Biomass burning as a potential source for atmospheric ice nuclei: Western wildfires and prescribed burns

Anthony J. Prenni,¹ Paul J. DeMott,¹ Amy P. Sullivan,¹ Ryan C. Sullivan,¹,² Sonia M. Kreidenweis,¹ and David C. Rogers³

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1. Introduction

The sources, abundance and nature of atmospheric particles that serve as ice nuclei (IN) for cold cloud formation remain some of the most important, yet poorly-characterized, features of aerosol-cloud interactions that indirectly affect climate. Although a great deal of effort has focused on characterizing the ice nucleating ability of mineral dusts, less is known about carbonaceous particles. A primary source for carbonaceous IN is from biomass combustion. Here we report new measurements of IN activity at −30°C and above water saturation from biomass burning generated particles from prescribed burns and wildfires in the western US. These measurements suggest a range of IN activity, with variability largely dependent on the intensity of the fire. Although the fraction of particles generated during burns which serve as IN is quite small, the large numbers of particles generated in fires make biomass burning a potentially important source of IN to the atmosphere. Citation: Prenni, A. J., P. J. DeMott, A. P. Sullivan, R. C. Sullivan, S. M. Kreidenweis, and D. C. Rogers (2012), Biomass burning as a potential source for atmospheric ice nuclei: Western wildfires and prescribed burns, Geophys. Res. Lett., 39, L11805, doi:10.1029/2012GL051915.

2. Experiment

Measurements of IN number concentrations were made using the Colorado State University (CSU) continuous flow diffusion chamber (CFDC) [Petters et al., 2009]. Particles were processed above water saturation at −30°C. Under these conditions, the CFDC directly measures IN activating by condensation/immersion freezing. For these processing conditions, ambient background IN number concentrations are often ≤10 L⁻¹. We made measurements during five fires in Colorado and Wyoming, and in one case of smoke from long range transport. All measurements were conducted during daylight hours, except for the Medano wildfire, which extended to 10 PM local time. Details of each sampling period are given in Table 1.

Many of the measurements were conducted using the CSU mobile air quality laboratory (Brown Specialty Vehicles, Inc.). Measurement locations were selected based on forecasted winds, but were restricted based on road access and safety considerations. Power was supplied by generators (Honda EU6500is, EU3000i) located downwind of the parked vehicle. Number concentrations of condensation nuclei (CN) were measured using a condensation particle counter (CPC, TSI 3776/3010). In some cases, measurements were made of particle size distributions (TSI 3010 CPC and TSI 3071 DMA), PM2.5 mass concentrations (Met One Environmental Beta Attenuation Monitor; E-BAM), and bulk smoke chemical compositions (PM2.5 Hi-Volume...
quartz filter samples). Two punches from each filter sample were extracted in deionized water and analyzed for potassium and calcium ion concentrations using cation-exchange chromatography [Sullivan et al., 2008]. For aircraft measurements, the CFDC was mounted in the NSF/NCAR C-130 aircraft as part of the Ice in Clouds Experiment - Layer Clouds (ICE-L). During ICE-L, a TSI 3760 CPC was used to determine CN concentrations and an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS, DMT) was used to determine number concentrations for particles 0.5–1.0 μm. All number concentrations were corrected to standard temperature and pressure (STP).

