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

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Keywords: organic aerosol, biomass burning, FTIR, AMS, heterogeneous reactions.
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Organic matter (OM) can constitute more than half of fine particulate matter (PM) and affect climate and human health. Natural and man-made biomass burning is an important contributor to primary and secondary OM (POA and SOA) with an increasing trend.

Aerosol mass spectrometry (AMS) and Fourier transform infrared spectroscopy (FTIR) are two complementary methods of identifying the complex chemical composition of OM in terms of mass fragments and functional groups, respectively. AMS offers a relatively higher temporal resolution compared to FTIR (performed on PTFE filters). However, the interpretation of AMS mass spectra remains complicated due to the extensive molecular fragmentation.

In this study, we used collocated AMS and FTIR measurements to better understand the evolution of biomass burning POA and SOA due to different mechanisms of chemical aging (e.g., homogeneous gas-phase oxidation and heterogeneous reactions). Primary emissions from wood and pellet stoves were injected into a 10 m^3 environmental chamber located at the Center for Studies of Air Qualities and Climate Change (C-STACC) at ICE-HT/FORTH. Primary emissions were aged using hydroxyl and nitrate radicals with atmospherically relevant exposures. PM_1 was analyzed by a high-resolution time-of-flight (HR-ToF) and was also collected on PTFE filters over 20-minute periods before and after aging for off-line FTIR analysis.

AMS and FTIR measurements agreed well with regards to the concentration of OM and some biomass burning tracers (levoglucosan and lignin; Yazdani A., 2020b) and the OM:OC ratio. Chamber wall loss rates were estimated using AMS OM concentration and were used to split the contribution of POA and SOA. The estimated FTIR and AMS spectra of SOA produced by reactions of biomass burning volatile organic compounds (VOCs) with OH were found to have prominent acid signatures. Organonitrates, on the other hand, appeared to be important for SOA produced by the nitrate radical. We found that with continued aging, SOA evolves and becomes similar to the oxygenated OA (OOA) in the atmosphere. We also found that POA composition also evolves with aging. Our estimates show that up to 10 % of POA mass undergoes aging. Biomass burning tracers such as lignin and levoglucosan in addition to hydrocarbons are among the POA compounds that are lost the most in biomass burning POA (up to 6 times more than OM decrease due to chamber wall losses; Fig. 1). This diminution is observed for both semi-volatile (levoglucosan and hydrocarbons) and non-volatile (lignin) POA species, implying the importance of gas-particle partitioning, heterogeneous reactions, and photolysis for POA evolution in the atmosphere. This result can be important since chemical transport models usually do not consider POA heterogeneous reactions.

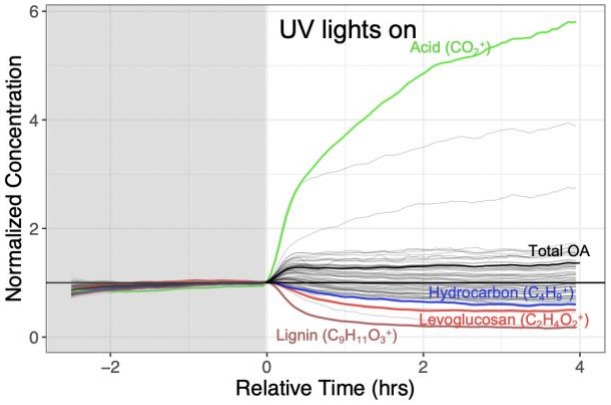

Figure 1. Trends of individual AMS mass fragments (with contribution to OM> 0.3 %) during aging with UV (starting from time zero). All mass fragments have been normalized by their concentration before the with start of aging and corrected for the chamber wall losses. Important mass fragments are shown in color.

This work was supported by the project PyroTRACH (ERC-2016-COG) funded from H2020-EU.1.1. - Excellent Science - European Research Council (ERC), project ID 726165 and funding from the Swiss National Science Foundation (200021_172923).

References

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