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Environmental Research Letters

LETTER

Does chronic nitrogen deposition during biomass growth affect atmospheric emissions from biomass burning?

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Abstract

Chronic nitrogen deposition has measureable impacts on soil and plant health. We investigate burning emissions from biomass grown in areas of high and low NO_x deposition. Gas and aerosol-phase emissions were measured as a function of photochemical aging in an environmental chamber at UC-Riverside. Though aerosol chemical speciation was not available, results indicate a systemic compositional difference between biomass grown in high and low deposition areas. Aerosol emissions from biomass grown in areas of high NO_x deposition exhibit a lower volatility than biomass grown in a low deposition area. Furthermore, fuel elemental analysis, NO_x emission rates, and aerosol particle number distributions differed significantly between the two sites. Despite the limited scale of fuels explored, there is strong evidence that the atmospheric emissions community must pay attention to the regional air quality of biomass fuels growth areas.

1. Introduction

Aerosols play a major role in the Earth's climate system. Aerosols can have health effects on humans and local ecosystems, can affect regional visibility, and have a large impact on the Earth's radiative forcing balance (Forster *et al* 2007). One major source of aerosol and trace gases to the atmosphere is biomass burning (Crutzen and Andreae 1990, Ramanathan *et al* 2001, van der Werf *et al* 2006, Akagi *et al* 2011, Hardy 2014). Biomass burning emits a complex mixture of gases and aerosol and emissions can vary in both composition and amount. Previous work has shown that the composition and moisture of the fuel, packing ratio of the fire, and the fire regime (e.g. flaming, smoldering, or mixed phase) affect the emissions properties of biomass burning (Ward 2003, Petters *et al* 2009, Thonicke *et al* 2010, Hosseini *et al* 2013). Additionally, photochemical aging of biomass burning emissions can effect aerosol properties such as hygroscopicity, light absorption, surface activity, and

morphology (Hennigan *et al* 2011, Engelhart *et al* 2012, Giordano *et al* 2013, Zhong and Jang 2014, Giordano *et al* 2015).

One caveat of the work done on biomass burning to date is that measured emissions vary greatly between experiments. Individual fire conditions such as differences in fire energy (fraction of fuel that burns flaming versus smoldering) and nitrogen content of fuels contribute to high uncertainties in both emission amounts and properties. Satellites can be used to better constrain the effects that fire energy has on NO_x emissions (Mebust *et al* 2011, Schreier *et al* 2014, Tanimoto *et al* 2015). However, constraining the impact that nitrogen content of fuels has on burning emissions must be done through carefully controlled laboratory experiments.

Wildland fires burn the accumulated dead material (foliage, wood) that is deposited in forests through normal tree growth processes. The recently deposited material is called litter while the older, partially decomposed accumulation is sometimes

referred to as duff. Forest litter is a primary fuel source for wildfires and plays a large role in nutrient cycling between vegetation, duff, and soil (DeBano 1991, Kauffman *et al* 1995). One factor that has not been studied extensively is the impact of local air quality on wildland fire smoke emissions. A limited number of studies have reported smoke emissions resulting from fires in areas of known pollution. In two studies, the Lodi prescribed burn located in the San Gabriel Mountains near Los Angeles, CA and fires in the mountains surrounding Mexico City, Mexico, high levels of nitrogen compounds were observed in the smoke emissions (Hegg *et al* 1987, Yokelson *et al* 2007) and in the stream water run off (Riggan *et al* 1994). In the case of these two studies, ambient sampling was performed to characterize emissions from these polluted areas; however designed experiments to directly determine the effect of air pollution on smoke emissions have not been performed to our knowledge.