3. Results
3.1. Prescribed Burns

[7] IN concentrations were measured for two days from the mobile laboratory during prescribed burns in Roosevelt National Forest. These burns were carried out by the US Forest Service near Sheep Creek, ~50 km northwest of Fort Collins, CO, with ~50 hectares burned each day. All sampling was done within 3 km of the fire, and at times the fire was within a few hundred meters of the sampling location. Aerosol potassium ion concentrations were indicative of the phase of the fire, with higher concentrations associated with flaming combustion [Echalar et al., 1995]. Potassium levels during the Sheep Creek burns suggested an active flaming phase (Table 1); ground based observations reported both flaming and smoldering concentrations. The sampling inlets were alternately in and out of the smoke plume, giving values within a factor of 4. IN concentrations were too high (>10^6 cm^-3) to measure directly with the TSI 3010 CPC; instead, number concentrations for particles 12–350 nm were derived from integrated size distributions. Although this method underestimates total CN, in all cases the mode diameter fell within this size range. Data from one sampling period are shown in Figure 1, where excursions in CN and PM2.5 concentrations indicate the presence of smoke. Measurements began ~40 min before biomass burning particles were first detected, as indicated by the initial spikes in CN at 14:15. A clear increase of IN concentrations inside of the plume is evident in Figure 1, and IN and CN concentrations were positively correlated (R^2 = 0.55 for two days of measurements). PM2.5 calcium ion concentrations, which can indicate the presence of lofted soil, did not show significant enhancement during the burns compared to pre-burn levels (3 days of measurements prior to the burn). Background IN concentration measured for the 40 min prior to the arrival of smoke was 3.4 L^-1, while the background CN concentration was 2300 cm^-3, making the fraction of all background particles which nucleated ice 1.5 × 10^-6, a typical value for background aerosol. Within the smoke, IN concentrations were elevated, such that if the plume were to reach cloud levels, these particles would likely influence freezing processes. The data in Figure 2 shows that IN concentrations increased with CN across the full range of CN concentrations, suggesting that the biomass burning produced particles themselves were responsible for ice nucleation. A simple linear fit of IN versus CN concentrations was used to

![Figure 1](image-url)

**Figure 1.** Data from Sheep Creek prescribed burns on July 23, 2009. IN (green) and CN (black) concentrations are corrected to STP. Also included are PM2.5 measurements (red), collected using an E-BAM. IN data are shown as 20 sec (0.5 L) averages. CN and PM2.5 data are shown based on the scanning times of the instruments, 3 min and 15 min, respectively.

Table 1. Summary of Fire Characteristics, Including Estimated Location of the Firea

<table>
<thead>
<tr>
<th>Name</th>
<th>Latitude/Longitude</th>
<th>Date</th>
<th>Sampling Time (min)</th>
<th>Major Fuels</th>
<th>Distance From Fire (km)</th>
<th>Potassium Ion (μg m^-3)</th>
<th>IN Fraction (IN/CCNavg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ICE-L RF01b</td>
<td>41°38'N 107°23'W</td>
<td>07 Nov 07</td>
<td>14</td>
<td>sage, aspen</td>
<td>11–23</td>
<td>NA^a 5 × 10^-6</td>
<td>2 × 10^-3</td>
</tr>
<tr>
<td>Sheep Creek</td>
<td>40°55'N 105°34'W</td>
<td>23 Jul 09–24 Jul 09</td>
<td>335</td>
<td>grasses, sage, bitterbrush, Douglas fir, Ponderosa pine, Lodgepole pine, aspen chaparral, mixed pines, sage brush, mahogany, scrub oak, Douglas-fir ponderosa pine, douglas fir, grasses</td>
<td>&lt;3 0.29 0.17</td>
<td>1 × 10^-6</td>
<td></td>
</tr>
<tr>
<td>Aged Smoke Ft Collins</td>
<td>34°15'N 118°12'W</td>
<td>01 Sept 09</td>
<td>156</td>
<td>chaparral, mixed pines, sage brush, mahogany, scrub oak, Douglas-fir ponderosa pine</td>
<td>1–3 0.05 4 × 10^-6</td>
<td>8 × 10^-6</td>
<td></td>
</tr>
<tr>
<td>Medano</td>
<td>37°44'N 105°31'W</td>
<td>25 Jun 10</td>
<td>82</td>
<td>80</td>
<td>0.16d 8 × 10^-6</td>
<td>8 × 10^-6</td>
<td></td>
</tr>
<tr>
<td>Four Mile</td>
<td>40°03'N 105°23'W</td>
<td>07 Sept 10</td>
<td>102</td>
<td>ponderosa pine, douglas fir</td>
<td>10 NA BDL</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ft Collins</td>
<td>40°03'N 105°23'W</td>
<td>09 Sept 10</td>
<td>226</td>
<td>6 0.06</td>
<td>BDL</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Four Mile Boulder</td>
<td>40°3'N 105°23'W</td>
<td>13 Sept 10</td>
<td>115</td>
<td>ponderosa pine, douglas fir</td>
<td>BDL</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reservoir Road</td>
<td>40°23'N 105°18'W</td>
<td>13 Sept 10</td>
<td>115</td>
<td>grasses, juniper, pine</td>
<td>BDL</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

^aSampling time designates the total sampling time, including measurements outside of the plume.

