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Biomass Burning Observation Project Science Plan

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September 2013



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Abstract

Aerosols from biomass burning perturb Earth's climate through the direct radiative effect (both scattering and absorption) and through influences on cloud formation and precipitation and the semi-direct effect. Despite much effort, quantities important to determining radiative forcing such as the mass absorption coefficients (MAC) of light-absorbing carbon, secondary organic aerosol (SOA) formation rates, and cloud condensation nuclei (CCN) activity remain in doubt. Field campaigns in northern temperate latitudes have been overwhelmingly devoted to other aerosol sources in spite of biomass burning producing about one-third of the fine particles (PM_{2.5}) in the U.S.

The Atmospheric Radiation Measurement (ARM) Climate Research Facility strategy of both long-term and intensive sampling offers a path forward. We propose to conduct a campaign to measure biomass burning emissions between June and October 2013, using the Department of Energy (DOE) ARM Aerial Facility's (AAF) Gulfstream-1 (G-1) aircraft. For most of this five-month period, the G-1 will be on call at its home base in Pasco, Washington, to sample large wildfires in the Northwest. During the month of August, at the peak of the wildfire season, scientists will be present. There will also be an intensive operational period (IOP) from late September to late October in Memphis, Tennessee, to sample prescribed agricultural burns. We will sample biomass burning plumes from ~0-5 hours downwind, over which rapid changes have been observed.

Key scientific objectives are to:

1. quantify the downwind time evolution of microphysical, morphological, chemical, hygroscopic, and optical properties of aerosols generated by biomass burning
2. use the time sequences of observations to constrain processes and parameterizations in a Lagrangian model of aerosol evolution
3. incorporate time evolution information into a single-column radiative transfer model for determining forcing per unit carbon burned.

At the heart of this proposal are requested additions that will provide unique capabilities and synergisms to the G-1's usual complement of aerosol and trace gas instruments. A fast integrated mobility spectrometer (FIMS) will yield size distributions at a frequency commensurate with rapid plume transects in the hard-to-sample diameter range below 60 nm. In order to determine the morphology of single light-absorbing particles, be they soot, tar ball, or some other form, we are requesting an aerosol sampler and electron microscopy. Data will be used to test theories of the relation between near-surface soot inclusions and recently observed single-particle soot photometer (SP2) negative lag times. A soot particle aerosol mass spectrometer (SP-AMS) will allow, for the first time, quantitative airborne measurements of the chemical composition of refractory and non-refractory components of light-absorbing aerosols. A time sequence of such measurements downwind of a fire will be used to constrain the Lagrangian model MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) to investigate growth mechanisms and evolution of black carbon (BC) mixing state. The problematic determination of aerosol MACs will be attacked using refractory carbon mass determined from an SP2 in combination with absorption measurements from a second-generation photothermal interferometer (PTI) and an improved photoacoustic spectrometer (PAS). The time evolution of scattering and absorption will be combined with compositional information from the SP-AMS and morphological data from electron microscopy to test various aerosol optical models and examine radiative implications.

Summary

This field campaign will address multiple uncertainties in aerosol intensive properties, which are poorly represented in climate models, by means of aircraft measurements in biomass burning plumes. Key topics to be investigated are:

1. aerosol mixing state and morphology
2. MACs
3. chemical composition of non-refractory material associated with light-absorbing carbon (LAC)
4. production rate of SOA
5. microphysical processes relevant to determining aerosol size distributions and single-scattering albedo (SSA)
6. CCN activity.

These topics will be investigated through measurements near active fires (0–5 hours downwind), where limited observations indicate rapid changes in aerosol properties and in biomass burning plumes aged > 5 hours. Aerosol properties and their time evolution will be determined as a function of fire type, defined according to fuel and the mix of flaming and smoldering combustion at the source.

The DOE G-1 aircraft is being requested from June 1 to October 30, 2013, to be based at its home location in Pasco, Washington, except for a four-week IOP in Memphis, Tennessee. A sampling strategy has been devised that will maximize opportunities to sample both fresh biomass burn emissions and aged plumes. This strategy consists of an extended deployment of the G-1 in Pasco from July 1–August, 31, 2013, during which time targets of opportunity will be exploited and an IOP in Memphis in September–October 2013, where prescribed agricultural burns will be sampled.

This field campaign will leverage the capabilities of several new instruments or instrument combinations that have not been previously used in aircraft. Morphological studies will be made by electron microscopy (offline) and SP2 analysis (Sedlacek et al. 2012). Growth of particles with diameters < 60 nm will be determined by the high-time-resolution measurements provided by the FIMS. Quantitative measurements of the refractory and non-refractory components of particles containing BC will be provided by the soot particle aerosol mass spectrometer (SP-AMS). Deployment of four instruments devoted to light absorption or extinction (particle soot absorption photometer [PSAP], photothermal interferometer [PTI], photoacoustic spectrometer [PAS], and cavity attenuated phase shift [CAPS]) will better quantify the inherently difficult aircraft measurement of light absorption and determination of MAC.

The primary measurement objective is to quantify the time evolution of microphysical, morphological, chemical, hygroscopic, and optical properties of aerosols generated by biomass burning from near the time of formation onward.

The extended deployment at Pasco together with the IOP at Memphis will allow an examination of the dependence of evolution of biomass burn aerosol properties on fuel type. These properties will also be measured in plumes aged several days and compared with those of younger plumes.

The primary scientific objectives are to investigate:

- SOA formation rates
- structure and/or configuration of biomass burn aerosol particles
- aerosol light absorption
- composition of brown carbon (BrC)
- time evolution of the composition of refractory black carbon (rBC)
- determination of MAC
- determination of the time-series for coagulation and condensation
- CCN evolution and relation to condensed organics
- radiative transfer of biomass burns.

These will be used to:

- constrain processes and parameterizations in a detailed Lagrangian model to reproduce the time-dependent microphysics and chemistry of aerosol evolution
- incorporate time evolution information into a single-column radiative model as a first step in translating observations into a forcing per unit mass carbon burned.

In the unlikely event that only a few fires can be sampled, a set of alternative objectives related to biogenic aerosols, new particle formation (NPF) and growth, and characteristics of black carbon-containing aerosols in various environments have been defined so that productive science can be performed.

Acronyms and Abbreviations

AAF	ARM Aerial Facility
ABLE	Atmospheric Boundary Layer Experiment
ARM	Atmospheric Radiation Measurement (Climate Research Facility)
ASR	Atmospheric System Research
ASU	Arizona State University
BAe146	British Aerospace 146
BBOA	biomass burn organic aerosol
BC	Black carbon
BIBLE	Biomass Burning and Lightning Experiment
BLM	Bureau of Land Management
BNL	Brookhaven National Laboratory
BrC	brown carbon
CAPS	cavity attenuated phase shift
CARES	Carbonaceous Aerosols and Radiative Effects Study
CCD	charge-couples device
CCN	cloud condensation nuclei
CHAPS	Cumulus Humilis Aerosol Processing Study
DDA	discrete dipole approximation
DOE	Department of Energy
DOS	dioctyl sebacate
EC	elemental carbon
EPA	Environmental Protection Agency
FIMS	fast integrated mobility spectrometer
GCM	global climate model
GFS	global forecast system
IOP	intensive operational period
IPCC	Intergovernmental Panel on Climate Change
LAC	light-absorbing carbon
LED	light-emitting diode
LVOC	low-volatility organic compounds
MAC	mass absorption coefficient
MAX-MEX	Megacity Aerosol Experiment - Mexico City
MOSAIC	Model for Simulating Aerosol Interactions and Chemistry
MRI	Meteorological Research Institute
NCEP	National Centers for Environmental Prediction
NPF	new particle formation

Nd:YAG	neodymium-doped yttrium aluminum garnet
NOAA	National Oceanic and Atmospheric Administration
OC	organic carbon
PACE-5	Pacific Atmospheric Chemistry Experiment
PCASP	passive cavity aerosol spectrometer probe
PI	principal investigator
PMF	Positive matrix factorizations
POA	primary organic aerosol
PSAP	particle soot absorption photometer
PTI	photothermal interferometer
rBC	refractory black carbon
RH	relative humidity
RRTM	Rapid Radiative Transfer Model
SBIR	Small Business Innovation Research
SAFARI92	South African Fire Atmosphere Research Initiative 1992
SAFARI2000	South African Fire Atmosphere Research Initiative 2000
SCAR-B	Smoke, Clouds, and Radiation—Brazil
SMPS	scanning mobility particle sizer
SOA	secondary organic aerosol
SOAS	Southern Oxidants and Aerosol Study
SSA	single-scattering albedo
STORMVEX	Storm Peak Laboratory Cloud Property Validation Experiment
TEM	transmission electron microscopy
TOA	top of the atmosphere
TRACE-A	Transport and Atmospheric Chemistry near the Equator—Atlantic
VBS	volatility basis set
VOCALS-REx	VAMOS Ocean Cloud Atmosphere Land Study Regional Experiment

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1.0 Project Description

This aircraft-based field campaign is designed to quantify the time evolution of the microphysical, morphological, chemical, hygroscopic, and optical properties of aerosols generated by biomass burning, all of which are poorly represented in models. To accomplish this we request the DOE G-1 for a five-month period from June–October, 2013. This field campaign will meet the Atmospheric System Research (ASR) program mission of “improving the fidelity and predictive capability of global climate models requires better understanding of a multitude of fundamental aerosol and cloud life cycle processes” (ASR 2010).

1.1 Introduction and Motivation

Aerosols from biomass burning (Figure 1) are recognized to perturb Earth’s climate through the direct effect (both scattering and absorption of incoming shortwave radiation), the semi-direct effect (evaporation of cloud drops due to absorbing aerosols), and indirect effects (by influencing cloud formation and precipitation, e.g., Kaufman et al. 2002, Andrea and Rosenfeld 2008). Biomass burning is an important aerosol source, providing an estimated 50% of anthropogenically influenced fine carbonaceous particles (Bond et al. 2004, Andrea and Rosenfeld 2008, de Gouw and Jimenez 2009). Number concentration of particles from biofuel and biomass burning are comparable to sulfate on a global average (Chen et al. 2010). Primary organic aerosol (POA) from open biomass burns and biofuel comprises the largest component of primary organic aerosol mass emissions at northern temperate latitudes (de Gouw and Jimenez 2009). Data from the IMPROVE (Interagency Monitoring of Protected Visual Environments; EPA 2012) network has been used to show that in large sections of the U.S., aerosols from fires (defined here to include agricultural burns and forest fires, both prescribed and wild) are a major fraction of aerosol mass, and their year-to-year variability dominates the overall variability of aerosol loading and radiative forcing (Park et al. 2007).



Figure 1. Sample plume above boundary layer while low altitude smoke is more from smoldering by a chaparral fire in California (Akagi et al. 2012).

Many large field campaigns have focused on biomass burning in tropical regions:

- Atmospheric Boundary Layer Experiment (ABLE)
- Biomass Burning and Lightning Experiment (BIBLE)
- Pacific Atmospheric Chemistry Experiment (PACE-5)
- Smoke, Clouds, and Radiation—Brazil (SCAR-B)
- South African Fire Atmosphere Research Initiative 1992 (SAFARI92)
- South African Fire Atmosphere Research Initiative 2000 (SAFARI2000)
- Transport and Atmospheric Chemistry near the Equator—Atlantic (TRACE-A).

