

# **AEROSOL PROPERTIES DOWNWIND OF BIOMASS BURNS**

## **Proposal to ARM Climate Research Facility**

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### **Principal Investigators**

Lawrence I. Kleinman  
Arthur J. Sedlacek  
*Brookhaven National Laboratory*

### **Co-Investigators**

William Patrick Arnott (*University of Nevada*)  
Peter R. Buseck (*Arizona State University*)  
Paul Davidovits (*Boston College*)  
Jeffrey Steven Gaffney (*University of Arkansas*)  
Ernie R. Lewis (*Brookhaven National Laboratory*)  
Wuyin Lin (*Brookhaven National Laboratory*)  
Hans Moosmüller (*Desert Research Institute*)  
Timothy B. Onasch (*Aerodyne Research*)  
John E. Shilling (*Pacific Northwest National Laboratory*)  
Jian Wang (*Brookhaven National Laboratory*)  
Robert J. Yokelson (*University of Montana*)  
Rahul A. Zaveri (*Pacific Northwest National Laboratory*)

### **Dates and Location of Proposed Activities**

**2013-2013**

**Pasco, WA & Little Rock, AR**

**ARM AERIAL FACILITY**

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## 2. 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 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 PM<sub>2.5</sub> in the US.

The ARM 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 DOE G-1 aircraft. For most of this five month period, the G-1 will be on call at its home base in Pasco, WA 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 Observational Period (IOP) from late-September to late-October in Little Rock, AR to sample prescribed agricultural burns. We will sample biomass burning plumes from ~0-5 hours downwind, over which rapid changes have been observed. Dr. Yokelson's extensive experience with aircraft studies of biomass burns and his contacts in this field will complement the knowledge that other team members have garnered in many hundreds of hours of G-1 use.

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 FIMS (Fast Integrated Mobility Spectrometer) from Dr. Wang 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 by Prof. Buseck's team. Data will be used to test theories of the relation between near-surface soot inclusions and recently observed SP2 (Single Particle Soot Photometer) negative lag times. An SP-AMS (Soot Particle Aerosol Mass Spectrometer), from Dr. Onasch, 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) from Dr. Zaveri to investigate growth mechanisms and evolution of 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 PTI (Photothermal Interferometry) provided by Dr. Sedlacek and an improved PAS (Photoacoustic Spectroscopy) provided by Dr. Arnott. 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.

### 3. Summary of the Proposed Activity Including Major Scientific Objectives

This proposal 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) mass absorption coefficients (MACs), 3) chemical composition of non-refractory material associated with light-absorbing carbon (LAC), 4) production rate of secondary organic aerosol (SOA), 5) microphysical processes relevant to determining aerosol size distributions and single scattering albedo (SSA), and 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, WA, except for a 4 week Intensive Operating Period (IOP) in Little Rock, AR. 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 Little Rock in September-October 2013, where prescribed agricultural burns will be sampled. We also request AAF instrumentation and instruments not available in the AAF inventory, as described in Section 4.4 that will provide measurements that are essential for the scientific objectives described in Section 4.5 and 4.6.

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 Single-Particle Soot Photometer (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 Fast Integrated Mobility Spectrometer (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 mass absorption coefficients (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 Little Rock 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 Mass Absorption Coefficients (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, and*
- *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 done.

## 4. Project Description

The proposed 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 DOE-ASR mission of “Improving the fidelity and predictive capability of global climate models requires better understanding of a multitude of fundamental aerosol and cloud lifecycle processes” (ASR, 2010).

### 4.1 Introduction and Motivation

Aerosols from biomass burning (Figure 4.1) 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).

Many large field campaigns have focused on biomass burning in tropical regions (e.g. ABLE, BIBLE, PACE-5, SCAR-B, SAFARI92, SAFARI2000, TRACE-A, etc). 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-I 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 SE 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



Figure 4.1. Sample plume above boundary layer while low altitude smoke is more from smoldering by a chaparral fire in California (Akagi et al., 2012).

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 mass absorption coefficients (MACs), chemical composition of non-refractory material associated with light absorbing carbon (LAC), production rate of secondary organic aerosol (SOA), microphysics processes relevant to determining aerosol size distributions and single scattering albedo (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 black carbon (BC) while smoldering combustion is dominated by the production of organic carbon (OC), including brown carbon (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 incandesces within a specified temperature range in a Droplet Measurement Technologies “Single Particle Soot Photometer” instrument, or 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 current proposal 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 4.5.