Though there have been no investigations into the effects of regional air quality on biomass burning emissions, chronic nitrogen deposition has been shown to have measureable, long term effects on ecosystems. Elevated atmospheric nitrogen encourages the formation of nitrogen oxides and ammonium which provides biologically available forms of nitrogen to ecosystems (Galloway *et al* 2004). While most ecosystems are defined as biologically nitrogen limited, an increase in biologically available nitrogen can lead to nitrogen saturated environments (Aber *et al* 1998). Nitrogen saturation has multiple deleterious effects on ecosystems including, but not limited to, a decrease in biodiversity, acidification of soil, an increase of secondary stressors on plants (especially through the formation of ground level ozone), and a negative impact on water quality and aquatic systems (e.g. Fenn *et al* 1998, Carroll *et al* 1999, Bowman *et al* 2008, Bobbink *et al* 2010). The effects of nitrogen saturation have been shown to have measurable impacts on biomass foliar N:C, N:P, and N:S ratios, foliar nitrate and ammonium ratios, soil C:N ratios, and soil nitrogen flux and mineralization (Fenn *et al* 1996). Ecosystems located near urban environments, such as Los Angeles, are especially susceptible to nitrogen saturation (Fenn *et al* 1996, 2000). Though nitrogen saturation has been an active area of research for ecologists and botanists for many years, the effects of nitrog en saturation on biomass burning emissions have yet to be explored.

A gradient of elevated ozone and nitrogenous pollutants has been documented in the San Bernardino Mountains, located east of the San Gabriel Mountains. The location offers an opportunity to compare emissions from wildland fuels subjected to different levels of chronic air pollution. The area is subject to oxidant and particulate air pollution from the Los Angeles air basin and has been of particular interest to plant and forest health researchers for over 50 years (Cobb and Stark 1970, Miller 1973, Fenn and Bytnerowicz 1993,

Grulke *et al* 1998, Arbaugh *et al* 2003). The pollution gradient is such that the concentration decreases from west to east (as the distance from Los Angeles increases) and has been well characterized (Miller *et al* 1986, Grulke *et al* 1998). Increased air pollution, oxidant exposure, and nitrogen deposition rates in the area have been shown to negatively affect the health of the mixed conifer forest that grows in the area (McBride *et al* 1975, Pronos *et al* 1978, Fenn *et al* 1996, Miller *et al* 1996, Grulke *et al* 1998).

The current state of scientific understanding is that elevated nitrogen deposition changes biomass health and nutrient content. Unfortunately, no study has yet to identify if such changes have a measureable impact on the emissions from biomass burning. This manuscript examines what, if any, effects local air quality has on the emissions resulting from the burning of biomass grown in nitrogen polluted and clean areas. Wildland fuels from the mixed conifer forest in the San Bernardino Mountains are burned and exposed to simulated daylight in an environmental chamber. Aerosol and gas-phase properties of the emissions are presented.

2. Experimental methods

Fuel samples were collected from two sites that were a part of the San Bernardino Mountains Gradient Study (SBGS, Arbaugh *et al* 2003) in southern California. The Camp Paivika (CP) site has a yearly nitrogen deposition rate of the site to be 31–45 kg N ha^{-1} and an average ozone concentration of 0.09 ppm-h (90 ppb-h) (Allen *et al* 1996, Grulke *et al* 1998, Fenn *et al* 2003) ('high' exposure). The Camp Osceola (CAO) site is a nitrogen limited site with a yearly deposition rate of 5–13 kg N ha^{-1}. CAO also experiences lower average ozone concentration (0.07 ppm-h, 70 ppb-h) and lower deposition rates and atmospheric concentrations of species such as NH_4^+, SO_2^-, NO_3^-, and HNO_3 than CP (Fenn and Bytnerowicz 1993, Grulke *et al* 1998, Fenn *et al* 2003) ('low/ intermediate' exposure). The forest at both locations is typical mixed conifer composed of ponderosa pine (*Pinus ponderosa* Lawson and C Lawson), Sierra white fir (*Abies lowiana* (Gordon and Glend.) A Murray bis), incense cedar (*Calocedrus decurrens* (Torr.) Florin), and California black oak (*Quercus kelloggii* Newberry). Plant names are presented here in the standard form contained in the International Code of Nomenclature for algae, fungi, and plants (ICN) where an author citation is provided after the Latin name (Wiersema *et al* 2015).

Care was taken during the collection of biomass to ensure that forest litter was collected from under a wide and approximately equal number of tree species at both sites. Additionally, after collection the samples were well-mixed in their containers. Separating individual coniferous needles between species would not

have been feasible but the authors believe each burn was a representative and equal mix of different tree species. The measurements presented here therefore represent an ensemble average of 'polluted' and 'clean' forest litter.

