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Biomass Burning Observation Project (BBOP) Final Campaign Report

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Executive Summary

The Biomass Burning Observation Project (BBOP) was conducted to obtain a better understanding of how aerosols generated from biomass fires affect the atmosphere and climate. It is estimated that 40% of carbonaceous aerosol produced originates from biomass burning—enough to affect regional and global climate. Several biomass-burning studies have focused on tropical climates; however, few campaigns have been conducted within the United States, where millions of acres are burned each year, trending to higher values and greater climate impacts because of droughts in the West. Using the Atmospheric Radiation Measurement (ARM) Aerial Facility (AAF), the BBOP deployed the Gulfstream-1 (G-1) aircraft over smoke plumes from active wildfire and agricultural burns to help identify the impact of these events and how impacts evolve with time. BBOP was one of very few studies that targeted the near-field time evolution of aerosols and aimed to obtain a process-level understanding of the large changes that occur within a few hours of atmospheric processing.

The G-1 research aircraft was stationed in two locations. From July to September 2013, the G-1 was deployed in Pasco, Washington, and took measurements during 17 naturally occurring uncontrolled fires across Washington, Oregon, Idaho, northern California, and western Montana. This location allowed the G-1 to gather data from smoke plumes under 5 hours of age. In October 2013, the G-1 was deployed to Memphis, Tennessee, where over 24 agricultural burns in the lower Mississippi River Valley were sampled. Measurements were taken from varied distances and ages to establish a clear picture of aerosol evolution. Aged smoke that covered large regions and plumes from five urban areas were sampled for contrast. Three flights included segments over and near the Mount Bachelor Observatory (MBO). One MBO flight examined a complex of forest fires in northern California, which were observed at MBO and sampled by the National Aeronautics and Space Administration aircraft¹ in collaboration with the G-1.

The G-1 was equipped with a suite of instruments for measuring aerosol, trace gas, and atmospheric parameters. Measurements included aerosol concentration, size distribution, optical properties, and radiative flux. The BBOP campaign also represented the maiden deployments of the 355-nm photoacoustic spectrometer, the 532-nm photothermal interferometer, the extended wavelength fast-integrating mobility spectrometer, and soot photometer–aerosol mass spectrometer, enabling measurements not previously available in airborne research.

We found that all fire plumes were dominated by organic aerosols, with soot accounting for only a few percent of the mass. After atmospheric aging times of 2 hours, aerosol concentration normalized for dilution had increased modestly, while light scattering typically had doubled, indicating that changes in particle size have major impacts on scattering and hence radiation. Single scattering albedos near a fire were usually consistent with a small net cooling effect that became significantly larger a few hours downwind. Aerosol absorption measurements (i.e., Angstrom exponents) indicated the presence of near-ultraviolet light absorbing brown carbon. Transmission electron microscopy showed that aged aerosol particles found in regional smoke haze often are dominated by high-viscosity organic aerosols (i.e., “tar balls”) while within the active plume, the particles were primarily low-viscosity organic aerosols.

¹ The National Aeronautics and Space Administration-sponsored Emissions and Atmospheric Composition, Clouds, and Climate Coupling by Regional Surveys dedicated research flights to study the California wildfire.

Aerosol Mass Spectrometer measurements indicated aerosol nitrate increases of up to an order of magnitude after a few hours of aging, while sulfate levels remained almost constant. There is a monotonic increase in aerosol O-to-C ratio, which is consistent with secondary organic aerosol formation accompanying primary organic aerosol loss as the net increase in organic aerosol is relatively minor. Finally, active gas-phase chemistry is implied by the very rapid loss of NO_x , which apparently does not proceed by $\text{OH} + \text{NO}_2$ as the change in the toluene-to-benzene ratio is minor. Ozone production can yield concentrations in excess of 100 ppb in some plumes and only minor increases in others.

Flight patterns for fires occurring in the Pacific Northwest included multiple transects at varying downwind distances with multiple repetitions of these transects. This flight pattern assisted in assessing the role of natural variability in comparison to downwind trends attributed to aging of emitted and secondary pollutants. Where possible, a centerline transect flown through the plume provided data on aerosol evolution without the inherent steps in plume age characteristic of aircraft-based Lagrangian sampling strategies (e.g., 30-minute, 1-hour step sizes). Because agricultural burns generally are smaller in size, the primary comparison is between fresh pollutants and regional haze caused by multiple burns.

Acronyms and Abbreviations

AAF	ARM Aerial Facility
AMS	Aerosol Mass Spectrometer
ARM	Atmospheric Radiation Measurement
ASR	Atmospheric System Research
BBOP	Biomass Burn Observation Project
BC	black carbon
BNL	Brookhaven National Laboratory
BrC	brown carbon
CAPS	Cloud, Aerosol, and Precipitation Spectrometer
CAPS PMex	Cavity Attenuated Phase Shift Particulate Matter Extinction
CCN	cloud condensation nuclei
CPC	Condensation Particle Counter
DOE	U.S. Department of Energy
FIMS	Fast-integrating Mobility Spectrometer
G-1	Gulfstream-1
MBO	Mount Bachelor Observatory
PAS	Photoacoustic Spectrometer
PbP	Particle-by-Particle
PCASP	Passive Cavity Aerosol Spectrometer Probe
PI	principal investigator
PNNL	Pacific Northwest National Laboratory
POA	primary organic aerosol
PTI	Photothermal Interferometer
SOA	secondary organic aerosol
SP2	Single-particle Soot Photometer
SP-AMS	Soot Photometer – Aerosol Mass Spectrometer
TEM	transmission electron microscopy
UHSAS	Ultra-high-sensitivity Aerosol Spectrometer
UTC	Coordinated Universal Time

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1.0 Background

The Biomass Burning Observation Project (BBOP) was undertaken to advance our understanding of how aerosols generated from burned biomass affect the atmosphere and climate. Using the resources of the Atmospheric Radiation Measurement (ARM) Aerial Facility (AAF), BBOP deployed the Gulfstream-1 (G-1) aircraft over active wildfire and agricultural smoke plumes to help identify the impact of agricultural burning and wildfires and how these particles evolve over time. Aircraft sampling focused primarily on determining the time evolution of fire-generated pollutants over a time period from nearly just-emitted to 4 hours of atmospheric aging. Air with a regional accumulation of fire-generated pollutants provided information on aging over longer time scales. Two intensive study periods were undertaken in 2013: 1) June 15 to September 13, during which the G-1 operated out of its home base in Pasco, Washington, with the primary objective of observing wildland shrub and forest fires in the Pacific Northwest; and 2) October 1-22, during which the G-1 was stationed in Memphis, Tennessee, and was used to observe agricultural burns in the lower Mississippi River Valley. During the first intensive period, surface observations were made at Mount Bachelor Observatory (MBO), a mountaintop facility in central Oregon that is frequently impacted by wildland burns. In addition to targeting wildfires and agricultural burns, BBOP flights were conducted in and around the five urban areas of Spokane, Washington; Seattle, Washington; Portland, Oregon; Memphis, Tennessee; and Nashville, Tennessee. Air samples around power plants near Nashville also were collected. These urban and power-plant flights served to determine the contrasting behaviors of carbonaceous pollutants emitted from different sources. Figure 1 shows the locations sampled and ground tracks of the G-1. During BBOP, a total of 35 flights were flown, three of which included sampling over and upwind of MBO and one that was flown in coordination with the National Aeronautics and Space Administration Studies of Emissions, Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys program.