#### **4.2 Campaign Timing and Location**

Fire locations, operational characteristics of the G-1 aircraft, and other logistic 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 US. In a six-year study (2003 – 2008), more than 90% of the burned area and emissions in the Western U.S. occurred during a 5-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, ID; Little Rock, AR; Nashville, TN; Pasco, WA; and Reno, NV. For each location the frequency of fires was calculated based on daily emissions of CO<sub>2</sub>, CO, NMHC, and PM<sub>2.5</sub> as determined by 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 (AK) and Pasco (WA) are shown in Figure 4.2.

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 sampling time and the latter, a one-way flight with one-hour 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.

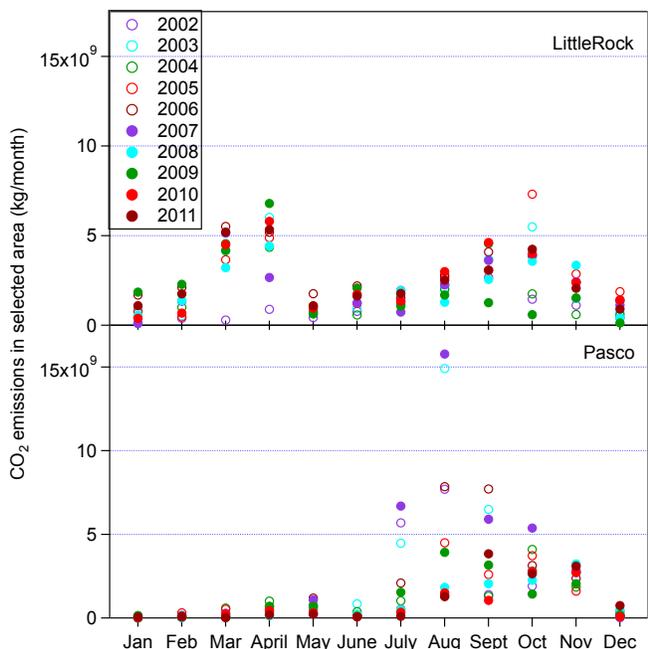


Figure 4.2. Monthly biomass burning CO<sub>2</sub> emissions for Little Rock, AR and Pasco, WA based on the MODIS fire products and FINNv1 emission inventory (Wiedinmyer et al., 2011). Increased CO<sub>2</sub> emissions in Pasco during July – September represent the biomass burn season, while two prescribed 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, WA, 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 datasets. 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 ID ([www.nifc.gov](http://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](http://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 4.2).

Little Rock was selected for a 4-week IOP in September to October using a similar strategy. This IOP will be focused on investigating agriculture residue, also known as “field” or “prescribed”, burns thereby providing a dataset on a different, important fuel source. Given the availability of the G-1 (June – October, 2013), the Little Rock IOP will target the second field burning season, as seen in Figure 4.2.

### **4.3 Biomass Burn Sampling Considerations and Strategies**

Much of our proposed sampling strategy follows from several tens of aircraft sampling campaigns that the PIs and co-I’s have participated in, including recent DOE-sponsored studies in Mexico City (MAX-MEX) and Sacramento (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 Kft) and monitoring dedicated fire information remote sensing websites, such as the NOAA firedetect 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 forecast 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. The logistics associated with research aircraft sampling near active fires will be greatly facilitated by co-I Yokelson’s experience.

#### *4.3.1 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 4.3 where the fraction of “thickly-coated” refractory black carbon (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.

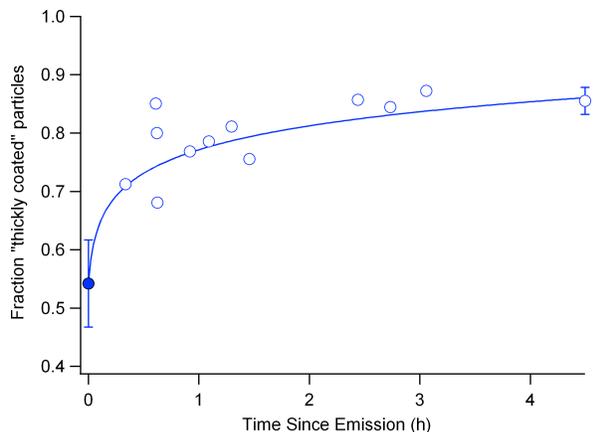


Figure 4.3. Fraction of “thickly-coated” rBC particles vs. time since emission during a chaparral fire in California. The mean fraction of source samples at  $t = 0$  h (solid circle) with an error bar reflecting the standard deviation of the mean. Points were fit to an empirical logarithmic function. (Akagi et al., 2012)

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 (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.