In contrast, relatively fewer and smaller-scale aircraft-based field campaigns focused on fire emissions have been carried out in the U.S., mostly by co-investigator Bob Yokelson (e.g., Yokelson et al. 1999, Goode et al. 2000, Burling et al. 2011). The relatively infrequent occurrence of fires in the U.S. compared to the Amazon, Africa, and southeast Asia (Wiedinmyer et al. 2011) has contributed to the comparative neglect of fire-related field campaigns in the U.S. This is particularly true when one excludes regional-scale campaigns in which biomass burning is sampled as a climatological component of the atmosphere, thousands of kilometers downwind of the source (e.g., Hecobian et al. 2011). The possibility and promise of fire observations within the U.S. directed at the temporal evolution of microphysical, morphological, chemical, hygroscopic, and optical properties of aerosols from biomass burning occurring on time scales that are amenable to study by aircraft have been demonstrated (e.g., Akagi et al. 2012, Gyawali et al. 2009, Marley et al. 2009). We intend to conduct aircraft observations to study the evolution of aerosol mixing state and morphology, black carbon MACs, chemical composition of non-refractory material associated with LAC, production rate of SOA, microphysics processes relevant to determining aerosol size distributions and SSA, and CCN activity.

Sampling biomass burning aerosols presents unique challenges. In addition to the sporadic and unpredictable nature of wildfires and the relative short lifetime of prescribed burns, fires may be spatially inhomogeneous, and part or all of a fire can contain a dynamic mix of flaming and smoldering combustion, each of which produces aerosol with different properties. Flaming combustion is characterized by the production of BC, while smoldering combustion is dominated by the production of organic carbon (OC), including BrC. BC and BrC are optically defined as light-absorbing compounds, but whereas for BC the imaginary part of their refractive index is independent of wavelength, for BrC there is a large increase in this quantity with decreasing wavelengths. Particle emissions from fires consist of, among other substances, soot, tar balls, light-absorbing organics, and refractory black carbon (rBC), which is operationally defined as a substance that incandescens within a specified temperature range in a SP2 (Schwarz et al. 2006). This rBC typically accumulates organic and inorganic substances through a combination of coagulation with smaller aerosol particles and condensation of substances from the gas phase. There is paucity of knowledge about the early stages of evolution of the properties and composition of aerosols from biomass burns, which has implications on parameterizations used in climate models.

The field campaign seeks to address this lack of knowledge through aircraft-based measurements of biomass burning aerosol properties during this early stage of evolution. The measurements that will be

obtained with the suite of instruments deployed during this campaign will allow numerous scientific questions to be addressed, as discussed in Section 1.5.

1.2 Campaign Timing and Location

Fire locations, operational characteristics of the G-1 aircraft, and other logistical concerns were considered in establishing a primary and secondary base of operations. The window of opportunity from June 1 to October 31, 2013, for using the G-1 fortuitously occurs during months with maximum fire activity in the western U.S. In a six-year study (2003–2008), more than 90% of the burned area and emissions in the western U.S. occurred during a five-month period from June to October (Urbanski et al. 2011).

To determine a location for this field campaign we examined studies in which emissions and/or fire counts were tabulated for different combinations of region, state, fire type, month, and year (Liu 2004; McCarty et al. 2007, 2009; Urbanski et al. 2011). Important criteria were proximity to regions where biomass burns typically occur (as determined by the range of the G-1), availability of aircraft support facilities, and logistics (such as location of U.S. Forest Service [USFS] field officers). Several candidate locations were assessed: Idaho Falls, Idaho; Little Rock, Arkansas; Nashville, Tennessee; Pasco, Washington; and Reno, Nevada. For each location the frequency of fires was calculated based on daily emissions of CO₂, CO, NMHC, and PM_{2.5} as determined by Moderate Resolution Imaging Spectroradiometer (MODIS) fire products and the FINNv1 (Fire INventory from NCAR) emission inventory (Wiedinmyer et al. 2011). Annual variability was estimated from 10 years of data. Fire counts and emissions were summed over areas within approximately 500 km of the candidate site. An example of the fire data products for Little Rock, Arkansas and Pasco, Washington are shown in Figure 2.

The effective range of the DOE G-1 was an important consideration for choice of deployment location. Upgrades to the G-1, including installation of more fuel-efficient engines and elimination of water/methanol tanks, are expected to increase the previous range of 400 and 1000 km, the former being a round-trip flight with close to two hours of sampling time and the latter a one-way flight with one hour of sampling time. Estimated ranges are based on a sampling speed of 100 m/s and do not take into account mission-specific factors or the additional range that would be obtained by ferrying to a burn site at high altitude. The one-way range would allow for sampling a plume at or near its source on one day, returning to base, and sampling it on the following day at its new location.

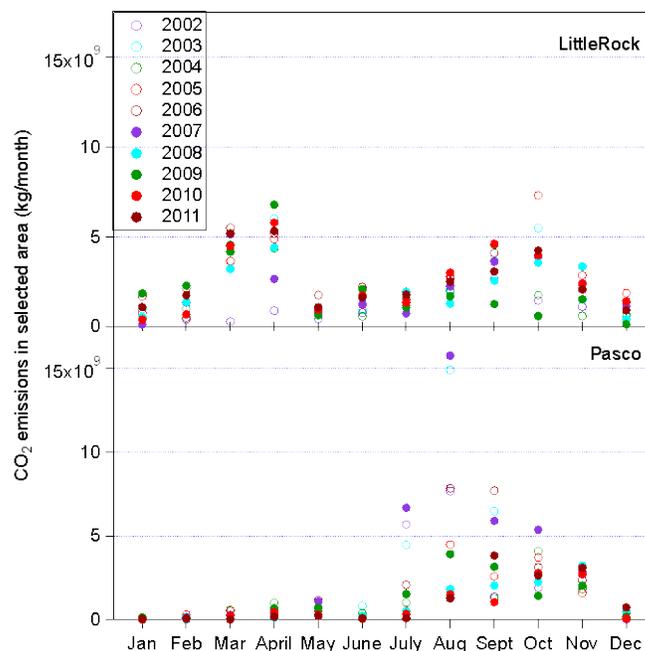


Figure 2. Monthly biomass burning CO₂ emissions for Little Rock, AR and Pasco, WA based on the MODIS fire products and FINNv1 emission inventory (Wiedinmyer et al. 2011). Increased CO₂ emissions in Pasco during July – September represent the biomass burn season, while two prescribed agricultural burn periods, March-April and September-October, are observed in Little Rock. As discussed in the text, measurement campaigns will be staged in Pasco during July/August and Little Rock in the September/October timeframe, contingent on the prior 3-month climatological forecast.

Pasco, Washington, the G-1’s home base, was selected as the long-term deployment (June to September) site based on extensive analysis of the FINNv1 emission inventory and fire count data sets. It is within range of wild land fires in Regions 1, 4, and 6 of the USFS and large areas managed by the Bureau of Land Management (BLM). Pasco offers access to a wide range of aerosol-processing environments including the humid coastal ranges and the deserts of eastern Oregon and Washington. Locating fire-sampling opportunities will be facilitated as all fire activity throughout this area is monitored closely by USFS personnel at the Ranger District and Forest Level, state dispatchers, and the National Interagency Fire Center located in Boise, Idaho (www.nifc.gov). We will stay in close contact with agency personnel and also monitor fire danger forecasts (www.spc.noaa.gov/products/fire_wx/overview.html). The Northwest wildfire season typically starts in July and finishes in September, falling within the time period of the proposal. The month of June will be devoted to instrument installation and start-up activities, leaving the outfitted G-1 available in July and August for fire-sampling opportunities. During the first part of this long-term deployment, AAF staff will be responsible for instrument operation when targets of opportunity are identified. In August, scientists will be convened at Pasco to take advantage of the heightened fire activity that typically occurs during this month (Figure 2).

Little Rock was selected for a three-week IOP using a similar strategy. This IOP will be focused on investigating agriculture residue, also known as “field” or “prescribed” burns, thereby providing a data set on a different, important fuel source.

1.3 Biomass Burn Sampling Considerations and Strategies

Much of our proposed sampling strategy follows from several tens of aircraft sampling campaigns that the principal investigators (PIs) and co-investigators have participated in, including recent DOE-sponsored studies in Mexico City (Megacity Aerosol Experiment—Mexico City [MAX-MEX]) and Sacramento (Carbonaceous Aerosols and Radiative Effects Study [CARES]) in which point source and area source plumes were followed (e.g., Springston et al. 2005; Kleinman et al. 2008, 2009; Zaveri et al. 2012). Particularly relevant to this proposal are studies of temperate zone fires, in which isolated plumes were followed in a Lagrangian frame from near a fire to several hours downwind (Akagi et al. 2012, Burling et al. 2011).

Selection of either wildfires or prescribed burns will depend upon fire type, frequency, timely knowledge of location, and ability to conduct aircraft sampling. Locating fire plumes will depend on maintaining close contact with agency personnel, fortuitous spotting from high altitude (~20,000 feet), and monitoring dedicated fire information remote sensing websites, such as the National Oceanic and Atmospheric Administration (NOAA) Fire Detection site (<http://www.firedetect.noaa.gov/viewer.htm>) and MODIS-based fire products from NASA (<http://lance.nasa.gov/data-products/modis-products/>). Prescribed fires require a spot weather forecast (special forecast issued to fit the time, topography, and weather of a specific fire; such forecasts are issued upon request of the user agency and are more detailed, timely, and specific than zone forecasts). These forecasts are posted on public websites along with telephone numbers for the conducting personnel. Permission to sample the fires is usually straightforward and consists mainly of establishing an air-ground radio frequency, as other aircraft on-site are rare. Wildfires require the establishment of a “fire-traffic area” to manage aerial fire-fighting assets. Permission to interact in a fire-traffic area with an approved radio frequency is commonly granted subject to non-problematic altitude constraints (most aerial fire assets operate at very low altitude, whereas a major fire can inject fresh smoke at high altitudes). Several miles from the plume only normal airspace regulations apply.

1.3.1 Active Fires

The near field of a biomass burn is characterized by rapid evolution of aerosol properties. As the aerosol leaves the source, it cools through dilution with unperturbed air, and this cooling affects condensation and evaporation dynamics, coagulation, hygroscopic growth, and the structure and morphology of the particles. An example of this is shown in Figure 3 where the fraction of “thickly coated” rBC particles (under the assumption of a uniformly distributed coating around an rBC core) measured by the SP2 is observed to increase rapidly following generation.