At the time of collection in late October 2013, both sites had been subject to similar temperatures and precipitation levels. While precipitation gauges are not available at either site, precipitation in the collection areas had been (as it continues to be) incredibly low. The closest NOAA rainfall observation post to the clean site is located at Big Bear Lake, CA. At Big Bear, NOAA observed 0.00 inches of rainfall in September 2013 and 0.07 inches of rainfall in October 2013. The polluted site is more influenced by air from the Los Angeles Basin. The measured precipitation from NOAA for downtown LA is lower at 0.00 and 0.02 inches for September and October 2013, respectively (http://cnrfc.noaa.gov/). Therefore, it is unlikely that precipitation could have large impacts on the deposition of air pollutants, and washing them down through the forest floor litter layer between the two collection sites.

Samples from the top of the litter-layer, primarily composed of conifer needles, were collected from both SBGS collection sites. The forest litter was collected from locations away from main roads and a minimum of 5 m away from secondary roads to avoid pollution and dust from vehicles that use the roads. The samples were collected by hand and stored in clean plastic containers. Special attention was paid to exclude materials such as cones and branches. The fuels were stored in their containers at ambient conditions for up to three weeks until burned. Fuel moisture content was measured once according to ASTM standard for each fuel. Both fuels contained negligible water content.

Elemental analysis of polluted and clean fuels was determined from triplicate samples at the University of California, Riverside Environmental Sciences Research Lab using a Flash EA 112 series elemental analyzer[5]. Samples were strained through 100 μm mesh sieve, and roughly 10 μg were measured for each sample. The Flash EA 112 determines the weight percent composition of carbon and nitrogen species. Aspartic acid is a standard compound used for comparison of results.

The experimental facility, the Atmospheric Processes Lab at the University of California, Riverside College of Engineering—Center for Environmental Research and Technology, consists of a wood stove connected to a large environmental chamber (Giordano *et al* 2013). A series of six total experiments (three polluted and three clean fuels, alternating) were conducted. Prior to an experiment, the chamber was flushed and filled with filtered air. 100 g of a fuel were

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[5] The use of trade or firm names in this publication is for reader information and does not imply endorsement by the US Department of Agriculture of any product or service.

measured and burned in the wood-burning stove outside of the lab. Smoke from the burns was diluted and injected into the 12 m^3 Teflon® reactor through a Venturi tube injection system. The smoke was injected with a 10:1 total flow to sample flow ratio through a 7 m copper transfer line. The injection was completed when combustion was finished and the remaining ash was collected and weighed. 100 g was chosen because it yielded a final particle concentration in the reactor of 50–100 μg m^{-3} which is within range of ambient and plume-like biomass burning conditions (Hennigan *et al* 2011). The first hour is used to establish a well-mixed reactor and observe the effects of no photochemical ageing. The reactor is surrounded by ultraviolet lights with peak intensity of 350 nm (350 BL, Sylvania) which were turned on approximately 1 h after the injection was completed. UV lights simulate atmospheric photochemical ageing known to produce secondary pollutants. Each experiment was operated until the reactor was depleted (generally 5–6 h). Fire regimes are confirmed via a flame-integrated modified combustion efficiency (MCE $\equiv (\Delta CO_2)/(\Delta CO_2 + \Delta CO)$) (Ward and Radke 1993). The MCE is presented here as a fire-integrated measurement for each individual burn. Fuel consumption was determined by calculating the difference in fuel mass at the start and end of each experimental run. Fuel consumption is used to report an emission-like factor. This normalization is useful for placing the experiments on the same basis for comparison.

Instruments sampled both gas-phase and aerosol phases of the smoke. Gas-phase monitoring included teledyne carbon monoxide (CO), ozone (O$_3$), and NO$_x$/NO analyzers as well as a Licor carbon dioxide (CO$_2$) analyzer. Aerosol-phase measurements included particle size distributions with a scanning mobility particle sizer (SMPS, TSI 3080/3081), particle mass measurements with an aerosol particle mass analyzer (APM, Kanomax), and particle volatility with a home-built volatility-tandem differential mobility (V-TDMA) analyzer (Wang and Flagan 1990, Orsini *et al* 1999 McMurry *et al* 2002). The V-TDMA thermodenuder was set to a temperature of 100 °C and set to select 50, 100, 150, and 200 nm particles for the first DMA and a volume fraction remaining (VFR) calculated from the second DMA operated in scanning mode.