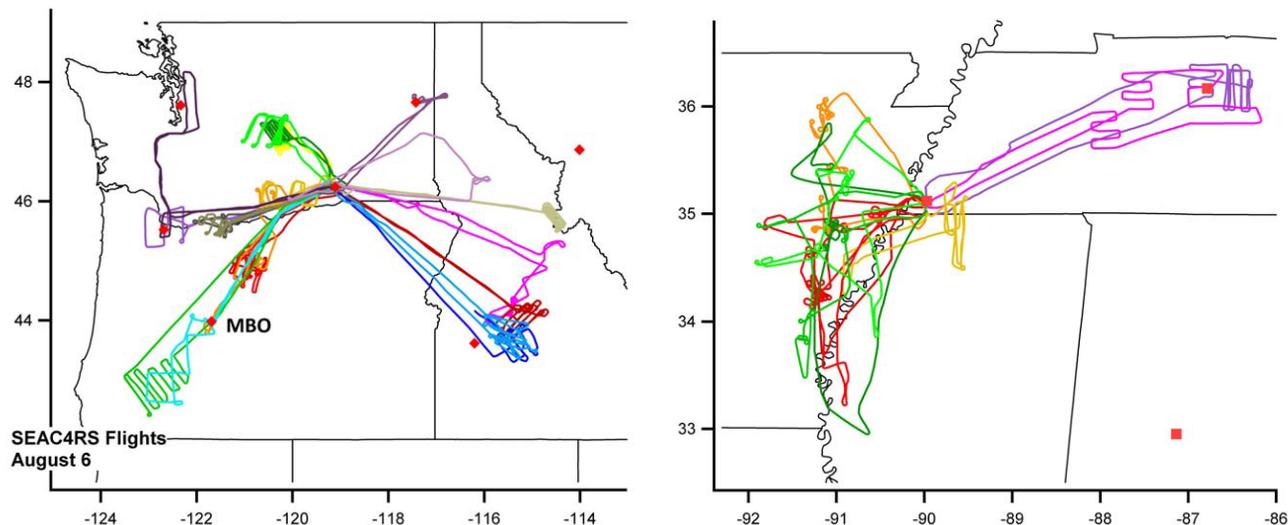


Figure 1. G-1 BBOP Flight Tracks. Left: Pacific Northwest. Right: Lower Mississippi River Valley and Nashville.

Table 1 lists the instruments provided for use in the G-1 by the ARM Facility and outside investigators. BBOP was the first aerial deployment of the Aerodyne Research, Inc., soot photometer–aerosol mass spectrometer (SP-AMS), the University of Nevada photoacoustic spectrometer (PAS), and Brookhaven National Laboratory’s (BNL) photothermal interferometer (PTI) and fast-integrating mobility spectrometer (FIMS). Instruments provided by the Jaffe group and by the Zhang group at MBO are listed in Table 2.

Table 1. G-1 Instrument Suite

Instrument	Measurement	Instrument mentor
3-λ Particle Soot Absorption Photometer	Aerosol light absorption. @ 467, 530, and 660 nm.	D. Chand, Pacific Northwest National Laboratory (PNNL)
3-λ Nephelometer	Aerosol light scattering, 450 nm, 550 nm and 700 nm	D. Chand, PNNL
Cloud Condensation Nuclei (CCN) counter (dual-column)	CCN concentration @ 2 SS (0.25% and 0.50%)	F. Mei, PNNL
PTR-MS	Trace volatile organic compound detection	J. Shilling, PNNL
Trace gas suite	NO, NO ₂ , NO _y , CO, SO ₂ , O ₃	S. Springston, BNL G. Senum, BNL
Meteorology	Wind direction; wind speed; air temperature; RH and rain fall	J. Hubbe, PNNL
Single-particle Soot Photometer (SP2)	Black carbon (BC) loading, size distribution, mixing state	A. Sedlacek, BNL
Ultra-high-sensitivity Aerosol Spectrometer (UHSAS)	Particle size distribution 60 – 1000 nm	J. Thomlinson, PNNL
Passive Cavity Aerosol Spectrometer probe (PCASP)	Particle size distribution 0.13 – 3 μm	J. Thomlinson, PNNL
Cloud, Aerosol, and Precipitation Spectrometer (CAPS)	Particle size distribution 0.5 – 50 μm	J. Thomlinson, PNNL
TSI-3010 Condensation Particle Counter	Particle counter; 10 nm – 1 μm	F. Mei, PNNL
TSI-3025 Condensation Particle Counter	Particle counter; 3 nm – 1 μm	F. Mei, PNNL
Cavity Attenuated Phase Shift Particulate Matter Extinction (CAPS PMex)	Particle extinction (Aerodyne)	A. Friedman, Aerodyne
PTI	Light absorption at 532 nm (BNL)	A. Sedlacek, BNL
FIMS	Particle size distribution (BNL)	J. Wang, BNL
Soot Photometer-Aerosol Mass Spectrometer SP-AMS	Particle and coating composition (Aerodyne)	T. Onasch, Aerodyne E. Fortner, Aerodyne
Electron microscopy	Two-dimensional/three-dimensional particle morphology and mixing state (Arizona State University)	P. Buseck, Arizona State University K. Adachi, Meteorological Research Institute
PAS	Aerosol light absorption @ 405 nm, 870 nm (University of Nevada)	P. Arnott, University of Nevada

