



Global Scale Simulations of Inorganic Gas/Aerosol Exchange with a new Hybrid Solver

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A new solver of inorganic atmospheric gas phase aerosol liquid phase exchange that combines a hybrid dynamical and equilibrium approach has been implemented in the modal version of the GLObal Model of Aerosol Processes (GLOMAP), which is itself embedded into the chemical transport model TOMCAT (Spracklen et al., 2005). Dissolving inorganic species are ammonia and nitric acid. Beneath these species the model particle liquid phase contains dissociating sulphuric acid and sea salt, which is supposed to consist of sodium chloride and sodium sulphate in proportions representative to standard seawater (Millero et al., 2007). All ionic inorganic species are allowed to interact chemically via the particle pH, the activity coefficients and the particle liquid water content (Topping et al, 2009). The solver is designed to associate high accuracy with low computational expense. Based on simple criteria the solver assesses for each grid box which modes are in equilibrium and which are not. In order to save computation time the solver places some non-equilibrium stiff modes in equilibrium. The equilibrium solution for these modes is ulteriorly corrected (=pseudo-equilibrium). Annual global experiments have been carried out with the solver set either to the standard hybrid regime, or to the pseudo-equilibrium or full equilibrium regime as transient dissolution is switched off. Also, experiments with the sticking efficiency of nitric acid and ammonia set to unity have been made.

Particles containing high loads of both ammonium and nitrate show a seasonal cycle with a trend from ammonium sulphate in winter to ammonium nitrate in summer. Sea salt appears to be an effective uptaking agent of nitrate, especially when nitrate is scarce and for those particles that contain little sulphuric acid. Over the southern ocean typically only between 10 and 20% of total nitric acid is present in the gas phase for this reason. Sticking coefficients have a marginal role only, as non-equilibrium modes tend to be too large to be in the molecular regime. Dissolution of inorganics as a whole, but also as to the choice of the dynamical regime in particular is shown to play a substantial role for the number of cloud condensation nuclei available.

The hybrid solver consumes a marginal fraction of total computation time only, of the order of less than 5%, regardless of the dynamical regime the solver has been set to. Results show the relevance of non-equilibrium effects on aerosol composition, especially under circumstances of high solubility of inorganics such as low temperature, high humidity, high pollution and low aerosol load. In the equilibrium regime, as well as to a smaller extent in the pseudo-equilibrium regime, the accumulation and especially the coarse mode tend to take up the bulk of the available dissolvable matter that is present in the atmosphere. Under this circumstance small aerosol particles tend not to get neutralized far enough by ammonia for the positive feedback to nitric acid solubility to become efficient. As a result small particles contain substantially less ammonium nitrate under polluted midlatitude conditions. In the wintertime arctic there is a corresponding tendency for small particles to contain high loads of nitrate when the model is run in the hybrid mode. Due to the fact that aerosol composition depends primarily on competition among size discretized aerosol particles that have distinct dynamical properties, the variation of the gas phase composition is less pronounced when hybrid runs are compared to equilibrium ones.

References:

Millero, F.J., et al., Deep Sea Research I, Vol. 55, p. 50-72, 2008.
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.