The APM system used here is the APM-DMA system described in Malloy *et al* (2009). Five different particle masses in the 50–200 nm electrical mobility particle size range were selected. The effective density of the aerosol was obtained by fitting the power-law function (Park *et al* 2003, Xue *et al* 2009, Nakao *et al* 2011):

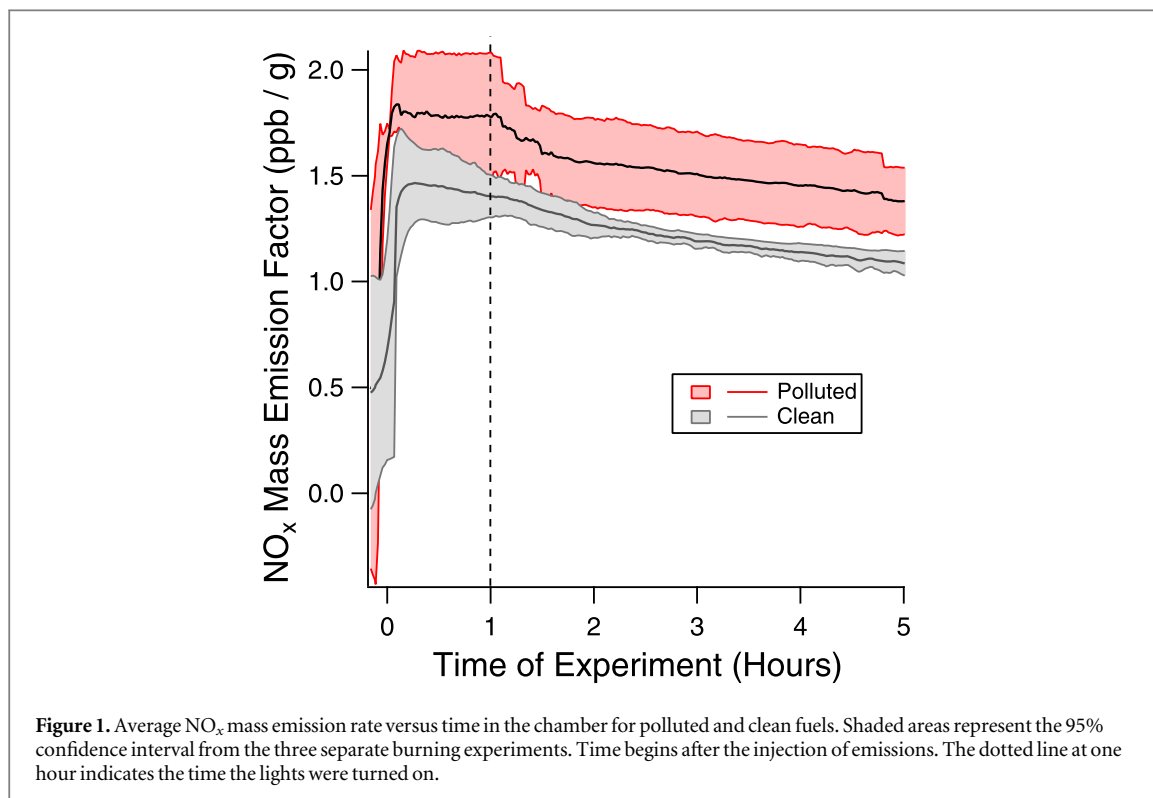$$\rho_{\text{eff}} = Cd_{\text{m}}^{D_{\text{f}}'-3}, \tag{1}$$

where ρ_{eff} is the effective density of particles, C a constant, d_{m} is the mobility diameter of the particles, and D_{f}' is the fractal-like dimension. Here, the fractal-

Table 1. Results of elemental analysis.