Table 2. Mount Bachelor Observatory Instrument Suite

Instrument	Measurement
Model 3563, TSI, Inc., St. Paul, Minnesota	Aerosol scattering coefficient @ 450, 550, 700 nm.
Particle Soot Absorption Photometer, Radiance Research, Seattle, Washington	Aerosol absorption coefficient @ 467, 530, 660 nm
Picarro G2502 Cavity Ring-down Spectrometer	Carbon dioxide
Picarro G2502 Cavity Ring-down Spectrometer	Carbon monoxide
Dasibi 1008-RS	O ₃
Tekran 2537A	Mercury (elemental)
Odd N suite	NO _y , NO, NO ₂ , PAN
Sunset Laboratories Semi-continuous Organic Carbon/Elemental Carbon field analyzer	Organic carbon/elemental carbon
NOAA flasks	~50 Volatile organic compounds and halogenated compounds
Eppley TUVB Radiometer	Ultraviolet radiance
Campbell Scientific HMP 45C	Temperature and relative humidity
Campbell Scientific HMP 45C	Water vapor
Vaisala PTB101B PRESSURE TRANSMITTER	Atmospheric pressure
Aerodyne HR-tof-AMS	Particle nonrefractory composition and size
Measurements by the Jaffe group, University of Washington, and Zhang group, University of California, Davis	

1.1 Additional Team Members

Instrument mentors within the ARM Facility and scientists from outside agencies that brought instruments to the G-1 or MBO are listed in Table 1 and Table 2. Other team members included Beat Schmid and Jennifer Comstock of PNNL and Peter Daum and Ernie Lewis of BNL. Special acknowledgement is called out to the AAF pilots, led by Mike Hubble, who safely and skillfully implemented the scientific goal of sampling fires near their source and at varying downwind distances. The AAF pilots met the BBOP sampling goals while working in an environment with severely reduced visibility and with the presence of fixed and rotary wing platforms used to combat the fires.

2.0 Notable Events or Highlights

Each wildfire was a memorable event. Much of our best data was collected over fires that individually burned several tens of thousands of acres. In 2013, the Rim Fire (named for its proximity to the Rim of the World vista point, a scenic overlook on Highway 120) near Yosemite, California, spread over 250,000 acres. The Rim Fire was beyond the range of the G-1, so in a collaborative effort, a National Aeronautics and Space Administration aircraft collected samples for that event.

Disregarding systemic problems, the number of instrument malfunctions experienced during the BBOP was in line with past expectations. Bad or questionable data are documented in the ARM Data Archive.

The Ultra-high-sensitivity Aerosol Spectrometer (UHSAS) flow controller had a leak that was not discovered until near the end of the campaign. When the leak was discovered, a flow calibration yielded new flows to be used in place of incorrect values. This still left us with several problems: the need to assume that the flow error did not vary since the start of the campaign and that the flow error is pressure (i.e., altitude) independent. The flow error tended toward providing a greater flow than indicated, causing

the UHSAS to experience coincidence problems at a factor of 4 lower concentration than would otherwise occur.

Based on the only published description of the SP-AMS prior to the BBOP (Onasch et al. 2012), we expected that the SP-AMS could be used to measure *only* the refractory and associated nonrefractory components of particles that contained black carbon (BC). However, this could not be done without physically removing the vaporization oven. Because of the instrument layout, the laser beam used for detecting BC unintentionally heats the oven. Even if the oven is off, it remains hot enough to vaporize most nonrefractory particles. Potential problems with running the SP-AMS in a mode requiring rapid (i.e., 1 minute) switching of the laser on and off were avoided soon after the project began when it was recognized that 1-minute switching was incompatible with rapid changes in plume composition.

The calibration unit for the trace gas suite is marginal at best for aircraft use and needs to be upgraded. The CO instrument was changed prior to BBOP. Its reliability, sensitivity, and accuracy were much improved over previous units; however, the response time was slower than that of the previous instrument. A response time improvement of more than a factor of 10 could be obtained with a larger (and heavier, hotter, and higher-powered) pump. The mentor is aware of these ongoing concerns. Instrument issues also are discussed in Section 3 Lessons Learned.

3.0 Lessons Learned

We expected that fire plumes would contain very high number concentrations of aerosol particles. Instruments deployed on the G-1 to measure number concentration all have upper limits beyond which individual particles cannot be distinguished. At a high aerosol number concentration, multiple particles are recorded as coincident, which leads to under counting and distortion of the size spectra. The affected instruments on the G-1 are the under-wing particle probes (UHSAS, PCASP, and CAPS) and the in-cabin condensation particle counters (CPC), CCN counters, SP2, and FIMS. To minimize coincidence problems, the AAF constructed a dilution system for the in-cabin instruments, but unfortunately not for the CCNs. Flow to the UHSAS could be adjusted, yielding the same effect as dilution. Because the PCASP and CAPS are only sensitive to larger particles, coincidence was less of a problem. Some fraction of our particle data is lost. The principal investigators (PI) are working to 1) quantify the coincidence problem, 2) determine whether additional processing would be useful for the SP2 (fitting multiple peaks) or UHSAS (using Particle-by-Particle [PbP] data), and 3) construct surrogate measures of particle concentration by folding in scattering data at multiple wavelengths.

Instruments sensitive to particle coincidence should be set up with a margin of error to accommodate unforeseen high concentrations. It is preferable to run instruments continuously at maximum dilution (or minimum flow for the UHSAS) and sacrifice signal-to-noise in clean regions rather than vary instrument settings. The risk that a clean air setting will be used in-plume outweighs the gain of making a better measurement in clean air as the latter was not a BBOP primary target.

Very rapid changes in concentration were observed, especially on transects close to wildland fire sources. Increased attention needs to focus on instrument response time and coordination of time bases between instruments. A suggested minimum accuracy for time measurements is 1 second. A discrepancy between instruments of 2 seconds is readily apparent as a loss of correlation. For some instruments, time responses

faster than 1 Hz are desirable, as shown by an analysis of particle clustering reported by Senum et al. (2015). For a certain broad set of analytical methods, the PIs believe that the casual data user could draw incorrect conclusions unless the varying time delays and time responses of different instruments are taken into account. The PIs are in the process of creating consistent merged data sets. For this campaign and for others, a decision should be made regarding how this responsibility should be split between AAF, instrument mentors, and campaign PIs. In view of the unique field campaigns that ARM supports, it would be beneficial to have a pre-campaign meeting of interested parties to discuss goals, products, and unique aspects of the deployment, and to clearly delineate responsibilities for achieving the goals and developing the products.

The PIs believe that the methods used to identify “bad” data and the way that bad data are treated in the data archive need to be revised. According to the International Consortium for Atmospheric Research on Transport and Transformation format followed by the ARM Facility, an invalid measurement is replaced with an entry of -9999. Extra columns are used in a data file to identify the reason a data entry is suspicious or invalid. Several classes of problems described below occurred in the assessment of data quality.