^bICE-L IN fraction are for –33°C.

^cNA designates that no potassium data were collected.

^dPotassium data were not co-located with the IN measurements, but rather were collected at Rocky Mountain National Park, which was also impacted by the fire.

^eCalculated from measurements with poor correlation between IN and CCN. In all cases, IN fractions derived from INavg/CCNavg in smoke gave values within a factor of 4.

^fBelow Detection Limit; no measurable increase in IN was observed.
determine the fraction of biomass burning particles that served as IN during our measurement period. The slope from this fit indicates an ice nucleation fraction of $2 \times 10^{-7}$, a lower fraction than was observed for the background air. This ice nucleation fraction was lower than that estimated to be needed for a large impact on the regional IN budget [Petters et al., 2009].

The Four Mile Canyon fire, ~8 km northwest of Boulder, CO, was a human-caused wildfire that spread to steep, rocky terrain. On September 7, 2010, this fire was very active, with substantial contributions from flaming combustion (http://www.inciweb.org/ and Table 1). On September 7, smoke reached Fort Collins, CO, ~80 km from the fire, with PM$_{2.5}$ in Fort Collins (http://www.colorado.gov/airquality/report/aspx) increasing from 1 μg m$^{-3}$ at 8 AM to 35 μg m$^{-3}$ by 1 PM (all times reported as local time). During the IN measurements, which started at 5 PM in our laboratory in Fort Collins, PM$_{2.5}$ had dropped to 16 μg m$^{-3}$, but with the sight and smell of smoke still present. For comparison, average PM$_{2.5}$ concentration on September 6 was 4 μg m$^{-3}$, and one hour values never exceeded 11 μg m$^{-3}$. The smoke near the surface sampling site appeared well-mixed, with a relatively narrow range of elevated CN concentrations observed (8300–10,600 cm$^{-3}$). On this day, IN and CN were not well correlated ($R^2 = 0.15$). This poor correlation may have resulted from the narrow range of CN observed, due in part to the fact that no pre-burn measurements were obtained. Average IN concentrations were $>50$ L$^{-1}$ at $-30^\circ$C, and the IN fraction derived from the slope of IN versus CN was $8 \times 10^{-6}$. With poor correlation between IN and CN, there is significant uncertainty in using the slope between IN and CN to determine IN fraction; determining this fraction by dividing average IN by average CN for the entire measurement period gave a comparable value ($5 \times 10^{-6}$). Although the IN fraction was relatively low, in this case the smoke had traveled 80 km to our sampling site, so that the large observed IN concentration itself implied a significant regional impact on the IN population.

**Figure 2.** IN concentration (L$^{-1}$) at $-30^\circ$C in the immersion/condensation regime versus CN concentration (cm$^{-3}$) from the Sheep Creek prescribed burns (circles). CN data are for 12–350 nm. Data are either 3 min averages or 5 min averages. Higher CN concentrations correspond to higher concentrations of biomass burning particles. Also shown are average background and smoke-affected concentrations for ICE-L RF01 (red square), Medano (blue square), Four Mile (Boulder measurements, green square), and Reservoir Road (magenta square) fires. The ICE-L RF01 are for $-33^\circ$C measurements.

**Figure 3.** Data from the ICE-L field study, RF01. All results are corrected to STP. IN data (green) were collected at $-33^\circ$C in the immersion/condensation regime. Also shown are CN (black) and aerosol concentration 0.5–1.0 μm (dashed) from the UHSAS.
[10] On September 9, 2010, we used the mobile laboratory to make measurements in Boulder, CO, closer to the Four Mile Canyon fire. By this time the intensity of the fire had decreased significantly, with ground-based observations describing the fire as primarily creeping and smoldering. Average PM$_{2.5}$ concentrations (http://www.colorado.gov/airquality/report.aspx) were ~6 μg m$^{-3}$ in Boulder. For these conditions, IN concentrations were at typical background levels ($IN_{avg} = 9 L^{-1}$), and we found no correlation ($R^2 < 0.01$) between IN and CN, even for a broad range of CN concentrations (8300–76,000 cm$^{-3}$). Thus, for the same fire, but different burn conditions, we observed very different impacts on the IN population. These observations are consistent with the assertion that flaming combustion is likely necessary for strong IN production in fires [Petters et al., 2009], who speculated that a large fraction of organic carbon, produced during the smoldering fire phase, may suppress IN activity.