Sample	% Carbon wt	% Nitrogen wt	C : N	
Aspartic acid	36.035 ± 0.025	10.462 ± 0.047	1.337 ± 0.006	(4:3)
Clean	50.777 ± 0.036	1.080 ± 0.015	12.965 ± 0.182	(13:1)
Polluted	49.962 ± 0.247	1.179 ± 0.012	11.873 ± 0.123	(12:1)

Figure 1. Average NO_x mass emission rate versus time in the chamber for polluted and clean fuels. Shaded areas represent the 95% confidence interval from the three separate burning experiments. Time begins after the injection of emissions. The dotted line at one hour indicates the time the lights were turned on.
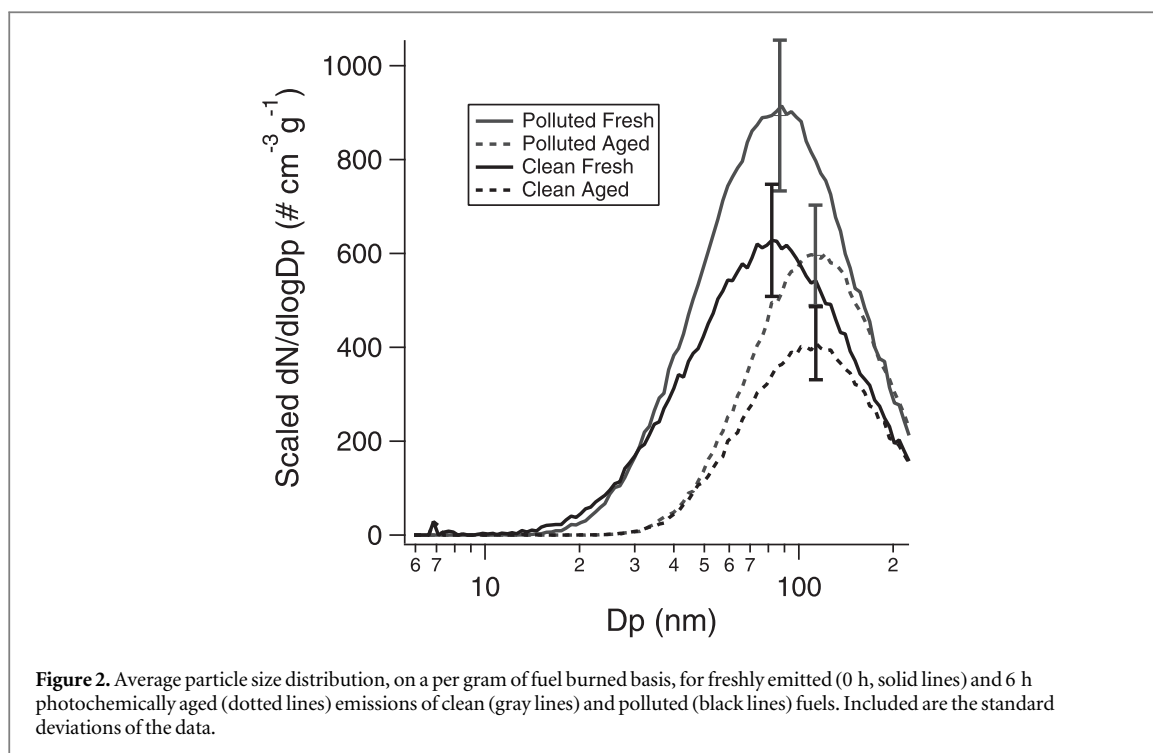
like dimension is a measure of the deviation from sphericity of the particle where $D_f' < 3$ indicates a non-spherical particle. Additional information on the fractal nature of biomass burning particles is reported in Giordano *et al* (2015).

3. Results and discussion

Elemental analysis showed a statistical difference in fuel composition (table 1). Fuels from low NO_x deposition areas have a larger C:N ratio than fuels collected in high NO_x areas. This finding is consistent with previous literature reporting that plants grown in areas of high nitrogen deposition rates have higher nitrogen content than plants grown in cleaner areas (e.g. Baddeley *et al* 1994). Up to an ~10% molar difference is observed in samples collected in high NO_x deposition areas compared to low NO_x areas. For simplicity, the fuels tested will be referred to as 'polluted' (from chronic high NO_x deposition areas) and 'clean' (low NO_x).