- Measurements of aerosol concentration in fire plumes were out of the range of that usually encountered. Even though fires are mainly a source of small particles, a fraction of particles that were larger than a few microns in diameter were present. Mentor quality-control/quality-assurance algorithms attributed time periods with high aerosol concentration to cloud droplets though clouds were demonstrably absent, which resulted in the replacement of valid data with a value of -9999.
- Comparisons between instruments were used to identify “out-of-bounds” results. The problem with this approach is that one does not always know which instrument is correct. For example, the UHSAS should always record a lower particle number concentration than the CPCs because the CPCs count particles over a larger size range. This comparison tended to fail in clean air, where fewer small particles and normal instrument precision could cause the UHSAS number concentration to exceed that measured by the CPCs by a margin large enough to be flagged as invalid data. The end result was that fields for valid data were replaced with -9999 entries.
- Useful information on instrument performance was often missing or obscure. In one case, instrument voltages identified error conditions.
- The most significant data-quality problem faced was that of coincidence counting by particle probes. A few instrument mentors added data-quality flags warning of high concentrations. Yet, in no case was information provided from which one could determine the counting error from Poisson statistics. There have been several versions of AAF data generated, prompted by the PIs observation that, in the first-look data generated soon after a flight, the data later deemed to be invalid often could not be distinguished from data later deemed to be valid. This comparison was only possible because the first-look data did not have any data removed. Most, but not all of the removed data, has been returned.

Two recommendations for improvements stand out:

- Instrument mentors must have approximated calibrations at all points in the campaign so that the consistency of measurements from multiple instruments can be assessed within a day or two of a flight. The number of required calibration points would vary by instrument. Availability of timely

calibration data would allow problems to be identified early and fixed during the campaign. For example, while large discrepancies between the UHSAS and nephelometer were identified in the first week of the campaign, they were incorrectly ascribed to UHSAS data being processed with old calibration factors.

- Data entries of -9999 should be reserved for situations in which an instrument clearly is not working or is not turned on. We recognize that the average user has no interest in instrument readings during instrument zeroes and calibration periods. A mechanism should be found to make that data available so an end-user could determine the quality if needed.

4.0 Results

4.1 Brown Carbon Light Absorption

One of the unique aspects of BBOP was the deployment of a 355-nm PAS and a 532-nm PTI to measure aerosol absorption. These two instruments provided in situ measurements of aerosol absorption and a wavelength combination that provided the opportunity to probe the contribution of brown carbon (BrC) at 355 nm. An example of this value is seen in the analysis of data collected on the Government Flats Fire Complex that was targeted on August 21, 2013.

Using CO as a conserved tracer to correct for dilution, the scattering-to-CO ratio was found to be constant near the source region, and then to increase with plume age (left plot of Figure 2). A similar analysis of BrC absorption-to-CO ratio at 355 nm, in which the BC contribution at this wavelength has been removed using the PTI absorption measured at 532 nm, reveals analogous behavior; that is, near constant ratio at the source followed by an increase downwind (right plot of Figure 2). To explain this observation, a two-component model of BrC absorption is proposed in which contributions are partitioned between primary organic aerosols (POA) created at the source and secondary organic aerosol (SOA) formation downwind. Support for a two-component model of BrC absorption also is found in a laboratory study of light absorption by organic carbon from wood combustion conducted by Chen and Bond (2010). Field data obtained during the BBOP provides first-time evidence of a two-component system of BrC.

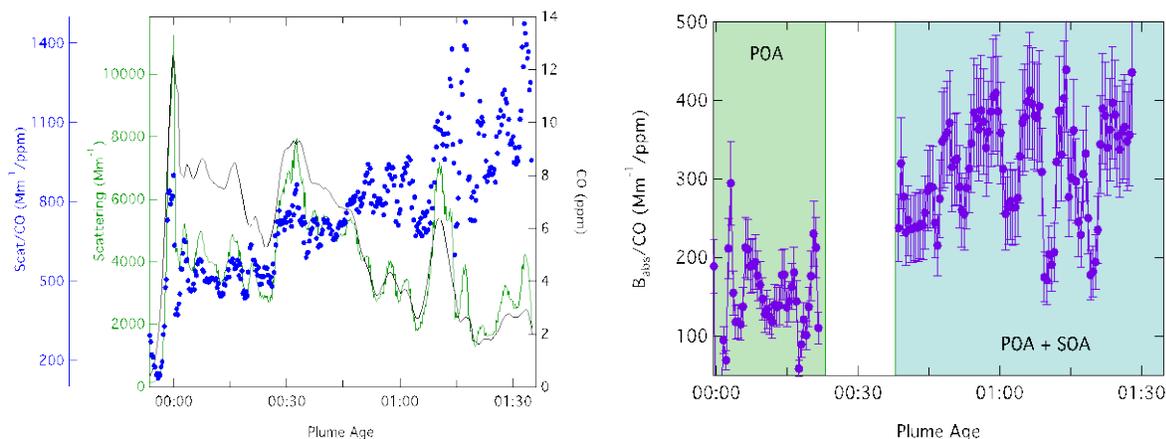


Figure 2. Evolution of BBOP for the Government Flats Complex Fire. The left plot shows aerosol scattering (green), CO (black), and scattering-to-CO ratios (blue) as a function of plume age, where CO is used to correct the scattering signal for plume dilution. The scattering-to-CO ratio is found to be constant during the first ~20 minutes, but then exhibits a steady increase. This increase suggests SOA production. A similar analysis of BrC absorption at 355 nm (right plot) reveals analogous behavior; that is, a near constant BrC absorption-to-CO ratio at the source followed by an increase in this ratio downwind. The data gap centered around 30 minutes is due to an instrument zero.

The apportionment of organic aerosol between POA and SOA is being investigated via positive matrix factorization analysis. Figure 3 shows fivefold increase in nitrate that tracks changes in scattering and absorption. The nitrate is likely in the form of organonitrates/nitroaromatics: a class of organics that are known to be produced during biomass burning events (Lin et al. 2015; Claeys et al. 2012; Iinuma et al. 2010). Nitrophenols have molecular absorption cross-sections in reasonable agreement with the enhanced absorption observed downwind (Chen et al. 2011).

Changes in the aerosol O-to-C ratio suggesting SOA production have been found for other flights (see Figure 4) and will be examined for the entire data set including the Government Flats Complex Fire.

