Alternating Photochemical and Dark Ageing of Biomass Burning Plumes in the Large Aerosol Chamber (PHOTO-LAC)

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Plumes from wildfires may be transported over large distances from remote to populated areas or threaten sensitive ecosystems. During atmospheric transport, plumes are processed by atmospheric oxidants and complex multiphase chemistry. At high plume concentrations, ageing of biomass burning (BB) aerosol differ from its atmospheric fate at typical ambient levels.

The Large Aerosol Chamber for photochemical aging (PHOTO-LAC) with a volume of 1,800 m^3 was used to investigate the chemical composition of BB aerosols under flaming and smoldering of 300 g pine mixed with 100 g forest debris. Equivalent particulate matter (ePM) and reactive gases O_3 and NO_x were monitored by a multi-angle / multi-wavelength nephelometer and gas analyzers, respectively. Quartz fiber filter samples were collected and analyzed by Fourier-transform Ion Cyclotron Resonance Mass Spectrometry (FT-ICR MS).

After conversion of NO into NO_2 by O_3 addition, UV lights were turned for 7 h, turned off for 13 h and turned on again for 10 h, resembling day- and night-time conditions over a total experimental duration of 30 h.. In both flaming and, ePM decreased by ~15% during photochemical, but reincreased during dark ageing, exceeding initial concentrations. Eventually, consecutive photochemical ageing increased ePM further to by 15% for flaming and 35% for smoldering.

The chemical composition of the flaming and smoldering aerosols changed during the entire ageing experiment by increasing elemental O:C and organic matter to organic carbon (OM/OC) ratio as well as decreasing aromaticity. Interestingly, OM/OC increased when ePM declined during first photochemical aging, hence fragmentation reactions play a significant role. However, secondary PM formation and increasing O-content, both well-known for ageing of BB aerosol, are apparently reduced compared to smog chamber experiments at levels closer to ambient PM, emphasizing the need of laboratory plume aging studies for a better understanding of the atmospheric fate of wildfire plumes.