Modeling the Urban- and Regional-Scale Dynamics of Gaseous Pollutants and Aerosols

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A new 3D numerical model has been developed for gas and aerosol dynamics in the regional and urban scales. The main modules of the model are atmospheric hydrodynamics, gas- and aqueous-phase chemistry, binary/ternary nucleation, condensation/evaporation, and coagulation (Aloyan & Piskunov, 2004).

The chemical model includes some 300 chemical reactions, and 44 gas-phase and 51 aqueous-phase chemical species. The kinetic models of condensation and coagulation use a non-equilibrium particle-size distribution function. The number of particle size bins in the model is 30, ranging from 0.05 to 1.5 µm. Using this model, numerical experiments were performed for condensation both on natural aerosol particles (Junge distribution) and on particles originated from binary/ternary nucleation.

The urban-scale calculations were performed for the industrial towns of Irkutsk (Russia) and Antwerp (Belgium). Along with photochemical transformation, the emphasis was on the possible formation of nucleation mode particles from precursor gases through the mechanisms of binary and ternary nucleation. The numerical calculations show that enhanced SO₂ emission levels in these towns contribute to the formation of nucleation-mode particles, with its rate being essentially dependant on seasons of the year. Then, the nucleation rate is used in the right-hand side of the kinetic equation of condensation and coagulation to solve the problem of aerosol dynamics.

The regional-scale numerical calculations were performed for the Lake Baikal area in Russia, using emission data of local industrial sources. The emphasis was on the variability of atmospheric gaseous pollutants and formation of aerosol particles having regard to their ion composition (Yermakov et al., 2007).

The calculated and measured data were compared for time periods when ammonium-sulfate particles had been registered. The numerical results show a good agreement between the calculated and measured data for the concentration of free protons in these particles. The oxidized SO₂ is largely (around 98%) stabilized in the atmosphere in the form of sulfate ions, while only some 2% is found as gaseous molecules of H₂SO₄.

An additional thermodynamic estimation of the ion composition of aerosols indicated that the specific volume of liquid water in equilibrium particles (gas-particle equilibrium) was 7x10⁻¹⁰ L/m³. It follows from here that the acidity of ammonium-sulfate particles was very large (~15 mole/l), which justifies the assumption that the acidity of aerosol particles collected during this episode is controlled exceptionally by the acidity of ammonium-sulfate particles. The agreement between kinetic calculations and observations is quite satisfactory for the mass concentration of nitrate ions (0.37 and 0.25 µg/m³, respectively).

Some discrepancy between calculated and observed data can be explained by the fact that there were no data of NH₃ natural emissions available at the Mondy background station and that remote sources of SO₂ outside the Baikal region may exist.

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