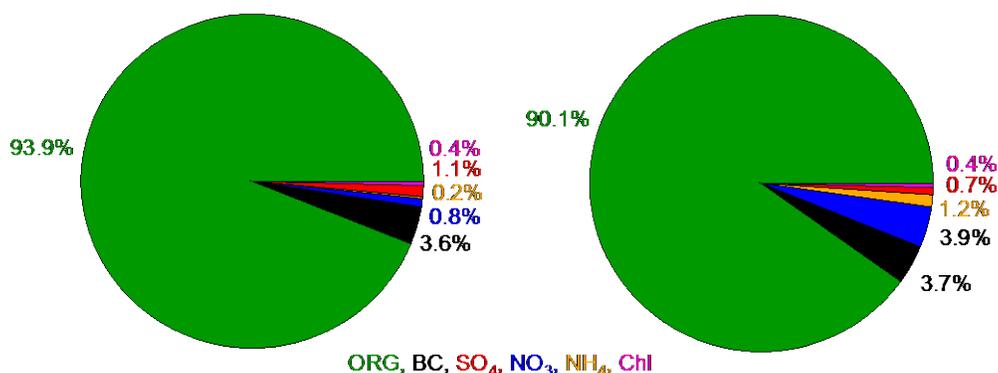


Figure 3. Aerosol Composition for the Government Flats Complex Fire. Aerosol compositions near the plume source (left pie chart) and downwind (right pie chart) from SP-AMS data. Downwind represents nominally 90 minutes of aging. Color mapping: organics – green; refractory black carbon – black; nitrates – blue; ammonium – yellow; sulfate – red; chloride – magenta.

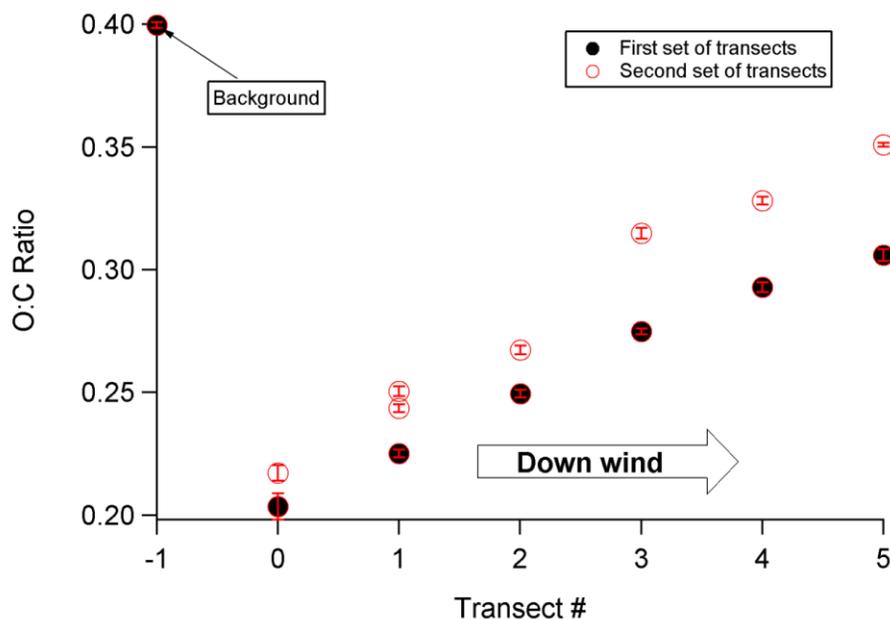


Figure 4. Aerosol O-to-C Ratio. Increase in aerosol O-to-C ratio as a function of downwind distance from the Colockum Tarps Fire (sampled on July 30, 2013).

4.2 Chemical Signature of Aerosol Aging

As shown in Figure 4, Onasch et al. (2015) found that the O-to-C ratio of organic aerosol in the Colockum Tarps Fire increased with downwind distance. This can be interpreted as a combination of POA evaporation and SOA formation.

Very rapid (up to an order of magnitude in 2 to 4 hours) disappearance of NO_x in wildfire plumes, about half of which involves conversion to gas or aerosol-phase odd N not detected as NO_y . The toluene-to-benzene ratio also decreased but by as much as an order of magnitude less.

In several fire plumes (not all have analyzed), aerosol sulfate appears to be a primary pollutant, while aerosol nitrate is formed downwind. Ammonium concentrations are stoichiometrically equivalent to nitrate, even though sulfate also is present as shown in Figure 5.

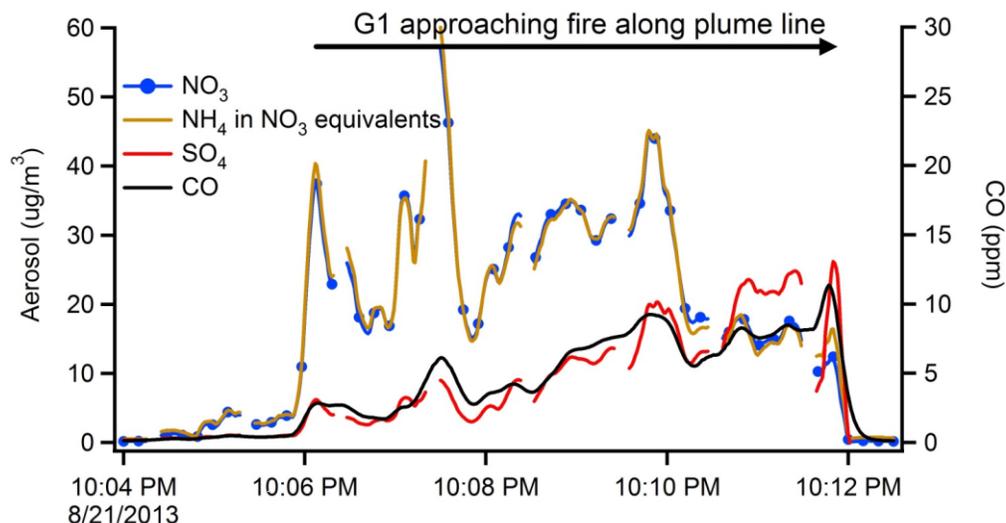


Figure 5. Trace Gas Evolution for the Government Flats Complex Fire. Concentrations of CO and aerosol constituents from the Government Flats Complex Fire, measured during an along-plume flight segment. Time is UTC, coordinated universal time. Transport time of the plume is approximately 2 hours. Ammonium concentration has been multiplied by a factor of $^{62}/_{18}$ to yield nitrate equivalents. There is an almost exact equality between nitrate and ammonium, requiring that symbols be added to the nitrate trace for visibility. Sulfate is seen to be proportional to CO indicating that it is a primary aerosol constituent. In contrast, the ratio of nitrate to CO increased by more than a factor of 5 during plume transport.

