Abstract

Human activities significantly altered the atmosphere of planet Earth during the past century. Each second, tons of man-made nitrogen oxides (NO_x) and volatile organic compounds (VOC) are emitted to the air around the globe. These species can react in the gas phase and form ozone and other secondary pollutants. They can also be transferred to the condensed phase, or may be removed by deposition at the Earth surface. The man-made emissions can also interact with naturally emitted gases and particles. Since the natural emissions are often larger than the man made ones, they are of great interest to atmospheric science. This thesis investigates several aspects of formation of secondary pollutants and removal of VOC by using numerical models and by analysing environmental measurements and experimental data.

Concentrations of 9 hydrocarbon species and ozone at background station Rörvik, situated south of Göteborg, Sweden, measured continuously during a two-year period, were analysed. The statistical analysis showed groups of individual species with high internal correlation and also a distinct seasonal variation of some species. Variation with the origin of the air mass showed, that the major part of hydrocarbons and ozone at the Swedish West Coast is transported from central and Western Europe or from the UK. The measured concentrations were simulated by a Lagrangean trajectory photochemical model. The sensitivity analysis performed illustrated well the importance of some processes described by the model.

Sea-salt is emitted in huge amounts to the atmosphere in the form of solution droplets. During the time the droplets stay suspended in the troposphere, they can release active halogen species to the troposphere. Such compounds may form atomic halogens, which can initialise oxidation of VOC and hence increase the oxidation capacity of the troposphere. A numerical model CCDSSP was developed to investigate the halogen release from the sea-salt particles and its effect on VOC degradation and photooxidant formation. The model includes a detailed description of both gas and aqueous phase chemistry with focus on the chemistry of chlorine and bromine. Mechanisms of halogen release and the effect on the coastal air and on acid deposition are discussed. The model results are compared with available measurements of active halogen species and of the deposited sea-salt.

Monoterpenes are biogenic VOC, emitted in large quantities from vegetation. A common phenomenon observed in terpene oxidation experiments is formation of particulate matter. Particle size distribution and particle number concentration data from experiments with oxidation of four monoterpene species were analysed and equilibrium vapour pressures of the main condensing products were estimated. The nucleation process was parametrised, using the measured data. A model simulating gas-to-particle conversion was developed. Processes affecting the measured final particle size distributions and implications for the environmental modelling are discussed.

Keyords: numerical model; data analysis; tropospheric chemistry; VOC; NMHC; halogens; Cl-atom; Br-atom; sea-salt; gas to particle conversion; BVOC; monoterpenes;

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