#### 4.3.2 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 of order 1 day or less, the photochemical age method (Kleinman et al., 2003) using  $\text{NO}_x/\text{NO}_y$  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  $\text{NO}_x/\text{NO}_y$  (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.

### 4.4 Instruments

Instruments requested for this field campaign that will ensure that the scientific goals will be realized are listed in Table 9.1 in Section 9. ACRF 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 ACRF instruments we are requesting instruments from outside investigators that will be mission critical to one or more of our scientific objectives.

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

*4.4.1 Aerosol Collection and TEM:* Prof. Buseck's group will collect aerosols on TEM grids during flight and analyze them off-line. 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, 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).

It is proposed that the electron microscopy be done in three places and institutions: Arizona State University, 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  $\sim 100$  particles in less than one hour. Electron diffraction and tomography measurements will be done on fewer particles.

*4.4.2 Fast Integrated Mobility Spectrometer (FIMS):* FIMS has been developed for fast measurements of aerosol size distribution (Kulkarni and Wang, 2006a and 2006b; Olfert et al., 2008; Olfert and Wang 2009), and was successfully deployed onboard G-1 during three major field campaigns (CHAPS, 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 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 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 G-1 to provide detailed characterization of the size spectrum of biomass burning plume in this proposed study.

*4.4.3 Soot Particle Aerosol Mass Spectrometer (SP-AMS):* The Aerodyne SP-AMS provides real-time, mass and size-resolved chemical composition of submicron refractory black carbon (rBC) and nonrefractory particulate matter. The SP-AMS is a standard high resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) with an added intracavity Nd:YAG laser vaporizer (1064 nm), based on the design used in the Droplet Measurement Technologies Single Particle Soot Photometer (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 (e.g. CalNex 2010 on R/V Atlantis, NYC 2009 on Mobile Laboratory, etc.).

Methods for 1) collecting data from AMS instruments (including the SP2-AMS to be used for this proposed work) and 2) deconvoluting high resolution mass spectra into chemical mass loading and generating organic matter to organic carbon (OM/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>.

*4.4.4. 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 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 contributed 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 MnO<sub>2</sub> will be used to remove NO<sub>2</sub> from the airstream thereby preventing interference with the 405 nm light absorption measurement

*4.4.5 Photothermal Interferometer (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 pathlength brought about by heat dissipation from this absorption. (Sedlacek and Lee 2007, Cross et al., 2010) Operation at 532 nm will compliment 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 pathlength and increased laser power. As with the PAS, an activated MnO<sub>2</sub> denuder will be used to remove NO<sub>2</sub> from the airstream. Performance data on the PTI will be provided by its deployment on the 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.

*4.4.6 Cavity Attenuated Phase Shift (CAPS) Extinction Monitor:* Aerodyne Research, Inc. with funding from the DOE 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  $\sim 1 \text{ Mm}^{-1}$  ( $2\sigma$ ) 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.

#### **4.5 Scientific Topics to be Addressed**

Wildland fires are dominated by two combustion processes, flaming and smoldering (Figure 4.1). 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 4.4a) 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 (Fig. 4.4b) (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).