4.3 Presence of Tar Balls and Their Detection by an AMS

A sequence of transmission electron microscopy (TEM) images taken at increasing distances from a wildland fire indicates that tar balls are a prominent part of the aerosol mixture, and their number concentration increases with atmospheric aging (Figure 6). Within the electron microscopy community, substances, such as tar balls, that can withstand an electron beam for several minutes are referred to as refractory. This term is used in a different context by the AMS community to denote an aerosol that is not evaporated by a 600°C tungsten oven within a sampling cycle (approximately 1 second). In a preliminary experiment, Kouji Adachi subjected a BBOP wildfire TEM sample to step increases in temperature, taking 15 minutes to reach 600 °C. Figure 6 illustrates the gradual shrinking, but not total evaporation, of tar balls as a function of temperature. Under sponsorship by the U.S. Department of Energy (DOE) Atmospheric System Research (ASR) program, Peter Buseck and Kouji Adachi will repeat this experiment using a microscopy stage capable of heating rates faster than the AMS. The objective is to determine if tar balls, or some fraction, are escaping detection in a standard AMS. Radiative impacts of tar balls depend on refractive index, estimates that have an imaginary component that span several orders of magnitude (Alexander et al. 2008, Chakrabarty et al. 2010, Gelencsér, 2015). The largest reported imaginary component (0.27i) suggests that tar balls could be detected by laser incandescence or light

absorption, but then it would be difficult to reconcile reports that tar balls are the dominant type of carbonaceous aerosol (China et al. 2013) with measured single scattering albedos.

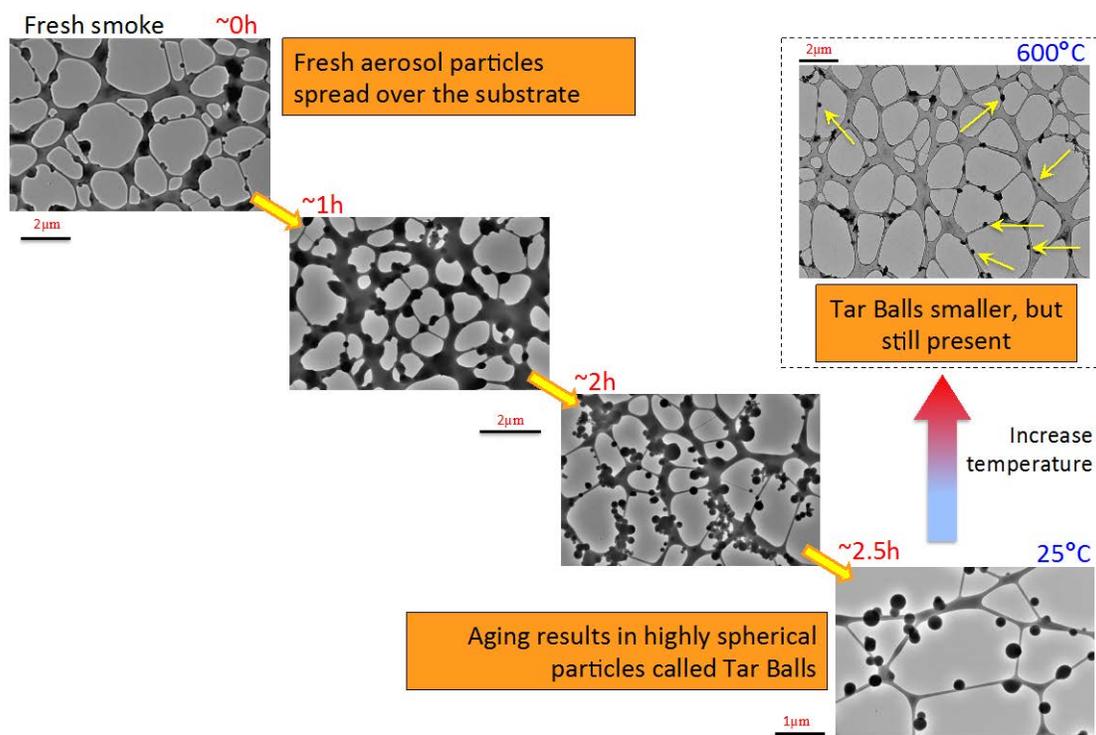


Figure 6. TEM Images of BBOP Samples. Rapid transformation of organic aerosol to tar balls observed for samples collected on flight 730b (Colockum Tarps Fire). In a separate experiment, a TEM grid was subjected to bulk heating from 25 to 600°C to examine the refractory character of tar balls (right hand side of figure). As evidenced by the image in the top right, while smaller, the tar balls show resiliency towards temperatures characteristic of the tungsten filament used in the AMS.

4.4 Spatial Homogeneity of Aerosols

The UHSAS characterizes aerosol particles by optical scattering providing a measure of atmospheric aerosol size and number density. The UHSAS also provides a PbP datastream, which measures the time of arrival for each aerosol and its size. This PbP data are then size binned for a specified time period to generate conventional UHSAS data. The aerosol time of arrival data can be used to calculate the spatial distribution of aerosols in the atmosphere. This generally is not of interest because almost all atmospheric aerosol spatial distributions follow a random Poisson distribution. A convenient parameter to test this is the Cluster Index, which is defined as $\text{Cluster Index} = \frac{\langle \text{variance}(iat) \rangle}{\langle \text{mean}(iat) \rangle} - 1$ where $\langle \rangle$ is an average over a time period and iat is the time between the arrival of one aerosol particle and the next. For most atmospheric aerosols, the spatial distribution is a random Poisson distribution, and therefore, the Cluster Index is zero. A Cluster Index greater than zero indicates that the particles are beginning to “cluster.” It is expected that aerosol particles would be initially clustered when they formed from fire processes. As an air mass ages, mixing will create a random Poisson distribution. The Cluster Index analysis has been applied to parts of the BBOP PbP data, and indeed, there are regions of clustered

aerosols. This analysis will be useful in discriminating between uniform smoke plumes and those that contain inclusions of plumes with different histories (e.g., due to downwind hot spots).

5.0 Opportunities

5.1 Radiative Forcing of Brown Carbon

Results presented here indicate that consideration should be given to treating BrC absorption as a two-component model consisting of a POA and SOA components instead of simply assuming all organic aerosols contribute equally to BrC absorption. The PAS measurements at 355 nm show that the component of BrC associated with SOA absorbs in the near-ultraviolet region and, as pointed out by Dickerson et al. (1997), will impact photochemical reactions. Impacts specific to biomass burning, including effects derived from photo-catalyzed aerosol-phase reactions, remain to be quantified.