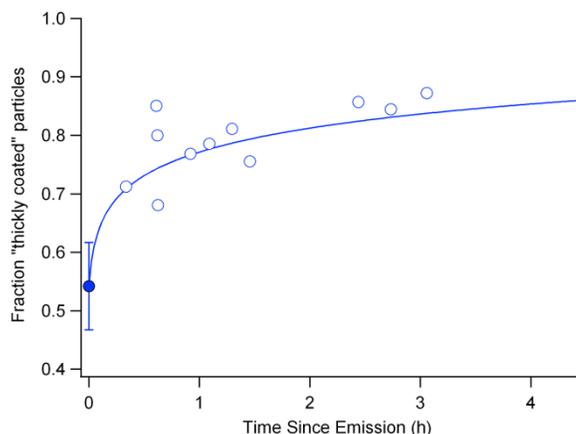


Figure 3. Fraction of “thickly-coated” rBC particles vs. time since emission during a chaparral fire in California. The mean fraction of source samples at $t = 0$ h (solid circle) with an error bar reflecting the standard deviation of the mean. Points w

Sampling in the near field will be conducted from as close to the fire as possible (hopefully within 1 km of burn) to as far downwind as the plume can be followed (typically 2–5 hours). Transects perpendicular to the plume will be flown at varying downwind distances to characterize near-source aerosol properties and the time evolution of these properties in the time range of less than 15 minutes to several hours, depending on source strength and dilution rate (using wind speed and plume size). Distinguishing time evolution of aerosol properties from changes at the source can be accomplished by flying transects in a Lagrangian frame, i.e., moving with the plume such that the aerosol sampled derived from the same time and location of the fire. Integrating across transects gives better signal-to-noise ratio (S:N) at a defined age (based on wind speed) and eliminates substantial uncertainty otherwise associated with different instrument response times. Transects will extend outside of the plume to determine properties of the unperturbed atmosphere. Spirals will also be flown to characterize the vertical development of the biomass burn aerosol properties.

1.3.2 Aged Biomass Burn Plumes

Aged biomass burn plumes can be distinguished from background by the presence of elevated concentrations of tracers such as acetonitrile (de Gouw et al. 2006, Akagi et al. 2011, Hecobian et al. 2011). The age of such plumes can be estimated by tracing back trajectories to regions with active fire detections. If concentrations are sufficiently above background and travel times on the order of one day or less, the photochemical age method (Kleinman et al. 2003) using NO_x/NO_y can be used to determine whether chemistry-based estimates of age are consistent with back trajectories. Other clocks can be investigated using high-resolution aerosol mass spectrometry (HR-AMS) such as the O:C ratio (ratio of oxygen atoms to carbon atoms), which has been found to correlate well with NO_x/NO_y (DeCarlo et al. 2008) and ratios of other hydrocarbons. The availability of these clocks permits intercomparison and evaluation to determine which clock offers greatest utility.

For times when there are no active fires within range of the G-1, attempts will be made to locate aged biomass burn plumes for sampling. Such plumes frequently impact large areas of the summertime Northwest and they are mapped on the NOAA fire detect site. These plumes will be sampled using cross wind transects and spirals to determine horizontal and vertical extent and variability.

1.4 Instruments

Instruments requested for this field campaign that will ensure that the scientific goals will be realized are listed in Table 2 in Section 3. ARM Facility instruments will be used for aerosol size distribution, optical properties, CCN activity, rBC size distribution and coating, and for trace gas measurements which are fire products and precursors to SOA. In addition to ARM instruments we are using instruments from outside investigators that will be mission-critical to one or more of our scientific objectives.

The use of new instrumentation, with new capabilities, and the synergism between instruments provides the opportunity for advancing our understanding of aerosols from biomass burning.

1.4.1 Aerosol Collection and Transmission Electron Microscopy

Prof. Buseck's group will collect aerosols on transmission electron microscopy (TEM) grids during flight and analyze them offline. The sampler has two impactor stages to collect coarse (aerodynamic diameter $>1 \mu\text{m}$) and fine (aerodynamic diameter 0.05 to $1 \mu\text{m}$) particles per sample and can collect up to 24 samples per flight. These samples will be collected during cross-plume transects at various distances from the source. TEM measurements provide unique, exquisitely detailed information on mixing state and morphology of individual particles (Pósfai and Buseck 2010). The morphological information will be both two-dimensional (2D), as is typical of most microscopy images, and three-dimensional (3D). Electron tomographic measurements will provide 3D data, including the presence and nature of pores, interstices, and whether the individual particles are coated by or embedded within other materials (Adachi et al. 2007, 2010). These microphysical properties will be determined for particles as a function of time and distance from the respective sources in order to obtain detailed information regarding the time evolution during aging (Adachi and Buseck 2011).

The electron microscopy will be done in three places and institutions: Arizona State University (ASU), Tempe, Arizona; the Meteorological Research Institute (MRI) in Tsukuba, Japan; and in TEMs available via the University of Pannonia in Veszprem, Hungary. ASU has the most complete set of TEMs for a variety of measurements, but the MRI has a new TEM that can automatically measure the compositions and sizes of ~ 100 particles in less than one hour. Electron diffraction and tomography measurements will be performed on fewer particles.

1.4.2 FIMS

FIMS has been developed for fast measurements of aerosol size distribution (Kulkarni and Wang 2006a, 2006b; Olfert et al. 2008; Olfert and Wang 2009) and was successfully deployed onboard G-1 during three major field campaigns (Cumulus Humilis Aerosol Processing Study [CHAPS], VAMOS Ocean Cloud Atmosphere Land Study Regional Experiment [VOCALS-REX], and CARES). The FIMS consists of a separator, condenser, and detector. Inside the separator charged particles are spread out based on their electrical mobility. The separated particles are then carried by a butanol-saturated sheath flow into the condenser, where a supersaturation of butanol is generated through cooling, and the classified particles grow into super-micrometer droplets. At the exit of the condenser, a laser sheet illuminates the grown droplets, and a high-speed charge-coupled device (CCD) camera captures their images. The images provide not only the particle concentration, but also the particle position, which directly relates to the particle electrical mobility. By simultaneously measuring particles of different

mobilities, the FIMS provides an aerosol size distribution in the diameter range of 30 to 100 nm at a time resolution of 1 Hz, nearly 100 times faster than traditional scanning mobility particle sizer (SMPS) systems. As individual particles and their positions are detected optically using the high-resolution CCD camera, the FIMS also offers high size resolution and good counting statistics. The FIMS will be deployed on the G-1 to provide detailed characterization of the size spectrum of biomass burning plume in this proposed study.

1.4.3 Soot Particle Aerosol Mass Spectrometer (SP-AMS)

The Aerodyne SP-AMS provides real-time, mass- and size-resolved chemical composition of submicron rBC and non-refractory particulate matter. The SP-AMS is a standard high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) with an added intracavity neodymium-doped yttrium aluminum garnet (Nd:YAG) laser vaporizer (1064 nm), based on the design used in the Droplet Measurement Technologies SP2 instrument (DeCarlo et al. 2006, Onasch et al. 2012, Schwarz et al., 2010). The laser vaporizes refractory substances, such as rBC, that are not detected in a standard AMS. The SP-AMS instrument can be operated with the laser vaporizer alone, with both the laser and tungsten vaporizers, or with the tungsten vaporizer alone. Operating with only the laser vaporizer provides a unique and selective method for measuring the size-resolved mass of the rBC (size discrimination subject to aerosol concentration and averaging time) together with the size-resolved mass and chemical composition of associated organic and inorganic material (Onasch et al. 2012). Operating with only the tungsten oven yields conventional mass spectra for non-refractory aerosol components (i.e., HR-ToF-AMS). Combining laser and oven vaporization provides quantification of refractory and non-refractory aerosol components.

The proposed work includes the first aircraft deployment of the SP-AMS. The laser vaporizer module of the SP-AMS does not interfere with any of the standard AMS hardware and has shown to be robust with respect to alignment and reproducibility across multiple recent field deployments from different measuring platforms.

Methods for (1) collecting data from AMS instruments (including the SP2-AMS to be used for this proposed work), (2) deconvoluting high-resolution mass spectra into chemical mass loading and generating organic matter (OM) to OC and O:C ratios, and (3) positive matrix factorizations (PMF) analysis techniques have been extensively developed and tested and are published in the scientific literature. For an annotated bibliography see <http://cires.colorado.edu/~jjose/ams-papers.html>.

1.4.4 Photoacoustic Spectrometer (PAS) with Scattering Module

Aerosol light absorption will be measured with a dual wavelength PAS operating at 405 nm and 870 nm. PAS measures the acoustic signal generated by particles upon dissipation of heat following light absorption. The 405-nm channel can detect enhanced absorption due to brown carbon, organic compounds that absorb strongly in the blue and Ultraviolet (UV) (Andreae and Gelencsér 2006, Moosmüller et al. 2009). Light scattering at these wavelengths will concomitantly be measured with an associated scattering module enabling SSA to be calculated directly from these scattering and absorption signals. The advantage of this approach is that the same particles contribute to the scattering and absorption.

Several modifications are or will be placed in this instrument package to improve performance on an aircraft platform. First, a new acoustical resonator has been constructed to accommodate a microphone sensor for the absorption measurement that has a frequency response much less influenced by turbulence from the aircraft aerosol inlet system. Second, a higher power and more compact 405-nm laser will be used. Third, the instrument zeroing function has been considerably sped up, minimizing data loss during this operation. Fourth, instrument drift will be minimized. An annular denuder coated with activated MnO₂ will be used to remove NO₂ from the airstream, thereby preventing interference with the 405-nm light absorption measurement.

1.4.5 PTI

A second-generation PTI will be deployed to measure light absorption at 532 nm. While possessing a similar “front end” as the PAS, the PTI relates aerosol light absorption to a change in optical path length brought about by heat dissipation from this absorption (Sedlacek and Lee 2007, Cross et al. 2010). Operation at 532 nm will complement the wavelengths employed by the PAS units.

Among the improvements made to the PTI are a new sample cell design to minimize turbulent flow, greater platform stability, and an increased detection sensitivity brought about through increased optical path length and increased laser power. As with the PAS, an activated MnO₂ denuder will be used to remove NO₂ from the airstream. Performance data on the PTI will be provided by its deployment on the British Aerospace 146 (BAe146) in the Amazon prior to the proposed campaign.

The measurements made with the PAS and PTI will be in addition to those made with a PSAP. As both of these instruments yield in situ measurements (and thus do not require filter changes), they have the unique advantage over the PSAP in that they do not suffer from filter-induced artifacts due to organic aerosols (Arnott et al. 2006, Lack et al. 2008). Measurements from all three instruments will allow quantification of these artifacts.

1.4.6 CAPS Extinction Monitor

Aerodyne Research, Inc. with funding from the DOE Small Business Innovation Research (SBIR) program, has developed a new instrument to measure the aerosol optical extinction with high precision and accuracy (Massoli et al. 2010). A light-emitting diode (LED) generates a square wave signal that is injected into one end of a sample cell is that bounded by two high reflectivity mirrors. Scattering and absorption by particles result in a distorted waveform that is characterized by a phase shift dependent on the amount of extinction. The monitor has a detection limit of $\sim 1 \text{ Mm}^{-1}$ (2σ) in 1-second integration time, which it can achieve as a result of its small sample volume (25-cm length cell) and near plug flow conditions.

1.5 Scientific Topics to be Addressed

Wildland fires are dominated by two combustion processes, flaming and smoldering (Figure 1). Flaming combustion of biomass fuel is the gas phase oxidation of organic compounds that are expelled from the fuel upon heating (Yokelson et al. 1996, Moosmüller et al. 2009). This process emits particles in the form of agglomerates that exhibit strong absorption over all wavelengths. These agglomerates (Figure 4)

consist of individual spherical monomers (spherules) of 20–40 nm diameter arranged in fractal-like morphology with a typical fractal dimension of 1.8 (Chakrabarty et al. 2006). Smoldering combustion of biomass is a combination of pyrolysis and surface oxidation of solid fuel (Moosmüller et al. 2009). During smoldering combustion, low-volatility organic compounds (LVOCs) are released (Ivlev and Popova 1973, Gao et al. 2003) and rapidly condense onto existing particles (Figure 4) (Ivlev and Popova 1973, Pósfai et al. 2003). This condensation results in spherical accumulation mode particles that consist nearly exclusively of OC and contain substantial amounts of light-absorbing BrC (Lewis et al. 2008, Gyawali et al. 2009, Chakrabarty et al. 2010).

