

Wood-burning emissions within a continuous-flow photooxidation reactor: Soot-Particle Aerosol Mass Spectrometer characterization

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Wood-burning stoves, used worldwide for heating and cooking, can be an important atmospheric source of primary and secondary aerosols. Current legislation restricts only the primary aerosol emissions of such stoves, which include primary organic aerosol particles (POA), ash, and soot. Such legislation neglects the potential formation of secondary organic aerosol (SOA), which may nucleate or condense after the atmospheric oxidation of volatile organic compounds. For example, smog chamber photooxidation experiments have observed wood-burning SOA concentrations up to four times higher than the corresponding POA emissions (Grieshop et al., 2009; Heringa et al., 2011).

To characterize the SOA-formation potential of wood-burning stoves and similar emissions, Keller and Butscher (2012) have developed a continuous-flow photooxidation reactor for monitoring potential SOA in real-time. The short residence time (seconds) of this chamber allows the time-resolved measurement of potential SOA. However, the small chamber also necessitates higher-than-ambient irradiation conditions.

Here, we present results from high-resolution Aerosol Mass Spectrometer (AMS) and Soot-Particle AMS (SP-AMS) analysis of wood-stove emissions under three different conditions: filtered and photooxidized (“SOA”), unfiltered and photooxidized (“OxPOA”), and unfiltered emissions (“POA”).

Figure 1 shows the increase in aerosol oxidation after photooxidation, as measured by the AMS. The figure shows the difference of the normalized SOA and POA organic mass spectra, averaged over multiple burns. High-resolution analysis has been used to identify organic species as C_xH_y (green), $C_xH_yO_z=1$ (purple), or $C_xH_yO_z>1$ (pink). However, data are presented on a unit-mass scale for clarity. As shown, the relative intensity of CO_x ions was higher for SOA, whereas C_xH_y was higher for POA. The pie charts (inset) summarize all three cases, showing $C_xH_yO_z$ increasing and C_xH_y decreasing, from left to right. For POA, the contribution of hydrocarbons C_xH_y was more than double that of SOA. Also, SOA contained more $C_xH_yO_z>1$ relative to OxPOA. Elemental analysis indicated that the shift of H:C and O:C ratios due to oxidation was comparable to, but significantly greater than, observations in Mexico City (Heald et al., 2010).

In these experiments, refractory species (those involatile below 600 °C) were also characterized with an SP-AMS laser equipped on the same AMS. High-resolution SP-AMS analysis yielded similar results to the

AMS, with the major spectral change due only to pure-carbon ions (C_x). Detailed AMS and SP-AMS results will be presented and compared.

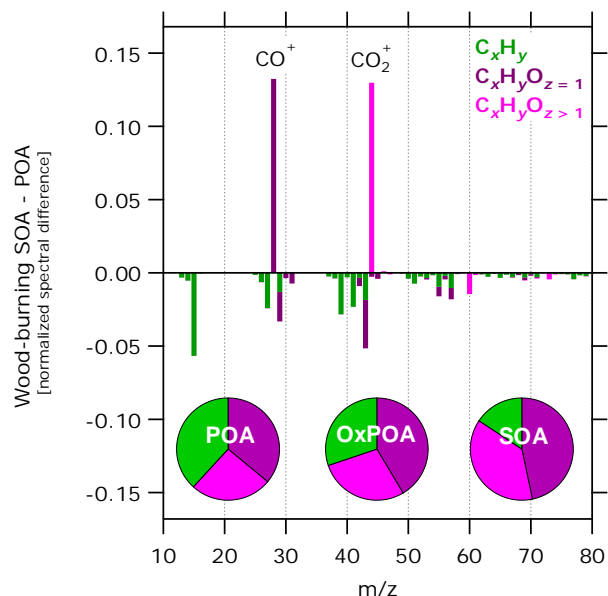


Figure 1. AMS difference mass spectrum for wood-stove SOA minus POA. Inset: pie charts of the organic composition for the three cases.

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