Atmospheric Processes in a young Biomass Burning Plume

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Radiation and Chemistry

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Abstract

Biomass burning contributes significantly to the global budgets of a number of atmospheric trace gases and particles. The gaseous emissions are involved in photochemical formation of tropospheric ozone. The particulate emissions contribute to the direct and indirect effect of the aerosol on the radiation budget of the earth-atmosphere system.

In this thesis, atmospheric processes in young biomass burning plumes in the first tens of minutes are investigated. For this purpose, the three-dimensional (3D) atmospheric plume model ATHAM is used to simulate the evolution of a biomass burning plume. The simulation represents the situation during the Quinault prescribed fire conducted during the Smoke, Cloud, and Radiation-C (SCAR-C) experiment. The model reproduces well the general appearance of the plume and the observed aerosol mass concentrations. Remaining differences between the model results and the measurements are attributed to limited meteorological and fire emission information. Remote sensing measurements indicate a lower limit for the single-scattering albedo, ω, of the emitted biomass burning aerosol at 550 nm of 0.94. The calculation of ω based on in situ measurements results in a significant lower value of 0.85. Possible reasons for this discrepancy are discussed.

Three-dimensional solar radiative transfer simulations show that horizontal photon transfer significantly influences the actinic flux in the center of the biomass burning plume. The magnitude of this 3D radiation effect depends on the absorbing properties of the aerosol and can influence photochemistry in biomass burning plumes and other phenomena of similar dimensions, e.g., convective clouds. For the interpretation of measurements of the upward irradiance above finite plumes, the use of one-dimensional (1D) radiative transfer models is inappropriate because of the decreasing solid angle of the plume with increasing altitude. This effect cannot be taken into account in 1D radiative transfer simulations.

Atmospheric photochemistry in young biomass burning plumes leads to the formation of ozone and nitrogen reservoir species. The simulated ozone mixing ratio of about 70 ppb agrees well with the observations from the Quin-

ault fire. Significant production of nitrogen reservoir species is simulated with HNO_3 and peroxyacetyl nitrate (PAN) accounting for about $\sim 60\%$ and $\sim 30\%$, respectively. The availability of radicals is the limiting factor for photochemistry in the plume. Production of radicals is dominated by photolysis of formaldehyde ($\sim 80\%$ of the total radical production). The concentrations of the alkenes are significantly reduced by oxidation in the plume. Neglecting the emission of formaldehyde from the fire leads to unrealistic low ozone concentrations. Decreasing the emissions of nitrogen oxides as well as neglecting aerosol absorption lead to an increase in the ozone concentrations within the range of observations.

Overall, it appears that young biomass burning plumes are a highly interesting research field for several disciplines in the atmospheric science. Plumes from vegetation fires include a number of atmospheric processes and offer the potential to combine field observations and modeling studies.