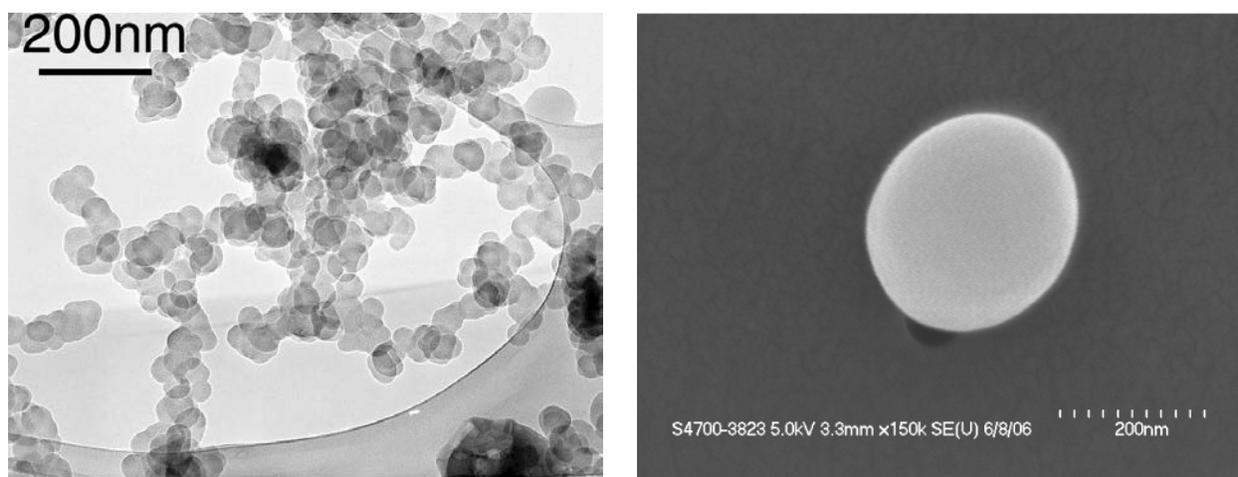


Figure 4. The left image is a typical, uncoated soot particle from biomass smoke, illustrating the fractal nature of aggregated nanospheres (from Li et al. 2003). The SEM image on the right is of a typical spherical OC particle emitted from the smoldering combustion of ponderosa pine duff (Chakrabarty et al. 2010).

In open biomass burning such as wildland fires, generally a flame front propagates across the fuel at speeds of 0.1 to several m/s, creating a convection column that lofts the flaming emissions and often most of the smoldering emissions generated by the fire. The lifetime of the flame front of prescribed fires is often only several hours, but for large wildfires it can be days to months, depending on fuel loading and conditions. For some fuel types smoldering that is not influenced by flame-induced convection can occur, producing unlofted emissions via a process known as residual smoldering combustion (Bertschi et al. 2003). Particle emissions per unit mass of fuel burned are much higher for smoldering combustion than for flaming combustion, and thus most of the emitted particles are generated by smoldering, despite the fact that flaming often consumes most of the fuel. Particles from both flaming and smoldering combustion are often collocated in turbulent plumes, resulting in the mixing of these two types and complicating source attribution.

Here we propose to observe the emitted particles once they have left the active fire region and to quantify their evolution as they travel downwind in the fire plume. Observations will include particle optical properties (i.e., absorption, scattering, extinction coefficients, and SSA) and the key underlying physical and chemical properties such as morphology and composition. These observations will allow us to improve understanding of physical and chemical processes that govern particle evolution in biomass burning plumes.

The measurements made in this field campaign will address several scientific topics, which for the present discussion are sorted into two categories: analysis of field data and modeling and radiation transfer.

1.5.1 Analysis of Field Data

1.5.1.1 SOA Formation Rates

Biomass burning events are a potentially significant source of both POA and SOA in the atmosphere. The paradigm established by Robinson and co-workers is that POA evaporates as it is released into the atmosphere and diluted, releasing large concentrations of SVOCs to the gas phase (Robinson et al. 2007). These SVOCs are subsequently oxidized, which reduces their volatility, causing them to condense back to the particle phase as SOA. This mechanism has been invoked to explain high concentrations of SOA downwind of urban areas (e.g., DeCarlo et al. 2010, Hodzic et al. 2010). Similar processes may also occur in biomass burning plumes. Indeed, laboratory studies have demonstrated significant SOA formation from wood-smoke emissions, with SOA formation accounting for up to ~4x increase in the total aerosol concentrations, with large differences being observed for different fuel, burner, and fire conditions (Grieshop et al. 2009, Hennigan et al. 2011, Miracolo et al. 2011, Heringa et al. 2011).

Downwind measurements of the ratio of OA mass to excess CO₂ or CO (Andreae and Merlet 2001, Yokelson et al. 2009) will provide an SOA formation rate as a function of photochemical age given by NO_x/NO_y (Kleinman et al. 2008) or transport age (Akagi et al. 2012). Measurements using the SP-AMS in its standard HR-AMS configuration will provide SOA data sets, including biomass burn organic aerosol (BBOA) markers. The AMS has sufficiently high resolution to allow the average elemental composition of the OA to be determined. The elemental composition, in particular the O:C ratio of the OA, provides information on aerosol aging mechanisms. PMF analysis on the AMS data set can separate the total OA mass into POA and SOA factors. The SOA produced from the fire plume may also be chemically and temporally distinct enough to allow for the separation of a “fire” SOA factor from the analysis, potentially allowing a distinction to be made several hours downwind between SOA produced from fire-derived VOCs and SOA produced from traditional biogenic VOCs (e.g., isoprene and monoterpenes) emitted for the unperturbed forest. There may be synergistic interactions between categories of SOA, as the admixture of fire-derived aerosol can provide additional solute mass that will lead to further condensation of biogenic precursors.

Observations of SOA production from fires have been interpreted to show a range of SOA mass from nearly zero (Capes et al. 2008, Akagi et al. 2012) to more than twice that of POA (Reid et al. 1998, Yokelson et al. 2009). Part of this range could be due to different processing environments (e.g., relative humidity [RH]), fuel, and fire types, whereas part may be rapidly changing near-source characteristics and the choice of the initial time (t=0). These large uncertainties in SOA production from biomass burning have substantial implications for accurately characterizing OA concentrations in the atmosphere. In addition to providing necessary inputs for determining optical and CCN properties of biomass burn aerosol, a characterization of SOA production from biomass burns will better constrain the atmospheric OA budget. Acquisition of gas phase species, including biogenic aerosol precursors and biomass burn tracers such as acetonitrile, through the deployment of the proton transfer reaction mass spectrometer (PTR-MS) and other instruments (de Gouw et al. 2006) will enable further investigation into the variable SOA production tentatively observed in biomass burning plumes.

Information on whether a substance is produced primarily by flaming or by smoldering combustion can be obtained from correlation matrices with excess CO₂ and CO in one dimension and PMF factors, individual AMS mass peaks, or other chemical concentrations in the other dimension. Substances produced in the active burning stage are characterized by higher correlation with CO₂, whereas those produced by smoldering are characterized by higher correlation with CO (Andreae and Merlet 2001, Yokelson et al. 2003).

1.5.1.2 Time Evolution of Refractory Black Carbon-Containing Particles

Recent observations from an SP2 of single particle incandescence and light scattering in which peak scattering occurred after peak incandescence (negative lag times) have been interpreted as indicating that rBC is located at or near the surface of a particle (Sedlacek et al. 2012). The high ratios (up to 60%) of such particles among all rBC-containing particles that have been measured during three different field campaigns in vastly different geographical locations (and presumably from different fuel sources) suggest that this configuration is strongly associated with biomass burning. This configuration may significantly lower light absorption compared with the simple core-shell configuration typically used in models. Direct observation of the structure of the particles using electron microscopy (as seen in tomographic reconstructions by Adachi et al. 2007) will allow the hypothesis that negative lag times result from near-surface BC to be tested. In addition to characterizing the structure of these rBC-containing particles, the time evolution of the particle morphology will be examined.

This field campaign will represent the first aircraft deployment of the Aerodyne SP-AMS instrument (Onasch et al. 2012). As discussed above, laser vaporization of rBC-containing particles enables targeted measurement of the non-refractory material associated with rBC particles and represents the first time that analysis of the composition of this material can be realized. The composition data obtained with SP-AMS will be combined with the SP2 to provide a robust data set that can be used to test various parameterizations that describe rBC aging and structure.

1.5.1.3 Relation Among rBC, Soot, LAC, and Elemental Carbon

BC has taken a variety of meanings in different fields. How it is described often depends upon the measurement, although most if not all GCMs treat BC as if it were a unique substance irrespective of how it was experimentally measured. LAC is based on light absorption whereas rBC is an operational definition based on the ability to heat to the point of incandescence. Elemental carbon (EC) is a chemical measurement, whereas soot, a well-known product in the combustion community, is a substance whose material and microphysical properties (e.g., morphology, composition, crystallographic structure, average dimensions, and mixing states) have been well described in the aerosol literature (Pósfai et al. 1999, Wentzel et al. 2003, Adachi et al. 2007, Adachi and Buseck 2008). The multiplicity of terms used to characterize what is presumed to be the same substance (although it might not be) has led to confusion and ambiguity. The wide variety of instruments that will be deployed as part of this field campaign will bring more clarity to the relationship among these definitions.

1.5.1.4 Aerosol Light Absorption

Aerosol light absorption measurements yield different results depending on the technique employed. It is most commonly measured with a filter-based technique such as a PSAP. However, artifacts due to scattering due to organic aerosols can result in absorptions that are a factor of two too high (Lack et al. 2008). The instrument suite on the G-1 will include, in addition to a PSAP, a PTI and a photoacoustic spectrometer (PAS), both of which provide direct in situ measurements of light absorption and therefore are not susceptible to the same artifacts as the PSAP. Although they have lower sensitivity than the PSAP, the PTI and PAS should have an excellent signal-to-noise ratio in high concentration fire plumes. Light absorption can also be calculated as the difference between extinction measured by the CAPS monitor (Massoli et al. 2010) and scattering measured by the 3- λ nephelometer or 2- λ PAS. Finally, theoretical values for light absorption can be obtained from calculations based on morphology determined by electron microscopy and so-called “coating” thickness and chemical composition determined by the SP2 and SP-AMS, although these values will differ depending on input parameters such as refractive indices. This will result in four observations of absorption and a set of theoretical predictions. The suite of instruments selected for this field campaign will permit comparison, evaluation, and validation among the measurements and theoretical predictions and provide insight into artifacts of filter-based techniques.