Burns were conducted in the smoldering and mixed phase fire regime. An MCE range of 0.85–0.91 was obtained for the series of six experiments. Figure 1 shows the average NO_x emitted during the burn for both the polluted and clean fuels, written as a mass emission-like factor: concentration emitted scaled by the amount of fuel consumed ($ppb\,g^{-1}$). The initial emission factor is calculated as a background corrected, fire-integrated assessment of the entire injection into the environmental chamber. The shaded areas of the figure represent the standard deviation of the averages and the time the lights were turned on is indicated. The polluted fuels exhibited higher initial NO_x emissions ($1.7\,ppb\,g^{-1} \pm 0.3\,ppb\,g^{-1}$ 95% confidence interval) than the clean fuels (1.3 $ppb\,g^{-1} \pm 0.15$ 95% confidence interval) for the same amount of mass burned. The difference in initial NO_x emissions may not be statistically significant but that may be due to the large range of NO_x emissions of the clean fuels. However, after photochemistry was initiated (one hour after injection), the NO_x emission factors for the clean fuels collapse onto each other. After approximately one hour of photochemical aging, the polluted and clean fuels become statistically significantly different (1.55 ± 0.2 $ppb\,g^{-1}$ 95% confidence interval for polluted fuels compared to 1.3 ± 0.03 $ppb\,g^{-1}$ 95% confidence interval for clean fuels). The results for NO were similar with polluted fuels having a higher mass emission-like factor than clean fuels (results not included here). The emission of

Figure 2. Average particle size distribution, on a per gram of fuel burned basis, for freshly emitted (0 h, solid lines) and 6 h photochemically aged (dotted lines) emissions of clean (gray lines) and polluted (black lines) fuels. Included are the standard deviations of the data.

higher NO_x from polluted fuels was consistent with larger elemental nitrogen fuel composition measured by elemental analysis. Thus, the climatic impacts of releasing more NO_x and NO from biomass burning of polluted fuels may be far reaching. An increase of tropospheric nitrogen oxides would have substantial impacts on regional OH concentrations. Up to a six-fold increase in OH has been predicted solely from biomass burning and deforestation in the tropics (Crutzen and Andreae 1990). If biomass from heavily polluted urban areas, such as Mexico City, releases 30% more NO_x as we show here, then current models may be under-predicting the impact of biomass burning if only emission factor values for clean fuels are used in the models.

Figure 2 shows how the average particle size distributions released from polluted and clean fuels changed with 6 h of photochemical aging. As with NO_x, the polluted fuels emitted more particles on a scaled basis but the difference was not statistically significant. The distributions for the fresh (0 h) and 6 h aged emissions were similar. When considering both fuels, the freshly emitted particle size distribution peaked at an average of 85 ± 5 nm while the photochemical aging process and coagulation grows the particle distribution peak to 110 ± 6 nm. MCE indicated that burning was consistently in the smoldering/mixed range and particle size distribution remained constant for both fuels (figure 2). Figure 2 agrees with previous work that shows the particle size distributions from biomass burning are much more sensitive to the fire properties (e.g., MCE) of the burns than the species being burned (Hosseini *et al* 2010). However, particle concentrations from polluted fuels had higher particle emission

rates than clean fuels. As with NO_x, utilizing a single emission factor for biomass burning may not capture the reality of particulate emissions if considering, for example, locations close to and far away from polluted megacities.