5.2 Systematic Determination of Emission Ratios

Approximately 17 wildland and 25 agricultural fires were sampled during the BBOP. Instruments listed in Table 1 can make more than 30 distinct chemical and physical measurements, many more if the AMS data are broken down into components such as by positive matrix factorization analysis or fragment ions associated with particular precursors. Each combination of measurement and fire leads to an emission ratio by normalizing that measurement to a conservative tracer such as CO or the sum of CO and CO₂. By sheer numbers, this data set represents a significant addition to our knowledge of boreal and agricultural fire emissions.

5.3 Lagrangian Models of Aerosol Time Evolution

BBOP was unique in obtaining multiple examples of the time evolution of aerosol and gas-phase pollutants from boreal fires over the time span between approximately 15 minutes after emission (limited by air control regulations and safety) and 2 to 4 hours downwind. As Akagi et al. (2012) reported, this is a dynamic period during which changes are rapid compared to the continued evolution over time periods of order half a day to several days. Among the most significant changes are of aerosol mass, size distribution, composition (e.g., BrC), and morphology (e.g., BC coating) as these attributes affect measured single scattering albedo. Sufficient measurements exist to determine the effects of different processes; for example, 1) evaporation of POA and formation of SOA, 2) relative importance of coagulation and condensation in shaping the aerosol size distribution, and 3) importance and possibly identity of BrC absorption. The relatively simple geometry of a plume characterized by means of transects at varying downwind distances is well suited to a Lagrangian model in which the computational effort is devoted to a detailed description of chemical and size-resolved microphysical processes. Dilution is accounted for by normalizing quantities to a conservative combustion tracer such as CO (e.g., Kleinman et al. 2008, 2009). Although not as well constrained, measurements were made in locations in which fire emissions had aged and spread out over a large region, showing the effects of atmospheric processing over longer time periods. Comparisons between the G-1 and surface observations at MBO have been used for the latter objective (Collier et al. 2015).

5.4 Differences between Emission Sources

BBOP was fortunate in sampling multiple wildland fires that obviously have different chemical outcomes over the course of a few hours. The campaign also provided the opportunity to contrast wildland fires with those from prescribed agricultural burns and to determine differences between uncontrolled fires and controlled combustion as seen in five urban areas and several power plants.

5.5 Photochemical Models

Although forest fires are thought to be a significant ozone source, the details have not been quantified. Observations of boreal fires indicate variable amounts of ozone formation with a lower limit of zero (Jaffe and Wigder 2012). Similar to previous observations, wildfires sampled in during BBOP showed a wide range in ozone production that ranged from nearly zero to almost 100 ppb. Data collected during G-1 flights will allow ozone production to be examined in the context of changing NO_x , hydrocarbon, and photolysis rates as functions of downwind conditions. Preliminary analysis has yielded the unexpected result that the NO_x -to- NO_y ratio can change by an order of magnitude over 2 to 4 hours, while the toluene-to-benzene ratio decreased by only circa 20%. Chemical pathways consistent with rapid removal of NO_x by processes other than direct reaction with OH will constrain mechanisms for ozone production. The different reaction rates of NO_x and toluene will have to be taken into account in interpreting aerosol changes in terms of photochemical age.

6.0 Public Outreach

The ARM Facility organized a media day and distributed press releases. Articles about BBOP stressing connections between aerosols generated in fires to climate change appeared in approximately 15 print outlets, and video presentations appeared on local and national news programs.

7.0 BBOP Publications

7.1 Journal Articles/Manuscripts

Collier, S, S Zhou, T Onasch, D Jaffe, L Kleinman, A Sedlacek, N Wigder, J Hee, E Fortner, J Shilling, D Worsnop, RJ Yokelson, C Parworth, X Ge, J Xu, Z Butterfield, D Chand, MK Dubey, M Pekour, S Springston, and Q Zhang. 2015. "Aerosol emissions influenced by wildfire combustion efficiency in the western United States." In preparation

Sedlacek, AJ, L Kleinman, WP Arnott, T Onasch, SR Springston, S Smith, and S Oatis. 2015. "Attribution of aerosol light absorption in wildfires." In preparation

Arnott, WP, AJ Sedlacek, and JM Hubbe. 2013. "Passive noise suppression methods for improvement of photo-acoustic and photo-thermal measurements of aerosol light absorption from mobile platforms." *Proceeding for the Air Quality Measurement Methods and Technology Conference 2013*, pp. 69-75, November 19-21, 2013, Sacramento, California. Available at <http://toc.proceedings.com/21116webtoc.pdf>.

7.2 Meeting Abstracts/Presentations/Posters

FY 2015

Sedlacek, AJ and L Kleinman. 2015. “Where there’s smoke, there’s fire (and climate change).” Presentation to New York University Science, Health and Environmental Reporting Program (SHERP), September 25, 2015, New York, New York.

Onasch TB, J Shilling, E Fortner, M Pekour, D Chand, S Collier, Q Zhang, L Kleinman, A Sedlacek, A Freedman, and D Worsnop. 2015. “Chemical composition of wildland and agricultural biomass burning particles measured downwind during BBOP study.” European Aerosol Conference, September 6-11, 2015, Milan, Italy.

Sedlacek, AJ and L Kleinman. 2015. “Biomass burning aerosol optical properties in the near-field.” 11th International Conference on Carbonaceous Particles in the Atmosphere (ICCPA) Meeting, August 10-13, 2015, Lawrence Berkeley Laboratory, Berkeley, California.

Sedlacek, A and L Kleinman. 2015. “Evolution of biomass burning aerosol optical properties in the near-field.” DOE Atmospheric Systems Research (ASR) Spring Meeting, ASR-CONF-2015, March 16-20, 2015, Vienna, Virginia.

Senum, G. and Tomlinson, J. (2015) Analysis of the UHSAS Particle-by-Particle Data in BBOP: an Indicator for New Aerosol Particle Formation, ASR Spring meeting, ASR-CONF-2015, March 16-20, 2015, Vienna, VA.

Adachi, K, AJ Sedlacek, L Kleinman, D Chand, and PR Buseck. 2014. “Transmission electron microscopy analysis of biomass burning aerosol particles during BBOP campaign 2013,” Meeting abstract for the 33rd Annual American Association of Aerosol Research Conference, October 20-24, 2014, Orlando, Florida.

Zhou, S, S Collier, J Hee, N Wigder, D Jaffe, L Kleinman, L., AJ Sedlacek III, and, Q Zhang. 2014. “Aerosol chemistry and processing at Mt. Bachelor summit: influences from wildfire plumes.” 33rd Annual American Association of Aerosol Research Conference, October 20-24, 2014, Orlando, Florida.