[11] We sampled smoldering combustion emissions from two more wildfires. The Medano wildfire in the Sangre de Cristo Mountains began with a lightning strike on June 6, 2010 near Great Sand Dunes National Park. Our measurements were conducted within Sand Dunes NP on June 25, when smoldering combustion dominated, as determined from visual observations and PM$_{2.5}$ potassium ion concentrations (Table 1). At the time of the measurements, the total area burned had reached ~2000 hectares. Although our sampling inlets were located at a lower elevation than the fire, measurements were possible due to an inversion, trapping smoke at lower levels. We were alternately in and out of the smoke plume throughout the measurement period, with large variability in CN (3100–128,000 cm$^{-3}$). Despite the high concentrations of particles, average IN concentrations were 13 L$^{-1}$, and IN and CN were not correlated ($R^2 < 0.01$), suggesting little impact on IN concentrations when smoldering combustion dominated. A similar result was found during the Reservoir Road wildfire, ~7 km west of Loveland, CO. This fire ultimately covered ~360 hectares; it was nearly this size at the time of the measurements. Although the fire was intense in its early stages, during the measurements its intensity had waned considerably, with primarily smoldering combustion (http://www.inciweb.org and Table 1). Average IN concentrations were elevated at 27 L$^{-1}$; however, no correlation was observed ($R^2 < 0.01$) between IN and CN, even for a broad range of CN concentrations (5100–43,000 cm$^{-3}$).

3.3. Long Range Transport

[12] Levin et al. [2009] used local particle measurements and the Navy Aerosol Analysis and Prediction System Global Aerosol Model to show that long range transport of smoke from the Station Fire in southern California impacted northern Colorado on September 1, 2009. This smoke traveled ~1600 km from reaching our laboratory, and was the result of intense flaming combustion (Table 1). On the morning of September 1, PM$_{2.5}$ levels reached over 60 μg m$^{-3}$ in Fort Collins (http://www.colorado.gov/airquality/report.aspx). Measurements were made later in the day, when PM$_{2.5}$ dropped to ~33 μg m$^{-3}$, but with the smell of smoke still present. The smoke was well-mixed, with CN concentrations of 3000–4200 cm$^{-3}$, and the correlation between CN and IN was again low ($R^2 = 0.05$). Average IN concentrations were elevated, ~27 L$^{-1}$, and the IN fraction was 1 × 10$^{-5}$, the highest that we observed for all of the fires. These data suggest a smoke impact on IN concentrations that extended across several states.

4. Conclusions

[13] Biomass burning particles have been shown to influence ice nucleation, although this impact may vary widely [Mossop, 1963; Petters et al., 2009]. In this study, we aimed to add to previous observations by determining IN concentrations during near-field sampling from 5 prescribed burns and wildfires, as well as from smoke arriving at our sampling site after long range transport. Our observations suggest two distinct regimes of IN production during biomass burning events. Smoke emitted during an active flaming phase showed positive correlation between IN (~30°C in the immersion/condensation-freezing regime) and total particle number concentration. Although the fraction of particles which nucleated ice was estimated to be too low to have large regional impacts [Petters et al., 2009], and was lower than that of background air, in all cases IN number concentrations were elevated by at least a factor of 2–3 over those typical of background air. In contrast, fires dominated by smoldering combustion showed no correlation between IN and CN number concentrations, and IN concentrations were not consistently elevated in the presence of smoke. As such, smoldering fires did not appear to have an impact on local IN populations. These measurements are, of course, limited in terms of the variety of fuel sources observed, but combined with laboratory work [Petters et al., 2009] paint a consistent picture regarding the importance of flaming combustion in generating ice nucleating particles. While the IN-active fractions of flaming-phase biomass smoke particles were observed to be low, the large total particle number concentrations produced by these fires, even when sampled long distances from the fire source, suggest that wildfires can produce wide-spread increases in atmospheric IN concentrations.

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