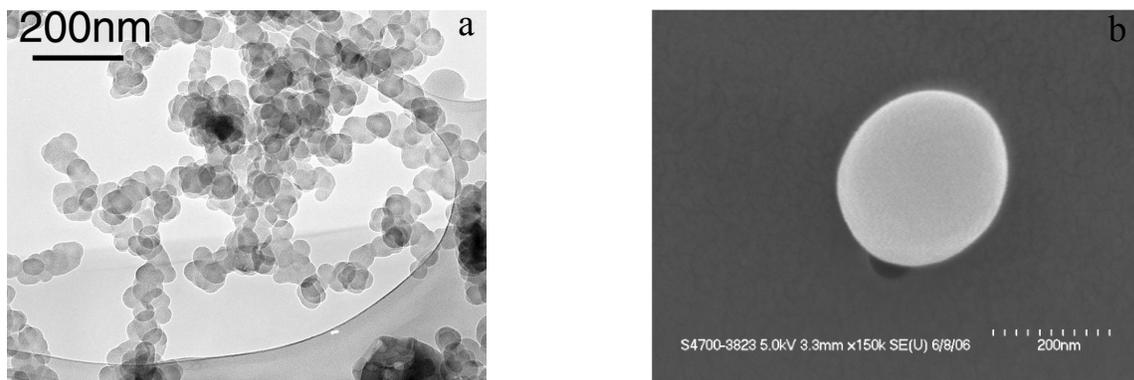


Figure 4.4. The left image is a typical, uncoated soot particle from biomass smoke, illustrating the fractal nature of aggregated nanospheres (From Li et al., 2003). The SEM image on the right is of a typical spherical OC particle emitted from the smoldering combustion of ponderosa pine duff (Chakrabarty 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 of 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 co-located 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*.

#### 4.5.1 Analysis of Field Data

**4.5.1.1 SOA Formation Rates:** Biomass burning events are a potentially significant source of both primary organic aerosol (POA) and secondary organic aerosol (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 organic compounds (primarily

semi-volatile or 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 organic aerosol (OA) mass to excess CO<sub>2</sub> 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<sub>x</sub>/NO<sub>y</sub> (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 datasets 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. Positive Matrix Factorization (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., 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 CO<sub>2</sub> 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 CO<sub>2</sub> whereas those producing by smoldering are characterized by higher correlation with CO (Andreae and Merlet, 2001; Yokelson et al., 2003).

4.5.1.2 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 lagtimes) have been interpreted as indicating that refractory black carbon (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 lagtimes 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 dataset that can be used to test various parameterizations that describe rBC aging and structure.

4.5.1.3 Relation Among rBC, Soot, LAC, and EC: Black carbon has taken a variety of meanings in different fields. How it is described often depends upon the measurement, although most if not all global climate models treat black carbon as if it were a unique substance irrespective of how it was experimentally measured. Light absorbing carbon (LAC) is based on light absorption whereas refractory black carbon (rBC) is an operational definition based on the ability to heat to the point of incandescence. Elemental carbon (EC) is 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.

4.5.1.4 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 Particle Soot Absorption Photometer (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 photothermal interferometer (PTI) and a photoacoustic spectrometer (PAS), both of which provide a direct *in situ* measurement 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 Cavity Attenuated Phase Shift (CAPS) monitor (Massoli et al., 2010) and scattering measured by the 3- $\lambda$  nephelometer or 2- $\lambda$  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/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 short-wave 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.

**4.5.1.5 Brown Carbon (BrC):** Brown carbon 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 mid-latitude fuels. However despite their presence in biomass burning plumes, much about BrC, 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, are not well characterized (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 ( $\beta_{abs}$ ) on wavelength ( $\lambda$ ) can be characterized by the Ångström absorption exponent ( $AAE$ ) defined by

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

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 single scattering albedo (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.

**4.5.1.6 Determination of Mass Absorption Coefficient (MAC):** The MAC is defined as the light absorption coefficient of an aerosol at a given wavelength divided by the mass concentration of black carbon. Global climate models 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 4.5).

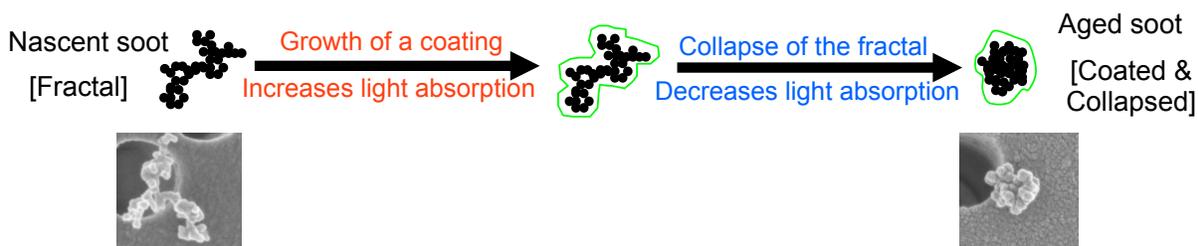


Figure 4.5 Atmospheric aging of soot through growth of an outer coating and collapse of the nascent fractal structure.