Jaffe, D, N Wigder, P Baylon, J Hee, Q Zhang, S Zhou, S Collier, L Kleinman, and AJ Sedlacek. 2014. “Observations of wildfire smoke and ozone at the Mt. Bachelor Observatory in central Oregon.” 33rd Annual American Association of Aerosol Research Conference, October 20-24, 2014, Orlando, Florida.

Sedlacek III, AJ, WP Arnott, D Chand, E Fortner, A Freedman, L Kleinman, TB Onasch, JE Shilling, SR Springston, and S Smith. 2014. “Evolution of biomass burning optical properties in the near-field.” Presented at the Annual American Geophysical Union Fall Meeting, December 15-19, 2014, San Francisco, California.

AJ Sedlacek was co-organizer for a session focused on “Biomass Burning Impacts on Composition, Clouds, and Climate: SEAC4RS, BBOP, SAMBBA, BORTAS, FLAME-4, and other Recent Studies” (A53) Yokelson, Sedlacek, Coe, and Dibbs, co-organizers of session on December 19, 2014.

Kleinman, L, AJ Sedlacek III, R Yokelson, T Onasch, E Fortner, K Adachi, PR Buseck, D Chand, MK Dubey, F Mei, J Shilling, S Springston, J Wang, R Zaveri, and Q Zhang. "Time evolution of biomass burning plumes observed in BBOP." Poster displayed at Atmospheric System Research Science Team Meeting. <http://asr.science.energy.gov/meetings/stm/posters/view?id=1309>.

Sedlacek, AJ, L Kleinman, WP Arnott, K Adachi, P Buseck, E Lewis, T Onasch, M Pikridas, J Shilling, S Springston, J Wang, and RYokelson. 2014 "Evolution of biomass burning aerosol optical properties in the near field." *Geophysical Research Abstracts* 16, EGU2014-9226. <http://meetingorganizer.copernicus.org/EGU2014/EGU2014-9226.pdf>

Fortner, E, TB Onasch, J Shilling, M Pekour, LI Kleinman, AJ Sedlacek III, and DR Worsnop. 2015. Chemical composition of wildland and agricultural biomass burning particles measured downwind during BBOP Study. Poster presented at the 2015 ARM/ASR User Facility PI Meeting, March 16-20, 2015, Vienna, Virginia.

Buseck, PR, K Adachi, D Chand, LI Kleinman, and AJ Sedlacek III. 2014. "Transmission electron microscopy analysis of tarball formation and volatility from biomass-burning aerosol particles during the 2013 BBOP Campaign." Presented at 2014 AGU Fall Meeting, December 15-19, 2014, San Francisco, California. <https://agu.confex.com/agu/fm14/meetingapp.cgi/Paper/13747>.

Sedlacek, AJ and L Kleinman. 2014. "Aerosol science and biomass burning observation program. Presentation made to the New York University Science, Health and Environmental Reporting Program (SHERP), October 10, 2014, New York.

FY 2014

Sedlacek III, AJ, L Kleinman, K Adachi, PR Buseck, ER Lewis, TB Onasch, M Pikridas, JE Shilling, SR Springston, J Wang, RJ Yokelson. 2014. "Evolution of biomass burning aerosols in the near-field." Presented at the European Geosciences Union General Assembly, April 27-May 2, 2014, Vienna, Austria. <http://adsabs.harvard.edu/abs/2014EGUGA..16.9226S>.

Sedlacek III, AJ and L Kleinman. 2014. "Evolution of biomass burning aerosols in the near-field." Presented at the 3rd Workshop of the Interdisciplinary Biomass Burning Initiative (IBBI), April 23-26, 2014, Schloss Ringberg, Bavaria.

Collier, S, S Zhou, J Hee, N Wigder D Jaffe, L Kleinman, AJ Sedlacek III, and Q Zhang. 2014. "Highly time-resolved chemical characterization of transported wildfire plumes at Mt. Bachelor summit, 2014." Presented at International Aerosol Conference, August 28-September 2, 2014, Busan, Korea.

Wigder, N, D Jaffe, P Baylon, H Gao, J Hee, Q Zhang, S Zhou, S Collier, L Kleinman, and AJ Sedlacek. 2014. "Wildland fire modified combustion efficiency, pollutant enhancements and fire radiative power." Presented at 94th American Meteorological Society Annual Meeting, February 2-6, 2014, Atlanta, Georgia.

Yokelson, RJ, C Stockwell, PR Veres, LE Hatch, KC Barsanti, IJ Simpson, DR Blake, M Alvarado, SM Kreidenweis, AL Robinson, SK Akagi, GR McMeeking, E Stone, J Gilman, C Warneke, AJ Sedlacek, LI Kleinman. "Recent progress and emerging issues in measuring and modeling biomass

burning emissions.” Presented at the American Geophysical Union Fall Meeting, December 9-13, 2013, San Francisco, California.

Sedlacek, AJ, L Kleinman, WP Arnott, PR Buseck, ER Lewis, W Lin., TB Onasch, JE Shilling, J Wang, R Yokelson, and R Zaveri. 2013. “Biomass Burn Observation Project (BBOP) overview.” Presented at the DOE ASR Working Group Meeting, November 4-8, 2013, Rockville, Maryland.

Kleinman, L, AJ Sedlacek, WP Arnott, PR Buseck, ER Lewis, W Lin., TB Onasch, JE Shilling, J Wang, R Yokelson, and R Zaveri. 2013. “Biomass Burn Observation Project (BBOP).” Presented at the DOE ASR Fall Working Group Meeting, November 4-8, 2013, Rockville, Maryland.

Butterfield, Z, C Arata, S Liu, A Aiken, M Dubey, J Hubbe, J Comstock, B Schmid, P Arnott, S Springston, AJ Sedlacek, and L Kleinman. 2013. “G-1 Picarro CO₂, CH₄, H₂O measurements for BBOP: MCE framework for plume resolving optical property analysis.” Presented at the DOE ASR Fall Working Group Meeting, November 4-8, 2013, Rockville, Maryland.

FY 2013

Sedlacek III, AJ, ER Lewis, T Onasch, A Lambe, P Davidovits, and L Kleinman. 2012. “Probing black carbon-containing particle microphysics with the single-particle soot photometer (SP2).” Presented at the American Geophysical Union Fall Meeting, December 3-7, 2012, San Francisco, California.
http://asr.science.energy.gov/meetings/stm/posters/poster_pdf/2013/P000786.pdf.

Sedlacek, AJ and L Kleinman. 2013. “BBOP overview presentation.” Presented at the Atmospheric Systems Research (ASR) Spring Meeting, March 18-21, 2013, Potomac, Maryland.
<http://asr.science.energy.gov/meetings/stm/2013/presentations/Sedlacek-BBOP-overview.pdf>.

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