The measurements obtained during this field campaign will allow closure tests and provide an optical characterization that can be used to choose between competing descriptions. For instance, mass-equivalent diameters of rBC-containing particles and amounts of non-rBC substances (measured by SP2 and SP-AMS) and morphological information (via TEM) can be used with a Mie scattering code or a discrete dipole approximation (DDA) model to compare theoretical and experimental determinations of light absorption. The wavelength dependence of light absorption determined with the PSAP and PAS will allow investigation of the shortwave absorption by BrC contained in organic aerosol (Marley et al. 2009, Lack and Cappa 2010) and of the optical effects of a non-absorbing shell (Gyawali et al. 2009), for instance, the extent of coating-induced lensing. Results could be as simple as a multiplicative factor that scales core-shell absorption in order to account for a more complicated reality.

1.5.1.5 Brown Carbon

BrC is a class of organic aerosol components that exhibit a strong dependence of light absorption on wavelength, often resulting in high absorption in the short wavelength visible and the near-UV spectral regions (Andreae and Gelencsér 2006). Such compounds can thereby modify radiative forcing (Solomon et al. 2007) and actinic flux (Jacobson 1998). BrC aerosol has been directly observed in primary (Lewis et al. 2008, Chakrabarty et al. 2010) and aged (Gyawali et al. 2009) biomass burning emissions. Real-time measurements indicated that aerosol particles containing BrC occur as amorphous, spherical “tar balls” (Pósfai et al. 2004, Chakrabarty et al. 2006, Adachi and Buseck 2011). In a recent pilot study Chakrabarty et al. (2010) observed the first direct large-scale production of BrC-containing aerosols from smoldering combustion of two common midlatitude fuels. However, despite their presence in biomass burning plumes, much about BrC is not well characterized, such as what chemical compounds constitute BrC, its production rates and sources, its absorption strength per mass of substance, and the time evolution of its properties (Andreae and Gelencsér 2006, Moosmüller et al. 2009). This lack of knowledge has resulted in large uncertainty in the radiative forcing of BrC-containing aerosols and ultimately has contributed to the large uncertainty of IPCC 2007 estimates of direct radiative forcing due to aerosols from both biomass and fossil fuel burning (Solomon et al. 2007).

The dependence of absorption (β_{abs}) on wavelength (λ) can be characterized by the Ångström absorption exponent (AAE) defined by

$$\beta_{\text{abs}} = a\lambda^{\text{AAE}},$$

where a is independent of wavelength. Sampling periods in fire plumes with a large fraction of LAC arising from BrC can be identified according to AAE. For instance, in situ spectral characterization of aerosols with multi-wavelength photoacoustic absorption measurements have demonstrated that combustion aerosols with high SSA can have AAE values up to 3.5 (Lewis et al. 2008). In situations characterized by high AAE values, additional information on the composition and structure of these BrC-containing particles can be provided by electron microscopy and mass spectrometry. The sampling protocols and instrument suite will yield measurements that can begin to address several BrC-related topics listed above.

1.5.1.6 Determination of MAC

The MAC is defined as the light absorption coefficient of an aerosol at a given wavelength divided by the mass concentration of BC. GCMs typically parameterize light absorption as the product of mass concentration of black carbon and the MAC, with an implicit assumption that each unit mass of BC absorbs the same amount of light. However, the MAC can be affected by the presence of non-absorbing substances and by the configuration of both the absorbing and non-absorbing substances (Figure 5).

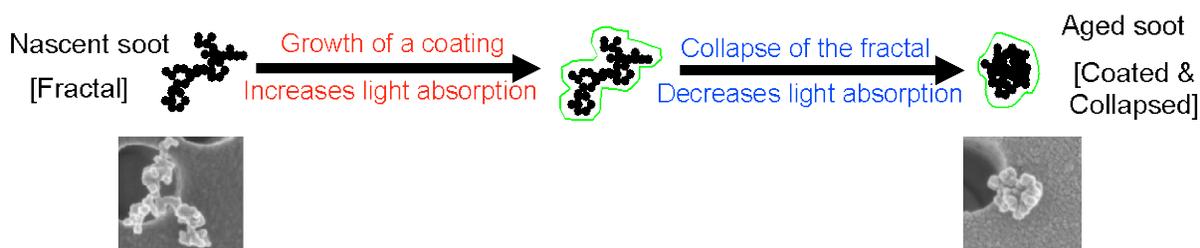


Figure 5. Atmospheric aging of soot through growth of an outer coating and collapse of the nascent fractal structure.

Measurement of the rBC mass as determined by the SP2 will be combined with measurement of light absorption as discussed above to provide values of MAC. The amount of rBC remains nearly constant with time, in contrast to the amount of BrC, which may change through chemical processing. Thus, changes in MAC can be attributed to changes in particle morphology and to changes in the amount and chemical composition of non-rBC material associated with the particles (“coating”). The presence of either absorbing (BrC) or non-absorbing coatings will increase the MAC (Bond et al. 2006, Lack and Cappa 2010) and also modify the Angstrom absorption exponent (AAE) (Gyawali et al. 2009). The sampling strategy outlined above will provide the necessary information to examine the time evolution of the MAC and its dependence on fuel type and other factors, thus improving representation of light absorption in models.

1.5.1.7 Determination of the Time-Scales for Coagulation and Condensation

Two processes important to the evolution of biomass burn aerosols are (1) condensation and evaporation of semi-volatile materials, including those from primary fire emissions and those present in the ambient

atmosphere, and (2) coagulation of individual particles. Condensation of OC or water will result in larger particles affecting light scattering and absorption and particle dynamics. Condensation might also alter chemical composition, also affecting optical properties and CCN activity. (Chamberlain et al. 1975, Hallett et al. 1989, Colbeck et al. 1990, Adachi et al. 2010). Coagulation will result in a change in the number concentration and size distribution, resulting also in changes in optical properties and CCN activity. The rates of these two processes are important for understanding of plume evolution and representation of aerosol properties in models. However, lack of high spatial and temporal resolution measurements of aerosol properties in biomass burning plumes have heretofore led to ambiguous descriptions of the particle growth mechanism (Johnson et al. 2008). The suite of measurements made during this field campaign will permit determination of these rates, therefore overcoming limitations of previous studies. The relative importance of coagulation and condensation to particle growth as a function of time can be determined by rapid measurements of aerosol number size distributions covering the diameter range 3 nm to 3 μm using CPCs, FIMS (Olfert et al. 2008), UHSAS, and PCASP (passive cavity aerosol spectrometer probe).

1.5.1.8 CCN Evolution and Relation to Condensed Organics

Biomass burn aerosols can influence radiative forcing through the indirect effect by increasing CCN concentrations and properties. While BC is typically hydrophobic, the cloud activation properties of biomass burn aerosol will be strongly dependent on the amount and composition of other substances, both inorganic and organic (Hennigan et al. 2012). These other substances are typically present. For example, the ratio of CCN/CN at 1% supersaturation for fresh biomass burning aerosol is 60–100% (Andreae and Rosenfeld 2008) consistent with the size of these particles (0.1–0.2 μm diameter) and the presence of soluble components. Andreae et al. (2004) reported that an increase CCN over the Amazon between September and November 2002 (burn season) led to a reduction in cloud droplet size that, in turn, caused the onset of precipitation at greater heights above cloud base compared to clean air conditions. Measurements of CCN activity will be made during the flights to investigate their time evolution and relation to organics as measured by the HR-AMS/SP-AMS and the PTRMS.

1.5.2 Modeling Activities Associated with Field Campaign

1.5.2.1 MOSAIC (Aerosol Box Model)

Quasi-Lagrangian observations within and outside of fire plumes will be interpreted with the comprehensive sectional aerosol box model MOSAIC (Zaveri et al. 2008). A completely constrained simulation of SOA formation is not possible at the present time due to lack of knowledge of all SOA precursor species and their volatilities and of the exact chemical and physical mechanisms of SOA formation. However, the observed evolution of SOA mass, chemical composition, and size distributions can be used to constrain the set of condensable species in MOSAIC.

SOA formation in MOSAIC is presently parameterized using a 4-bin volatility basis set (VBS) representation of condensable organic species. Gas-particle partitioning of these four SOA species is performed dynamically to size-distributed aerosols (as opposed to bulk equilibrium). Thus, in addition to simulating the mass of SOA that is formed, MOSAIC will simulate the evolution of aerosol size distribution and composition during particle growth to further constrain the underlying mechanism(s). For instance, reactive uptake growth would be controlled by aerosol surface area, while Raoult's Law-

based partitioning and particle-phase chemistry would be controlled primarily by aerosol volume, and the size distributions resulting from these processes would be quite different. An accurate description of the aerosol time evolution should therefore be able to reproduce not only the total SOA mass but also the size distribution and size-distributed chemical composition of the aged particles. We will test the sensitivity of the predicted size distribution evolution to the assumed distributions of species within the VBS framework.

Once SOA formation rates are accurately represented in the model, we will use the particle-resolved version of MOSAIC (PartMC-MOSAIC; Riemer et al. 2009, Zaveri et al. 2010) to explicitly simulate the evolution of BC mixing state, particle size distribution, and composition on the optical and cloud nucleating properties in fire plumes. The PartMC-MOSAIC model will be initialized and evaluated using SP-AMS and SP2 observations of BC mixing state.

1.5.2.2 Radiative Transfer Calculations

The Rapid Radiative Transfer Model (RRTM, Mlawer et al. 1997) will be used to translate the observed optical properties into radiative forcing. The RRTM uses a correlated-k method with 224 quadrature points across 14 spectral bands between 0.2-12.2 μm to calculate radiative transfer (RRTM_SW, Clough et al. 2005). The extinction optical depth, SSA, and the asymmetry parameter for the aerosol species will be combined for each spectral band in the radiative transfer calculation. We will calculate the radiative effects at the surface and the top of the atmosphere (TOA). Vertical profiles of atmospheric heating rates can also be calculated. The atmospheric temperature and moisture that are needed in the calculation will use the nearest analysis data in space and time from the National Centers for Forecast Prediction (NCEP) global forecast system (GFS, Kalnay et al. 1990). The magnitude and sign of biomass burning shortwave (SW) radiative effect is highly sensitive to surface albedo. We will report results using both the MODIS and AVHRR retrievals (Jin et al. 2003, Abel et al. 2005).

Areas of special focus will be the variation of radiative effects following the aging of the biomass burning aerosols, the sensitivity to vertical profiles of aerosols (results of initial emission heights and follow-up dispersion and downwind transports), and the sensitivity of radiative effects to typical prescribed size distributions (e.g., lognormal) that are used in popular climate models to account for the optical properties of biomass burning aerosols.

1.6 Plan for Situation of Few or Limited Biomass Burns

For periods with low fire activity, objectives are specified as a contrast to measurement in fires and to ensure productive use of resources.

- NPF events are often characterized by high concentrations of ultra-fine particles over wide regions. The spatial and temporal extent and variability of such high concentration events can be determined from measurements of the particle number concentration using the CPC and size distribution in the 30–100 nm diameter range using FIMS.
- Biogenic SOA formation rate could be determined by measuring the change in SOA concentration at increasing downwind distance from a forested region.

The Southern Oxidants and Aerosol Study (SOAS) sponsored by NOAA, NSF, and EPA plans to conduct aircraft and ground-based measurement campaigns in the early summer of 2013, focusing on a suite of aerosol issues including anthropogenic-biogenic interactions that foster the production of biogenic SOA. One of the staging areas for this study will be the Southeastern Aerosol Research and Characterization (SEARCH) site in Centerville, AL, ~500 km from Little Rock. Biogenic emissions decrease significantly from June to October and the G-1 would be ideally positioned to study seasonal variations in the production and properties of anthropogenic and biogenic aerosol.