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  $AAE$  (Gyawali et al. 2009). Our 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.

**4.5.1.7 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  $\mu\text{m}$  using CPCs, FIMS (Olfert et al., 2008), UHSAS (Ultra-High Sensitivity Aerosol Spectrometer), and PCASP (Passive Cavity Aerosol Spectrometer Probe).

**4.5.1.8 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  $\mu\text{m}$  diameter) and the presence of soluble components. Andreae et al. (2004) reported an increase CCN over the Amazon between September and November 2002 (burn season) lead 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 condition. 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.

#### 4.5.2 Modeling Activities Associated with Field Campaign

**4.5.2.1 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 4 SOA species is performed dynamically to size-distributed aerosols (as opposed to bulk equilibrium). Thus, in addition to simulating the mass of SOA 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.

**4.5.2.2 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  $\mu\text{m}$  to calculate radiative transfer (RRTM\_SW, [Clough et al., 2005](#)). The extinction optical depth, single-scattering albedo, 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 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.

#### **4.6 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.

- New particle formation (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 plan 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 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. We received a letter from Prof. Annmarie Carlton of Rutgers University; PI of the SOAS campaign affirming her interest in collaborating with DOE.

## 5. 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 the majority of black carbon (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 primary organic aerosol (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 IMPROVE (Interagency Monitoring of Protected Visual Environments) 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 black carbon 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 vs. 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 4. 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. We are requesting funding for a postdoctoral fellow for 3 years at Brookhaven National Laboratory (BNL) to 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?*

**5.1 What is the evolution of the structure and morphology of rBC-containing biomass burn aerosol particles (i.e., the configuration and location of the refractory Black Carbon (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 5.1, left) in which a spherical core of black carbon 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 5.1, 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.

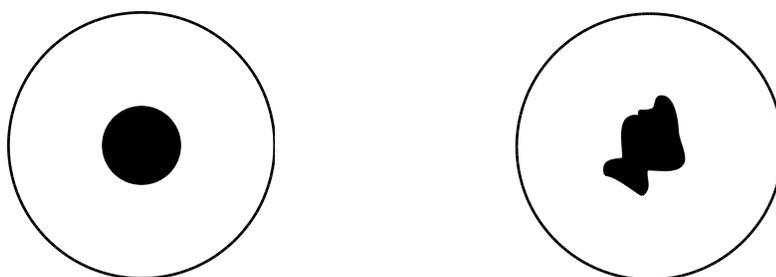


Figure 5.1. Idealized concentric core-shell configuration (left) used to model aged black carbon. Even black carbon aggregates that possess highly irregular shapes are thought to eventually acquire a configuration that is core-shell like.

However, despite the wide use of this structure in the aerosol modeling community, black carbon-containing aerosol particles observed by transmission electron microscopy (TEM) rarely, if ever, occur in such configurations; e.g., Figure 5.2a/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 5.2c) of aerosols collected during the DOE 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 km at a sampling speed of 100 m/s. Such distances could easily extend beyond the biomass burning plume.

A new analysis methodology using the Single-Particle Soot Photometer (SP2) recently described by Sedlacek et al. (2012) can distinguish particles that more closely resemble a core-shell configuration from those particles where the black carbon aggregate is located near or at the

