Effects of mineral dust and sea salt on global nitrate concentration field

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Inorganic particulate nitrate constituents contribute significantly to the total aerosol mass; i.e. over Europe particulate nitrate accounts for about 10–20% of the total dry aerosol mass (Putaud et al., 2004). While nitrate is predominantly present in the submicron mode, coarse mode aerosol nitrate can also be produced by adsorption of nitric acid onto sea salt particles and soil particles. Naturally emitted particles affect the phase partitioning of nitrate and ammonium, especially in areas where dust or sea salt comprises a significant portion of total particulate matter, and the simulation of these effects can considerably improve model predictions. However, most thermodynamic models used in global studies lack a realistic treatment of crustal species. Furthermore, assuming thermodynamic equilibrium between the gas and aerosol phases may not be appropriate for coarse mode aerosols in global models.

The project aims to improve the representation of nitrate aerosols in a global chemistry climate model, the ECHAM/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2006), and address the shortcomings of previous models. EMAC calculates the aerosol microphysics and gas/aerosol partitioning by using the Global Modal-aerosol eXtension (GMXe) aerosol module (Pringle et al., 2010). Aerosol microphysics are treated using an extended version of the M7 modal aerosol scheme (Vignati et al., 2004), which describes the aerosol size distribution by 7 interacting lognormal modes (4 hydrophilic and 3 hydrophobic modes). An advanced dust emission module also accounts for the soil particle size distribution of different deserts worldwide (Astitha et al., 2012). Gas/aerosol partitioning is simulated using the EQSAM (Metzger and Lelieveld, 2007) and ISORROPIA-II (Fountoukis and Nenes, 2007) thermodynamic equilibrium models. The EQSAM model is a simplified, non-iterative, treatment of gas/aerosol partitioning that uses analytical expressions based on the species solubility. EQSAM can be run in a range of complexities; in this work we consider the interaction of K^+-Ca^{2+}-Mg^{2+}-NH_4^+-Na^+-SO_4^{2-}-NO_3^--Cl^--H_2O aerosol components. ISORROPIA-II uses the same suit of components and solves for the equilibrium state by considering the chemical potential of the species.

Simulation of the effects of mineral dust and sea salt on nitrate aerosol partitioning and size distribution requires an accurate description of the aerosol dynamics in the coarse mode, while it is desirable for the model to remain computationally efficient. In this project we use the hybrid method for aerosol dynamics developed by Capaldo et al. (2000) in which only the aerosol particles in the fine (nucleation, aitken, and accumulation) mode are simulated assuming equilibrium by applying EQSAM or ISORROPIA-II. For coarse particles the mass transfer differential equations are solved using the gas-phase concentrations calculated by the equilibrium step.

The proposed modeling framework combines the accuracy of the dynamic approach with the speed and simplicity of the equilibrium approach, which makes it ideal for the accurate prediction of the nitrate global concentration fields. The new model is tested in long-term simulations covering the years 2005-2008. Model predictions are compared with data from the European Monitoring and Evaluation Programme (EMEP), the Clean Air Status and Trends Network (CASTNet), the Acid Deposition Monitoring Network in East Asia (EANET)

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