2.0 Research Plan

Aerosols from biomass burning are often a major, if not dominant, source of particle number, mass and absorbing material to the atmosphere. It is estimated that that the majority of BC in the atmosphere originates from burning biomass (40%) or biofuels (20%), with the balance attributed to fossil fuels (Ramanathan and Carmichael 2008, Bond et al. 2004). The number concentration of particles from biofuel and biomass burning are comparable to sulfate on a global average (Chen et al. 2010), and POA from burning biomass are estimated to be the largest organic aerosol emissions at northern temperate latitudes (de Gouw and Jiminez 2009). Additionally, biomass burning contributes substantially to the mass concentrations measured at Interagency Monitoring of Protected Visual Environments (IMPROVE) sites in the U.S. (Park et al. 2007).

Aerosols from biomass burning contribute to the direct effect through scattering and absorption of radiation, the semi-direct effect through cloud dissipation brought about localized heating by light-absorbing aerosols (Koren et al. 2008, Ackerman et al. 2000), and the indirect effects of extending cloud lifetimes and reducing precipitation. (Kaufman et al. 2002) However, the radiative forcing contribution from BC generated in biomass burning still retains large uncertainties. These uncertainties arise from two primary reasons. The first is the difficulty in characterizing and classifying the biomass burns themselves due to their complex nature, as they may have different burn conditions (flaming versus smoldering) and a variety of fuel types (different materials, water content, etc.). The second reason is the inherent difficulty in measuring properties of aerosols from biomass burns across multiple time-scales.

A proposal for a field campaign to obtain a data set pertinent to determining properties of aerosols generated in biomass burns and the scientific justifications for such a data set were described in Section 1. An abundance of new data would result from this campaign, even if there are few burn events. These data will enable several lines of scientific inquiry to be pursued. A postdoctoral fellow at Brookhaven National Laboratory (BNL) will use these data to investigate three overarching research questions that focus on improving our process-level understanding of biomass burn aerosols:

1. What is the evolution of the structure and morphology of rBC-containing biomass burn aerosol particles, and what factors affect/determine this structure and morphology?
2. How can the structure and morphology be described/parameterized/ modeled for considerations of light scattering and absorption, and what is the sensitivity of radiative forcing to these descriptions/parameterizations/models?
3. How can the atmospheric processing of biomass burn aerosols be quantified by the use of tracers?

2.1 Structure/Morphology Evolution

What is the evolution of the structure and morphology of rBC-containing biomass burn aerosol particles (i.e., the configuration and location of the rBC within the particle), and what factors affect/determine this structure and morphology?

It has typically been assumed that a black carbon-containing particle has a core-shell structure (Figure 6, left) in which a spherical core of BC is surrounded by a concentric spherical coating composed of other substances, mainly non-refractory organic and inorganic substances such as sulfates. Even particles that are not initially spherical can, through condensation of material from the gas phase, attain configurations that are approximated by this structure (Figure 6, right). This structure is widely assumed by theorists and modelers because it is easily characterized (two radii, two indices of refraction), and its simple geometry permits analytical calculation of light scattering and absorption.

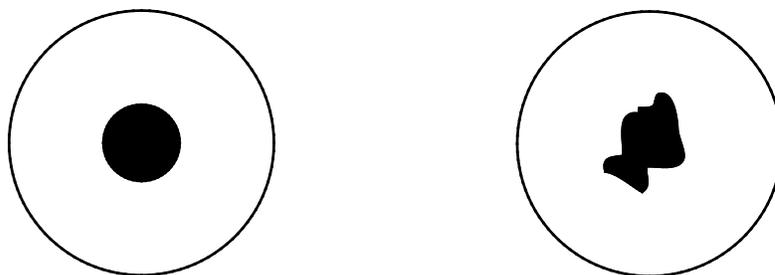


Figure 6. Idealized concentric core-shell configuration (left) used to model aged black carbon. Even black carbon aggregates that possess highly irregular shapes are thought to eventually acquire a configuration that is core-shell-like.

However, despite the wide use of this structure in the aerosol modeling community, black carbon-containing aerosol particles observed by TEM rarely, if ever, occur in such configurations (e.g., Figure 7a/b (Adachi et al. 2010)). An even more striking example of a non-core-shell configuration is provided by a scanning electron microscopy image (Figure 7c) of aerosols collected during the CARES field campaign which show BC aggregate located on the surface of the particle (unpublished results, Mazzoleni 2012). These images and other electron microscopy studies of soot-containing particles (Adachi et al. 2010, Pósfai and Buseck 2010, Adachi et al. 2011) demonstrate that the core-shell configuration may not be as common as is typically assumed.

Although electron microscopy (EM) can provide detailed information on configuration and morphology and on chemical composition of individual particles, it is labor-intensive and suffers from poor time resolution and counting statistics, thereby rendering correlations between particle configurations and field observations difficult. Collection of ambient particles onto an EM sample grid typically occurs over one minute or longer, which in the case of the DOE G-1 translates to 6 kilometers at a sampling speed of 100 meters per second. Such distances could easily extend beyond the biomass burning plume.

A new analysis methodology using the SP2 recently described by Sedlacek et al. (2012) can distinguish particles that more closely resemble a core-shell configuration from those particles where the BC aggregate is located near or at the surface of the non-refractory host with the temporal resolution and counting statistics commensurate with aircraft studies. This technique can complement EM to better interpret observed BC light absorption in ambient particles. Given the inconsistencies in the aerosol

literature regarding terminology (Bond and Bergstrom 2006), e.g., BC, soot, LAC, we adopt the nomenclature advocated by Schwarz et al. (2010) and widely used by the SP2 community that the SP2 measures rBC.

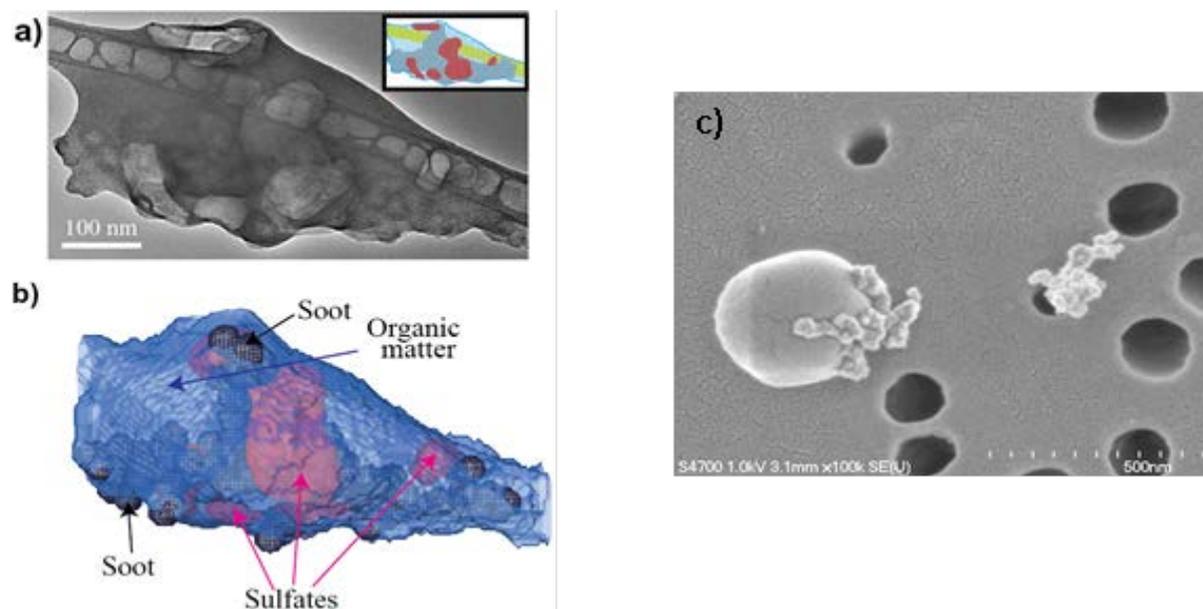


Figure 7. a) 2D TEM view of soot embedded within host aerosol particle (from Adachi et al. 2010). Inset in the upper right shows a schematic drawing of the components; blue dots denote organic material, gray dots denote soot, red dots denote voids where beam-sensitive material (presumably ammonium sulfate) was present, and yellow dots denote lacey-carbon substrate. b) 3D isosurface image of same particle (from Adachi et al. 2010). c) SEM image from CARES field campaign showing an uncoated soot particle together with a soot particle that is located on the surface of a non-refractory host (unpublished results courtesy of Mazzoleni 2012).

The SP2 determines the mass of the BC and proxies for the amount of so-called coating (i.e., non-BC material) in individual particles containing BC (Schwarz et al. 2006, Schwarz et al. 2008, Moteki and Kondo 2008, Subramanian et al. 2010) by the following methods, both of which implicitly assume a core-shell configuration.

Particles are injected into a 1064-nm laser beam whereupon the BC absorbs radiation (most particles are in the Rayleigh regime and thus Mie ambiguities do not occur). This absorbed energy is released through evaporation of the non-absorbing material, after which the BC incandesces and vaporizes. The peak intensity of incandescence is used to determine the mass of the BC. The initial scattering signal of the particle as it enters the laser beam (i.e., before much evaporation has occurred) can be used to estimate the initial radius of the particle (typically with the assumption that the particle has a single index of refraction of an equal mixture of sulfate and BC). This estimate of the initial radius and the mass of the BC yield an estimate of the coating thickness.

Another estimate of the amount of non-rBC material can be obtained from consideration of the timelag, defined as the time difference between the peak of the incandescence signal and that of the scattering signal (Schwarz et al. 2006, Moteki et al. 2007, Subramanian et al. 2010). As incandescence cannot occur

until all non-absorbing material has evaporated (according to the core-shell assumption), the timelag will be larger for thicker coatings. Typical scattering and incandescence time signals are shown in Figure 8 for thinly coated (left plot) and thickly coated (middle plot) rBC-containing particles. As with the previous method of estimating coating thickness, lack of knowledge of the properties of these non-rBC substances (index of refraction, specific heat, latent heat, etc.) prevents accurate determination of their amount.

During an IOP based on Long Island, New York, last summer, lagtime observations from an SP2 revealed the presence of particles for which the scattering peak occurred after the incandescence peak, resulting in negative lagtimes (Figure 8, right plot). Such negative lagtimes do not seem consistent with a core-shell configuration, and have been interpreted by Sedlacek et al. (2012) as indicating that rBC is located at or near the surface of such particles.

