 2011
EGU General Assembly 2011

A New Hybrid Solver for Inorganic Aerosol Liquid Phase Atmospheric Gas Phase Exchange for Use in Global Models.

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Within this study the formalism, degree of accuracy and computational efficiency of a newly developed hybrid solver of inorganic gas/liquid phase exchange are presented. At its current version the solver is for use with modal aerosol models and allows for a maximum of three dissolving inorganic species. The aerosol inorganic liquid phase may contain the cations H+, NH4+ and Na+, as well as the anions HSO4-, SO42-, Cl-, and NO3-. Chemical interaction, as given by the activity coefficients, sulphuric acid dissociation and the aerosol liquid water content, is assessed according to Topping et al. (2009).

Though conceptually related, the approach of the solver is novel with respect to the existing hybrid solver of Capaldo et al. (2000) as the dynamical regime the individual aerosol modes are in is a time function that is determined at the level of the internal timestep of the solver. Decision criteria follow inter-modal mass fractionation, prospective equilibration time, and computation time efficiency considerations. Non-equilibrium modes are treated with the transient dissolution method detailed by Jacobson (1998). Substantial savings in computation time are obtained as only those modes are effectively placed in non-equilibrium that require no more than a couple of internal timesteps. Chemically stiff non-equilibrium modes are primarily equilibrated with the gas phase. Their composition is then corrected with a simple method that considers their estimated equilibration time (=pseudo-equilibration). The equilibrium solver works on two sub-solvers. For equilibration that is driven by the variation of the gas phase pressure a solver based on a differential method has been developed. All aerosol modes are equilibrated conjointly with the gas phase for a certain species, whereas inter-species equilibration is taken into account iteratively. Modes whose equilibration is driven by chemical interaction are treated with an analytical solver that supports a maximum of three diffusing species. Non-linear chemical interaction is handled with a limited number of mode-internal iterations, whereas intermodal interaction is dealt with separately with external iterations. Decision on the equilibrium sub-solver is made after each complete iterative cycle.

Apart from the standard hybrid mode, the solver may be run as well in full equilibrium and in pseudo-equilibrium. In the box model version purely dynamical runs may be performed using the Jacobson transient dissolution solver. The fully dynamical configuration serves as benchmark to assess the accuracy of the other configurations. The computational efficiency of the solver is demonstrated through the fact that hybrid runs take sensibly the same, or even less, computation time as full equilibrium runs. Under non-stiff gas phase driven conditions the hybrid configuration renders sensibly similar results to the ones obtained with full dynamics. Under increasingly stiff conditions the difference between the hybrid and the pseudo-equilibrium runs tends to vanish as modes are put in pseudo-equilibrium to save computation time. However, it is shown that the pseudo-equilibrium regime catches reasonably well the dynamics of the simulated system. The importance of non-equilibrium considerations for aerosol particle composition, as well as the capacity of the hybrid solver to catch this property, is well demonstrated throughout the tested parameter space by the difference of aerosol composition between the hybrid and the full equilibrium regimes.

References:

Capaldo, K.P., Pilinis, C., and Pandis, S.N., Atmospheric Environment, No. 34, pp. 3617-3627, 2000.
Jacobson, M.Z., Fundamentals of Atmospheric Modeling, Cambridge University Press, 1998.
Spracklen, D.V., et al., Atmospheric Chemistry and Physics, No. 5, pp. 2227-2252, 2005.
Topping, D., Lowe, D., and McFiggans, G., Journal of Geophysical Research, Vol. 114, doi:10.1029/2008JD010099, 2009.