Both gas-phase and aerosol-phase measurements confirmed that chronic local air pollution during biomass growth can measurably alter the emissions produced by biomass burning. Measureable differences in specific particulate physical properties, volatility and density, existed in these experiments. Figure 3 shows the average VFR at 100 °C of both the polluted and clean fuels as a function of aging in the chamber. The 95% confidence interval is again shown as the shaded regions surrounding the averages. The periodicity that is exhibited in both fuels is due to the operating mode of selecting discreet particle sizes in the VTDMA system. The trends should be looked at as a whole. Both fuels exhibited an increasing VFR with photochemical aging indicating a change in the composition of the aerosol, consistent with literature (Kalberer *et al* 2004). Both fuels increased their VFR at approximately the same rate throughout the photochemical aging. Initially, the polluted fuels were composed of more low volatility products (VFR = 0.38 ± 0.05 95% confidence interval) than the clean fuels (VFR = 0.27 ± 0.05 95% confidence interval). As photochemical ageing occurred, polluted fuels became increasingly nonvolatile as compared to clean fuels (VFR = 0.7 as compared to VFR = 0.5, respectively). Changes in particle volatility indicate that polluted fuels had a different aerosol composition than the clean fuels both before and after oxidative aging. It is possible that the decreased volatility of polluted fuels
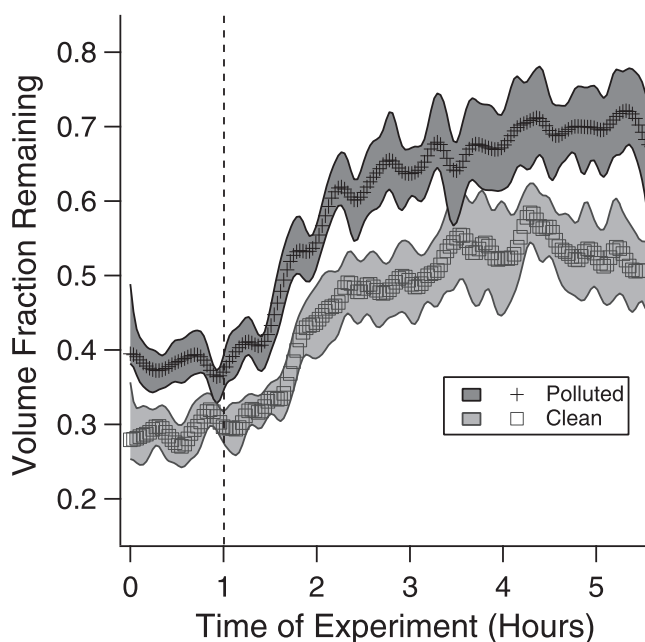
Figure 3. Volume fraction remaining (VFR) of the aerosol versus time of experiment. The average values for clean fuels (gray) and polluted fuels (black) are shown with their 95% confidence intervals as shaded areas. The dotted line represents the time the lights are turned on in the experiment. Aerosol from polluted regions were less volatile than cleaner fuels. Photochemical ageing produced aerosol of lower volatility.
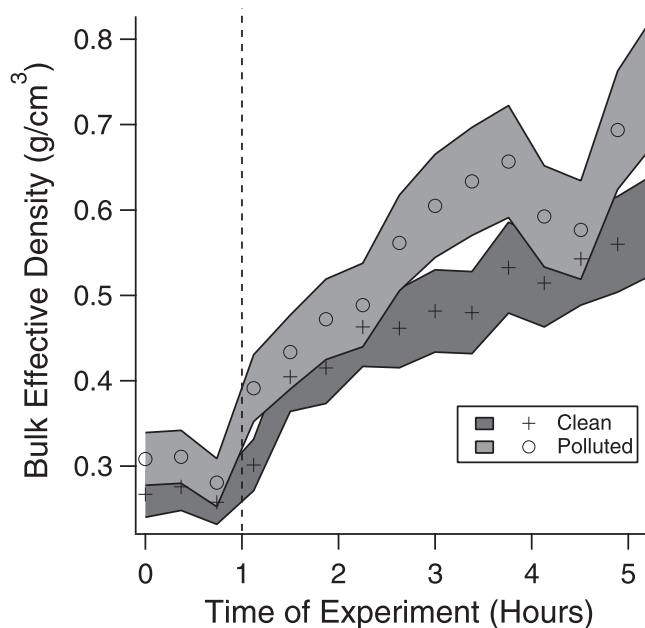

Figure 4. Average bulk effective density for clean fuels (gray) and polluted fuels (black). The standard deviations are represented as shaded areas. The dotted line represents the time the lights were turned on in the experiment. Photochemical ageing modified the effective bulk density of aerosol emitted from biomass burning.

was related to changes in initial gas-phase oxidant concentration and gas-particle partitioning of NO/NOx with photochemical aging. Further aerosol chemical speciation is required. Figure 4 shows the average calculated effective density from the APM system for the clean and polluted fuels. Both fuels were emitted at low bulk effective densities (0.2–0.3 g cm^{-3}) and exhibited effective densities that increased with time.