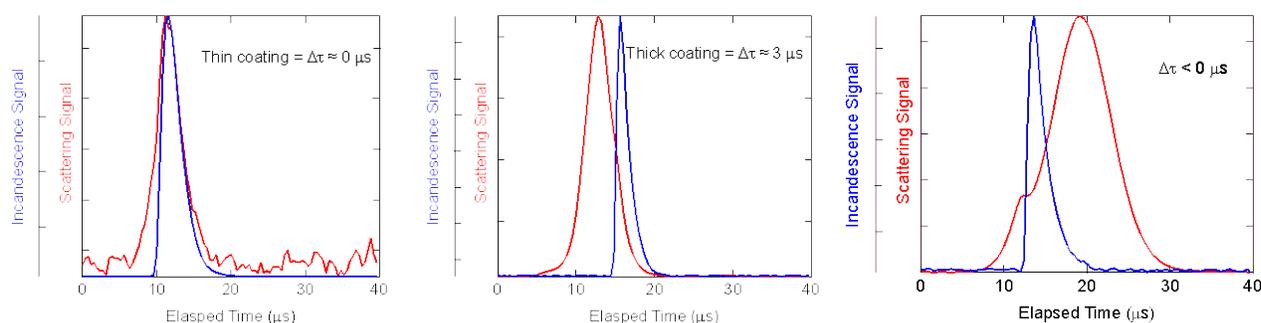


Figure 8. SP2 incandescence and scattering signals for thinly coated (left), thickly coated (middle), and near-surface rBC-containing particles (right), adapted from Sedlacek et al. 2012.

Of particular interest to the present proposal is that these negative lagtimes were observed in an air mass containing biomass burn markers. Time-series for the fraction of the rBC-containing particles that exhibited negative lagtimes, Φ_{ns} , and the ratio of the mass concentrations of biomass burning tracers $\text{C}_2\text{H}_4\text{O}_2^+$ ($m/z=60$) and $\text{C}_3\text{H}_5\text{O}_2^+$ ($m/z=73$) to total mass concentration of organics determined by HR-ToF-AMS for August 2, 2011, are shown in Figure 9. The shaded areas in the figure delineate two episodes of high rBC loading ($> 350 \text{ ng/m}^3$). The most striking features of this figure are the pronounced correlation between Φ_{ns} and biomass burning tracers and lack of correlation of Φ_{ns} with rBC loading, strongly suggesting that the rBC-containing particles exhibiting negative lagtimes derive from biomass burning events.

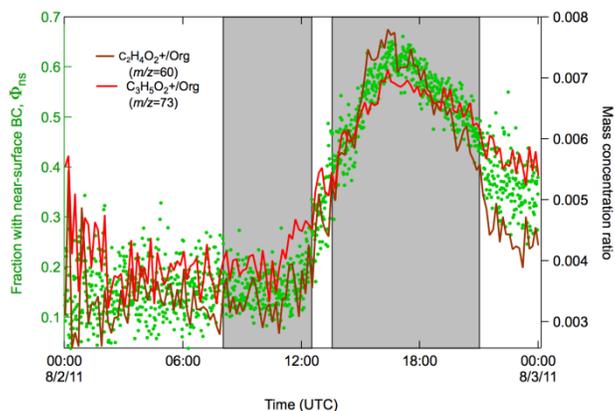


Figure 9. Fraction of near-surface rBC containing particles (green dots) and the ratio of the mass concentrations of biomass burning tracers to total mass concentration of organics. Grey boxes delineate episodes when the rBC loading exceeded 350 ng/m^3 . Adapted from Sedlacek et al. (2012).

Recent analysis of SP2 data sets collected during the CARES and Storm Peak Laboratory Cloud Property Validation Experiment (STORMVEX) field campaigns also support this hypothesis. An increase in the fraction of negative lagtimes of rBC-containing particles was observed for both a grass fire sampled during CARES and the plume from the New Mexico/Arizona wildfires sampled during STORMVEX. Taken together, these observations of negative lagtimes during three different field campaigns from three different geographical regions in the U.S. with three different fuel sources suggest that negative lagtimes, and hence the presence of rBC near the surface of particles, may be unique to or strongly associated with biomass burns. This suggestion raises several questions:

- In what fraction of rBC-containing particles is the rBC near the surface of the particle? What is the time evolution of this fraction, and what are its controlling factors?
- What mechanism is responsible for the formation of near-surface rBC-containing particles in biomass burns? What are the relative importances of coagulation and condensation at the source followed by evaporation and phase-separation?
- How does atmospheric processing (oxidation, humidification/dehumidification cycles) affect the structure and configuration of rBC-containing particles?

These questions will be investigated using analysis of field measurement data and focused laboratory experiments that allow critical aspects of this complex system to be controlled and studied in a systematic way.

An example of how laboratory experiments can be used to augment field measurements is the ongoing collaboration between BNL and Boston College with co-investigator Dr. Davidovits where investigations were carried out to explore the role of coagulation in the formation of near-surface BC-containing particles. These preliminary experiments have demonstrated that coagulation of regal black (surrogate for collapsed soot) and DOS (dioctyl sebacate; surrogate for organic material) can result in particles exhibiting negative lagtimes with the SP2. The ability to recreate negative lagtimes in the laboratory similar to those observed in the field (Figure 10) will not only enable study of the formation mechanism but also permit a careful examination of the optical properties in a systematic fashion.

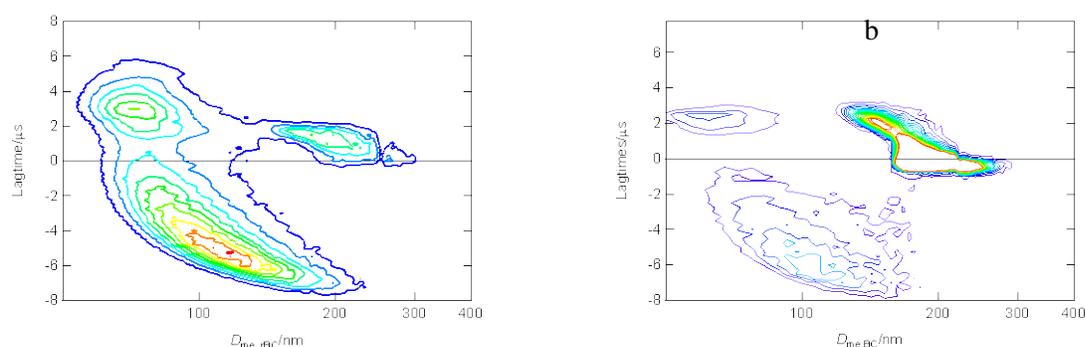


Figure 10. Incandescence lagtimes as a function of mass equivalent diameter of rBC, $D_{me, rBC}$, observed (a) on August 2 during the summer field campaign held at BNL and (b) through coagulation of regal black with DOS particles in a laboratory experiment conducted at Boston College. Measurement of regal black-DOS particles conducted after 120 minutes of mixing.

The mechanism responsible for the formation of near-surface rBC-containing particles will be investigated by comparing size distribution data from the BNL FIMS (Olfert et al. 2008) with the measured fraction of negative lagtime particles. The size distributions collected by FIMS are at a frequency commensurate with rapid plume transects and in the hard-to-sample diameter range below 60 nm. This data set will also be used in conjunction with Lagrangian calculations to identify the relative importance of coagulation and condensation to particle growth as a function of time in these biomass burn plumes.

The analysis conducted on evolution of the structure and morphology of rBC-containing particles will permit the development and testing of more refined parameterizations of optical properties of these particles. Given the large BC emission inventory that is attributed to biomass burning, such refinements in BC-related parameterizations are expected to improve model descriptions of BC radiative forcing.

2.2 Structure/Morphology Parameterization

How can the structure and morphology be described/parameterized/modeled for light scattering and absorption and what is the sensitivity of these descriptions/parameterizations/models to radiative forcing?

The structure and morphology of BC-containing particles exert a strong influence on their optical and microphysical properties that, in turn, influence their direct and indirect radiative forcing (Ackerman and Toon 1981; Bond and Bergstrom 2004; Fuller 1999; Jacobson 2000, 2001, 2012). Calculations have shown that enhancement of light absorption, for some combinations of core size and coating thickness, can easily approach a factor of two or more (Ackerman and Toon 1981, Bond and Bergstrom 2004). Measurements also yield a large enhancement in light absorption for coated BC-containing particles, as shown, for example, in Figure 11 for flame-generated soot coated with dioctyl sebacate (Cross et al. 2010 and references therein).

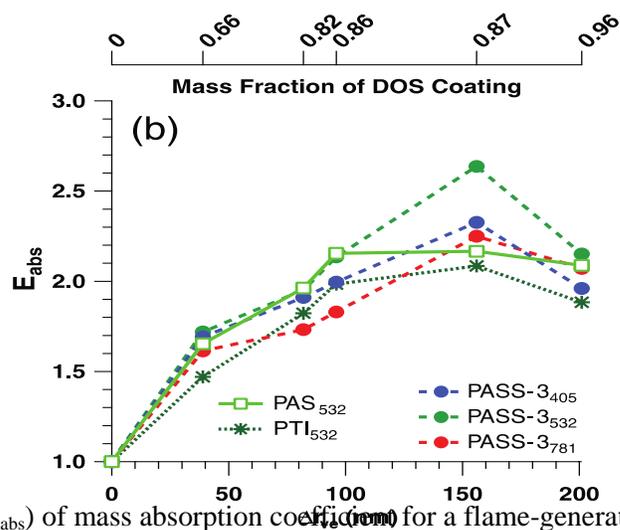


Figure 11. Ratio (E_{abs}) of mass absorption coefficient for a flame-generated soot particle coated with DOS to uncoated soot versus the change in the volume equivalent radius of the particle, Dr_{ve} , for a particle with a fractal soot core with mobility diameter $d_m = 168$ nm measured by PASS at 781, 532, and 405 nm; PTI at 532 nm; and PAS at 532 nm (Cross et al. 2010).

In addition to the influence exerted by the coating, the morphology of the BC aggregate itself can alter the optical properties of such particles. Whereas newly created BC particles are highly fractal (fractal dimension $D_f \sim 2$, Maricq and Xu 2004, Park et al. 2004), as these particles age, atmospheric interactions with water vapor, cloud drops, and organic/inorganic material can induce a restructuring of the aggregates to a more compact form (Abel et al. 2003). The more fractal structure of newly formed aggregates will exhibit a higher MAC because of the ability of most of the primary particles (spherules) to participate in light absorption, whereas the collapsed aggregate will exhibit reduced light absorption because of screening of some spherules (Penner 1986, Schnaiter et al. 2003, Saathoff et al. 2003). These two mechanisms, coating and chain aggregate collapse, will have opposite effects on absorption, greatly increasing the difficulty of quantifying the radiative forcing attributed to these particles.

The sensitivity of light absorption of BC-containing particles to their configuration is predicted by Fuller (1999) who examined the light absorption of soot with sulfate under the limiting cases of coagulation, resulting in soot and sulfate side-by-side, and of a core-shell configuration. He showed that a coagulated soot-sulfate particle would exhibit moderate enhancement in light absorption on the order of 30%, whereas the core-shell structure would exhibit an increase in light absorption by a factor of two. Schnaiter et al. (2003) explicitly examined light absorption of coagulated diesel soot/ammonium sulfate particles and reported a minor amplification in light absorption (a factor of 1.05), even less than the calculations of Fuller (1999). Recently, Bauer (2012) communicated that calculated light absorption for BC mixtures treated as a simple core-shell is higher than that estimated from AeroNet (<http://croc.gsfc.nasa.gov/aeronet/>), with the strongest absorption enhancement being associated with organic coatings. Additionally, comparison of PAS measured light absorption of ambient BC-containing particles with those that have been heated to remove associated non-refractory material appear to yield absorption enhancements of less than 10% far below those which would be predicted based on a simple core-shell configuration (Cappa et al. 2012). The fact that both the assumptions of a core-shell configuration and an internal mixture can appreciably overestimate light absorption of BC-containing particles provides a compelling argument that such configurations are not those typically encountered.

