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## Chemical evolution of primary and secondary biomass burning aerosols during daytime and nighttime

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Fine particulate matter (PM) affects visibility, climate, and public health. Biomass burning (BB) in the forms of residential wood burning, wildfires, and prescribed burning is a major source of primary and secondary organic matter (OM, an important fraction fine PM), and brown and black carbon (BrC and BC). The contribution of BB to the atmospheric fine PM is only expected to increase in the foreseeable future. Recent studies have highlighted the enhancement in the biomass burning organic aerosol (bbOA) concentrations with aging and reported on the chemical composition of the secondary biomass burning organic aerosol (bbSOA) formed under different conditions. However, the chemical processing of the primary biomass burning organic aerosol (bbPOA) with aging is not well characterized. This chemical processing can potentially alter the chemical composition of bbOA drastically and render its identification and quantification in the atmosphere difficult.

We used aerosol mass spectrometry (AMS) and Fourier transform infrared spectroscopy (FTIR) as two complementary methods to quantify the bbPOA aging in this study. AMS measures the bulk composition of OM with a relatively high temporal resolution but provides limited parent compound information due to the extensive fragmentation. FTIR, carried out on PTFE filter samples, provides detailed information about the functional group composition of the OM and certain bbOA markers at the expense of a relatively low temporal resolution. In a series of aging experiments at the Center for Studies of Air Qualities and Climate Change (C-STACC), primary emissions from wood and pellet stoves were injected into an environmental simulation chamber. Primary emissions were aged using hydroxyl and nitrate radicals simulating the atmospheric day-time and night-time oxidation. A high-resolution time-of-flight (HR-TOF) AMS was used to identify the composition of non-refractory PM<sub>1</sub>. PM<sub>1</sub> was also collected on PTFE filters before and after aging for the off-line FTIR analysis.

AMS and FTIR agreed well in terms of the measured bbOA mass concentrations, elemental ratios, and the evolution of biomass burning tracers. We developed a procedure to quantify the bbPOA aging using AMS and FTIR. Using AMS, we found that up to 17 % of the bbPOA mass underwent some form of transformation with aging. This transformation was more intense under day-time conditions. FTIR detected a more extensive oxidation (up to two times that of AMS), suggesting a substantial processing of bbPOA, and revealing the limitations of AMS to capture bbPOA aging due to the extensive fragmentation. Different bbOA-related ion fragments were observed to decay at different rates under different conditions (e.g., oxidants and relative humidity). These different decay rates can potentially be used to identify the extent of bbPOA aging in the atmosphere. The bbSOA formed during the daytime oxidation was dominated by acid contributions, resembling certain atmospheric biomass burning samples. The unique, acid-dominated FTIR spectrum of bbSOA can potentially be used as another indicator of the aged bbOA in the atmosphere.