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PD-FiTE - an efficient method for calculating gas / liquid equilibria in atmospheric aerosol particles

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Assessing the impact of atmospheric aerosol particles on the environment requires adequate representation of appropriate key processes within large scale models. In the absence of primary particulate material, interactions between the atmospheric gaseous components and particles means that the chemical nature of the particles is largely determined by the availability of condensable gaseous material, such as sulphuric and nitric acids, and by the ambient environmental conditions. Gas to particle mass transfer of semi-volatile components,driven by a difference in equilibrium and actual partial pressures above an aerosol particle, is an important factor in determining the evolving chemical composition of the particle and is necessary for predicting aerosol loading and composition. The design of an appropriate framework required for parameterizations of key variables is challenging. These thermodynamic frameworks are often numerically very complex, resulting in significant computational expense. Three dimensional chemical and aerosol transport models demand that computational expense be kept at a minimum,resulting in a trade-off between accuracy and efficiency. To calculate the equilibrium vapour pressure above a solution requires treatment of solution nonideality. This is manifest through activity coefficients of components pertinent to each condensing specie. However, activity coefficients are complex functions of the solution composition. Parameterisation of activity coefficients provides the main focus of this work largely because reducing the numerical complexity whilst retaining a good level of accuracy is very challenging. The approach presented here, the hybrid Partial Derivative Fitted Taylor Expansion (PDFiTE) (Topping et al 2008), builds on previously reported work, with an aim to derive parameters for an accurate and computationally efficient framework through coupling with a complex thermodynamic model. Such a reduction in complexity is important as it is necessary to further include an as yet unspecified number of condensing organic species, thus increasing the computational burden of any existing framework. Using this coupled approach we derive optimised model parameters describing the interaction between different chemical components, resulting in a significant increase in computational performance, in some cases giving a four fold decrease in the required number of floating point operations, whilst remaining accurate. Overall, comparisons with the most accurate inorganic activity coefficient model available (Clegg et al1998) and an existing parameterisation (MTEM) (Zaveri et al 2005) indicate that the methodology behind PD-FiTE gives an increase in accuracy for calculating the vapour pressure of all condensing gases when averaged over the parameterisation space of the system H+-NH4+-Na+-SO42–HSO4–NO3–Cl-. Following this, the reduced parameterisation is coupled into a combined chemistry / microphysical aerosol model in a dynamical simulation of aerosol composition. This demonstrates the robustness of our model and also demonstrates the implications of its use. As a simple test case we investigate the response of a typical marine aerosol passing through a polluted environment. This shows the robustness of PD-FiTE and illustrates its usefulness in capturing the fine details of important phenomena such as the outgassing of HCl in response to HNO3 uptake by sea-salt particles. Further, since semi-volatile organic compounds are ubiquitous and secondary organic aerosol is thought to be a major fraction of submicron aerosol mass, the inclusion of organic compounds into the framework will be reported.

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