The presence of near-surface rBC-containing particles associated with biomass burning aerosols discussed above and the work of Schnaiter et al. (2003) suggest that little to no enhancement of light absorption would be expected from rBC-containing particles from biomass burns. This hypothesis will be explored by combining the SP2 data set with aerosol optical properties measured by the 3- λ nephelometer, the 3- λ PSAP, the two single-wavelength PAS instruments, the PTI, and the single-wavelength CAPS probe for total extinction. The availability of measurements at several wavelengths will also provide information on composition. For example, we will also examine the perturbative influence of coatings that are partially absorbing in the near-UV (BrC) on near-surface rBC-containing particles. This work will be performed at BNL and, as necessary, through laboratory experiments at Boston College.

Parameterizations of BC-containing particles used in global climate models (GCM) are often restricted to three aerosol/particle configurations: (1) external aerosol mixture, (2) core-shell configuration, or (3) an internally well-mixed BC particle. As part of the proposed investigation, we will develop and test parameterizations for optical properties of near-surface BC-containing particles for use in models. This effort will leverage the ongoing collaboration between BNL and the NASA Goddard Institute for Space Studies (GISS), where past work has resulted in the development of the Multiconfiguration Aerosol Tracker of Mixing State (MATRIX) model (Bauer et al. 2008).

2.3 Use of Tracers to Quantify Atmospheric Processing of Biomass Burn Aerosols

How can the atmospheric processing of biomass burn aerosols be quantified by the use of tracers?

The ability to quantify the time evolution of an aerosol plume is a critical component to successfully achieving realistic climate modeling. As part of this study into the evolution of biomass burn aerosols, we will explore the possibility of determining a set of biomass burning-specific tracers that can serve as proxies for the time since aerosol formation. A version of this approach was developed by Kleinman et al. (2008, 2009) to quantify the time evolution of urban aerosol plumes in Mexico City using the photochemical age, defined as the negative logarithm of the ratio of the concentration of NO_x to that of NO_y [$-\log_{10}(\text{NO}_x/\text{NO}_y)$]. In the application of this photochemical clock, CO is used as a conservative tracer in order to correct for the effects of dilution. This approach has proven highly effective (Figure 12), and photochemical age shows good correlation to O:C ratios in downwind measurements in Mexico City (DeCarlo et al. 2008).

Figure 12. Dependence of excess aerosol volume (above background) per excess urban CO (above background) on photochemical age given by $-\text{Log}(\text{NO}_x/\text{NO}_y)$, adapted from Kleinman et al. (2009). Each data point is the reduced major axis slope from a linear regression of aerosol volume versus CO.

The success of such an approach suggests that a similar method should be explored for characterizing biomass burning aerosol aging. Typically only a few tracers are used to determine if an aerosol plume contains contributions from biomass burning (Alfarra et al. 2007), and there is little knowledge of how these tracers vary with factors such as fire source and/or water content, strength, and temperature, air temperature, and actinic flux. One possibility is to use potassium, a conservative tracer that is characteristic of biomass burning, and organic compounds that are specific to biomass burns, such as $\text{C}_2\text{H}_4\text{O}_2^+$ and $\text{C}_3\text{H}_7\text{O}_2^+$, and their ratios to total organic aerosol loading (Alfarra et al. 2007). As with NO_x and NO_y , the concentration of the organic tracers is expected to decrease with time through oxidation. In the evaluation of the use of these biomass burn tracers as a photochemical clock, potassium will be used to correct for dilution. The SP-AMS when operating in the HR-AMS mode will be able to provide concentrations of the tracers mentioned above for a variety of well-characterized biomass burns sampled during the field campaign, in addition to concentrations of other quantities that might function as tracers. The time evolution of these concentrations from different burn events allows evaluation of the utility of the tracers that have traditionally been used and permits the possibility to devise new ones.

The need for additional tracers to understand processing of biomass burning aerosols is shown by the following example. In the time-series of organic and biomass burn markers ($\text{C}_2\text{H}_4\text{O}_2^+$, $\text{C}_3\text{H}_5\text{O}_2^+$, and potassium) collected by the HR-AMS deployed during the Aerosol Lifecycle IOP from July 23–July 24, 2011, shown in Figure 13, the potassium marker indicates that two biomass burning aerosol-containing air masses intersected the IOP site. While good correlation between the three biomass burn markers (potassium, $\text{C}_2\text{H}_4\text{O}_2^+$, $\text{C}_3\text{H}_5\text{O}_2^+$) is observed for July 23, on July 24, the peak in the potassium mass concentration lags behind the two other biomass burn tracers as well as the organic mass loading (Zhang 2012, unpublished). Based on three-day Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) back-trajectory calculations (Draxler and Rolph 2011), it is likely that the air masses encountered during the IOP experience several days transport time from the plume, and only measurements from a single location for a poorly characterized biomass burning event were available. Thus it was not possible to explain the difference in ratios and/or timing of the peaks of the concentrations of the different quantities. However, from the measurements that will be collected, this time evolution

will be available and applied to such data to help explain this observation. The ability to correlate changes in the optical and microphysical properties of biomass burn aerosols with atmospheric processing time will find immediate value in climate modeling where realistic parameterizations of biomass burn aerosol aging are needed.

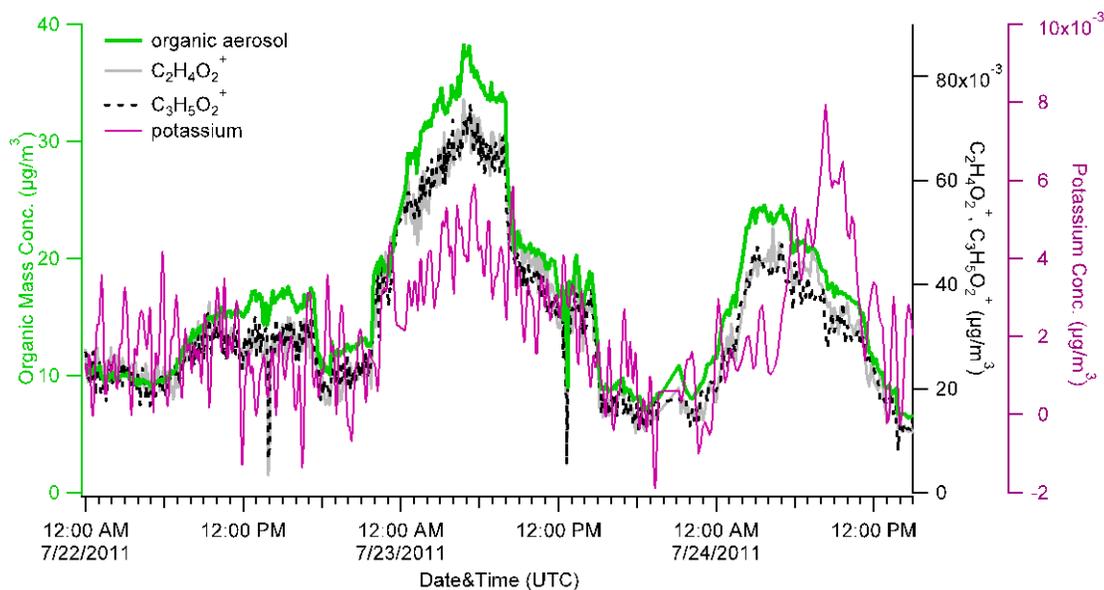


Figure 13. Time series of concentrations of organic mass and of $C_2H_4O_2^+$, $C_3H_5O_2^+$, and potassium collected by the HR-AMS deployed during the Aerosol Lifecycle IOP. On July 23, 2011, there is good correlation among the four quantities, whereas on July 24, the peak in the potassium mass concentration lags behind the other three concentrations (Zhang 2012, unpublished).

3.0 ARM Resources Required

1. One hundred and twenty G-1 flight hours are requested and will be divided between Pasco and Memphis (see Table 1) and AAF instrumentation (see Table 2) for the G-1 to sample biomass burning plumes and carry out secondary objectives. The requested flight hours are based on the following projection: 65 flight hours for June–September deployment at Pasco; 35 flight hours for the IOP based out of Memphis, TN, in October; 15 flight hours to ferry the G-1 to and from Memphis in October; 5 flight hours for pre-campaign installation and testing in June. These estimates represent an upper limit.
2. Specialized instruments not provided by AAF described in Section 1.4, that are mission-critical in addressing one or more of the scientific objectives listed in Section 1.5. Equipment requested includes (i) an aerosol sampler with offline EM performed on samples, (ii) a PTI and PAS for aerosol light absorption, (iii) a FIMS for rapid size distributions in sub-optical size range, and (iv) an SP-AMS for a quantitative determination of rBC coating composition and molecular information on rBC composition.
3. Internet connectivity to allow timely remote access of data (including calibrations) collected on the G-1 after a flight.
4. Instrument mentors or scientists cross-trained on other equipment to maintain instruments between flights and to operate instruments in flight.
5. A dilution system may be required. Particle number concentrations are expected to span one or more orders of magnitude, depending on proximity to the emission source. Alternate systems are being explored.
6. Fire forecasting and mission planning services are required. We strongly recommend that Bob Yokelson’s group at the Fire Laboratory at the University of Montana provide daily briefings on fire opportunities and sampling strategy. The field of aircraft sampling of fire plumes is a very specialized subset of aircraft operations, and Bob Yokelson is arguably the leading expert in the U.S. We propose that services provided by the University of Montana group be considered in the same category as scientists bringing mission-critical instruments to the field.

Table 1. Proposed field campaign timeline and requested flight hours for the DOE G-1.

Activity	June	July	August	September	October
Instrument Staging and G-1 Test Flights	5 hours				
Deployment at Pasco (WA)		15 hours	35 hours	15 hours	
3-week IOP at Memphis (TN)					35 hours
Round-trip Ferry Memphis to-Pasco					15 hours

Table 2. Instrument resource request.

Instrument	Measurement
3- λ PSAP	Aerosol light absorption at 450, 550, and 700 nm
3- λ nephelometer	Aerosol light scattering at 450, 550, and 700 nm
CCN counter (dual-column preferred)	CCN concentration @ 2 SS (0.25% and 0.50%)
PTRMS	Trace VOC detection
Trace gas suite	NO, NO ₂ , NO _x , CO, SO ₂ , O ₃
Meteorology	Wind direction; wind speed; air temperature; RH and rain fall
Single particle soot photometer (SP2)	BC loading, size distribution mixing state
UHSAS	Particle size distribution
PCASP	Particle size distribution
TSI-3010	Particle counter (10 nm–1 micron)
TSI-3025	Particle counter (3 nm–1 micron)
CAPS	Particle Extinction (Aerodyne)
PTI	Light absorption at 532 nm (BNL)
FIMS	Particle size distribution (BNL)
SP-AMS	Particle and coating composition (Aerodyne)
Electron microscopy	Two- and three-dimensional particle morphology and mixing state (ASU)
Photoacoustic Spectrometer (PAS)	Aerosol light absorption at 405 nm, 870 nm (University of Nevada)

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