Laboratory and Ambient Studies of Water Uptake by and Optical Properties of Wildfire Smoke: Role of Fuel Chemistry, Combustion Phase, and Age

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Abstract
Carbonaceous aerosols (CAs) from biomass burning (BB) have increased substantially with the observed warming and drying of the US. While wildfires are projected to intensify in the future missing knowledge of BB CAs hampers assessments. Observations show that warming effects of BB CAs can dominate over cooling effects due to enhanced light absorption by internal mixing. However, if internal mixing reduces the aerosol lifetime it would lower their atmospheric burden. We report observations of BB smoke to help elucidate mechanisms that control this tradeoff.

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Laboratory Results: Hygroscopicity
The mean hygroscopicity of BB CAs from various fuels ranged from nearly hydrophobic (φBB = 1) to very hydrophilic (φBB = 2.1) values typical of pure diquelscent salts. We also measured some φBB values <1 indicative of particle collapse at high RH. The k values varied from 0.004 to 0.18 and correlated well with fuel and smoke inorganic content. Invasive halophytes with high salt content exhibit greater water uptake than native coniferous species with low inorganic content. Combustion temperature and phase play a secondary role. High temperature ignition methods create flaming conditions that enhance hygroscopicity while lower temperature smoldering conditions diminish hygroscopicity. We correlate the measured smoke or wood inorganic content with k to construct an empirical relation.

Laboratory Results: Optical Properties
We measured light scattering and absorption at 405, 532 and 780nm with a photoacoustic extinction spectrometer (PASS-3), refractory CA mass with a SP2, and composition with a SP-AMS. The mean hygroscopicity of BB CAs from various fuels ranged from nearly hydrophobic (φBB = 1) to very hydrophilic (φBB = 2.1) values typical of pure diquelscent salts. We also measured some φBB values <1 indicative of particle collapse at high RH. The k values varied from 0.004 to 0.18 and correlated well with fuel and smoke inorganic content. Invasive halophytes with high salt content exhibit greater water uptake than native coniferous species with low inorganic content. Combustion temperature and phase play a secondary role. High temperature ignition methods create flaming conditions that enhance hygroscopicity while lower temperature smoldering conditions diminish hygroscopicity. We correlate the measured smoke or wood inorganic content with k to construct an empirical relation.

Ambient Fire Observations
We sampled smoke from 5 wildfire plumes that included fresh plumes from 4 small to moderate sized fires and control burns near Los Alamos, NM and an aged plume (>3 days) from the large labor day Pacific NW fires in 2017. The Los Alamos fires burned coniferous and mixed-coniferous species including ponderosa, pinion, juniper, and deciduous aspen. Ambient light scattering measured during the larger smoke impacted events, occurring on June 27th, was determined to have a k ext = 0.024 ± 0.005 (φBB = 1.15). Using laboratory calculated hygroscopicity for the aforementioned fuels, a linear combination of water uptake was calculated as k ext = 0.022 ± 0.011 (φBB = 1.15). This good agreement is promising for the use of mixing rules to predict water uptake by biomass burning from fuel composition data. The effects of aging the long-range Pacific NW fire smoke that burned similar fuel was small. However, we do see variability in k ext for local fires, particularly presnicrinated burns could that result from differences in fuel type and combustion phase that we are investigating.

Smoke f(RH), Optical & SP-AMS Measurements
We measured light scattering and absorption at 405, 532 and 780nm with a photoacoustic extinction spectrometer (PASS-3), refractory CA mass with a SP2, and composition with a SP-AMS. We find that the observed hygroscopicity was intimately linked to the chemical composition of the fuel. We compare our laboratory results with ambient fire results that spanned smoke ages of less than an hour and 3 days to evaluate mixing rules and the chemical changes. Our goal is to develop a mechanistic framework to predict water uptake and optical properties of smoke as a function of fuel, fire intensity and age.

Conclusions
• We demonstrate that fuel inorganics drives smoke hygroscopicity to 1st order and report an empirical relation between them for use in models.
• Flaming fire smoke exhibits higher hygroscopicity than smoldering fires with other detectable 2nd order effects: Large Inorganic Fraction >> Little Inorganic, Flaming >> Smoldering, Leaves >> Woods and Barks Deciduous >> Evergreen, and Invasive >> Natives.
• Mixing rules can predict k ext for fresh ambient wild-fire smoke. Prescribed burns k ext differs from wildfires and aging can also effect it.
• Optical properties of our burn experiments can be interpreted using Mie theory. For grasses, we observe large brown carbon aerosols at ignition that transition to mixed organics and finally black carbon at the end.
• Our new SP-AMS quantifies aging effects on chemistry of BB CAs.


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