The difference between the final measured effective densities of the polluted and clean fuels were 0.8 g cm^{-3} and 0.55 g cm^{-3}, respectively. The density results agreed well with the volatility data presented in figure 3. As in previously published work, freshly emitted particles were likely made up of a fractal backbone of black carbon agglomerate. As particles aged, a covering of low-volatility secondary organic aerosol

formed on the backbone. The covering filled the void spaces in the agglomerate, which resulted in a higher effective density. The results are consistent with previous work on the photochemical exposure of both biomass burning aerosol and diesel exhaust (Nakao *et al* 2011, Giordano *et al* 2015) that showed particles become denser with photochemical aging. Like the volatility results, the rate of change of the bulk effective densities between the two fuels are similar for the first two hours of photochemical aging. After two hours of aging, the rates diverge with the polluted fuels attaining a higher effective density within the time of experiment. The divergence at two hours of aging may be due to surface phenomena on the aerosols themselves or may be due to mass limitations in the gas phase. As with the volatility results, further aerosol and gas-phase speciation is required to determine which process drives aerosol density changes. Like particle size distribution, small differences between the clean and polluted particle densities generated in the smoldering phase suggested that physical fire properties are important in the resulting bulk effective density of a combustion particle.

4. Summary and implications

Biomass burning is one of the largest contributors to the global carbonaceous emissions budget. Various biomass species and fire properties affect both the amount and physical properties of biomass burning emissions. Despite the limitations of repeated sampling and a lack of direct aerosol chemical speciation, the results of this pilot-scale study are conclusive: the regional air quality in the location of biomass growth also affects the burning emissions. Plants grown in areas of 'high' NO_x deposition are shown to have greater elemental nitrogen content. This result is consistent with previous studies that have shown measurable impacts on biomass grown in nitrogen saturated areas. Here we show that the increased nitrogen fuel content modified emission properties as compared to fuels grown in areas of lower NO_x deposition. In particular, significant differences in the per mass emission of gaseous nitrogen species, total particle number, and the volatility of the aerosol emitted are effected by the exposure of biomass fuels to NO_x.

Much more work is required to continue to test these findings. However, the results from this work show that there is a measurable and systematic compositional difference between aerosols generated by the burning of clean and polluted fuels. To address one shortcoming of this work, direct aerosol chemical speciation should be performed. Aerosol chemical speciation would allow for nitrogen mass balance and should be a main goal of future work. Understanding where nitrogen partitions in burning emissions may prove to be essential in predicting the global impacts of biomass

burning. Beyond measurement techniques, biomass from ecosystems beyond coniferous forests should also be examined. Ecologists have performed nitrogen saturation measurements in coniferous, deciduous, and wetland ecosystems so the burning of hardwoods, softwoods, and grasses should all be tested. Additionally, the tree species sampled here are all temperate species collected from a precipitation-limited ecosystem. As such, the results here should be applicable to similar ecosystems near large urban (polluted) areas, such as central Mexico's southwestern oak savannahs. However, the results may not extend to tropical, water-abundant ecosystems. Water-abundant, high nitrogen deposition areas may be especially relevant when modeling biomass burning in the Amazon and parts of Asia and Africa where biomass burning seasons are annual occurrences. A larger, extensively funded study, would be well suited to exploring these questions in more depth.

Currently, measured emission factors based on fuel species and types are used to assess global biomass burning emissions inventories. Emission factors used to predict the anthropogenic impact of biomass burning should also account for the local air quality in which the fuels are grown (e.g. in polluted or non-polluted areas). Furthermore, an additional feedback loop for biomass burning predictions should be considered in regards to anthropogenic impacts on climate. Since the rate of the photochemical evolution of both polluted and clean fuels were shown to be remarkably similar, current data on the evolution of modeled emissions need not be altered. The results of this study indicate that only a difference in initial emission factors for clean and polluted growing areas need to be considered in emissions inventories and models.

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