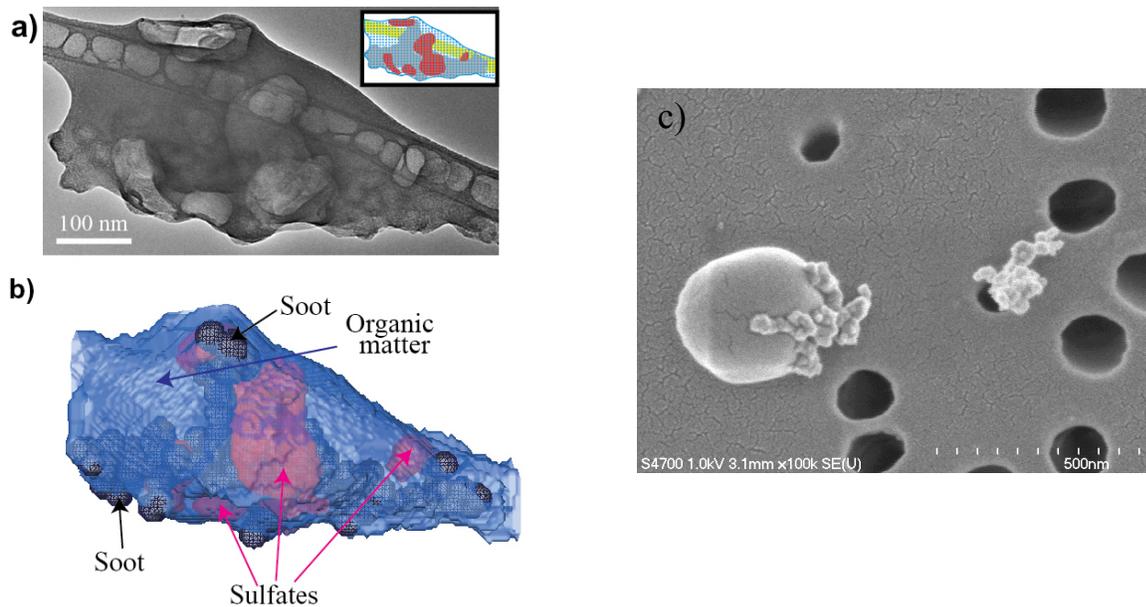


Figure 5.2. a) 2D TEM view of soot embedded within host aerosol particle (from [Adachi et al, 2010](#)). Inset in the upper right shows a schematic drawing of the components; blue dots denote organic material, gray dots denote soot, red dots denote voids where beam-sensitive material (presumably ammonium 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 DOE-sponsored 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](#)).

surface of the non-refractory host with the temporal resolution and counting statistics commensurate with aircraft studies. This technique can compliment 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, light-absorbing carbon (LAC), we adopt the nomenclature advocated by Schwarz et al. ([2010](#)) and widely used by the SP2 community that the SP2 measures refractory black carbon (rBC).

The SP2 determines the mass of the black carbon and proxies for the amount of so-called coating (i.e., non-BC material) in individual particles containing black carbon ([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 black carbon 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 black carbon incandesces and vaporizes. The peak intensity of incandescence is used to determine the mass of the black carbon. 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 black carbon). This estimate of the initial radius and the mass of the black carbon 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 5.3 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.

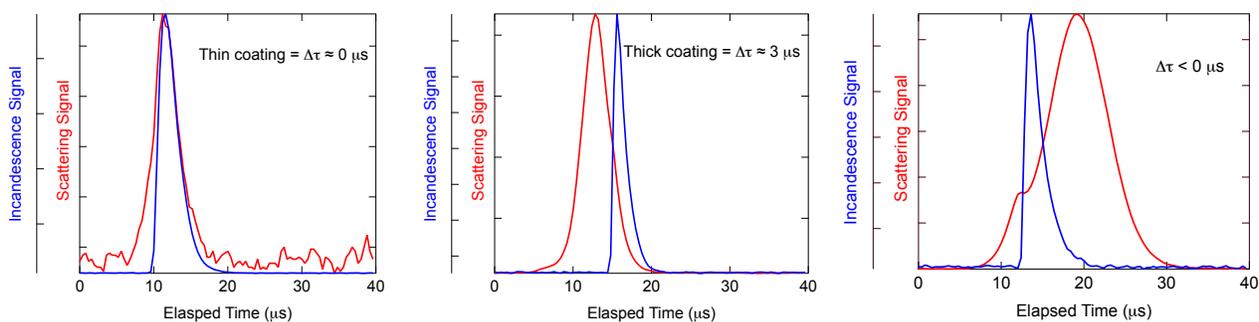


Figure 5.3. SP2 incandescence and scattering signals for thinly-coated (left), thickly-coated (middle), and near-surface rBC-containing particles (right), adapted from Sedlacek et al., 2012.

During an intensive observational period (IOP) based on Long Island, NY 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 5.3, 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.

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,  $\Phi_{ns}$ , and the ratio of the mass concentrations of biomass burning tracers  $C_2H_4O_2^+$  ( $m/z=60$ ) and  $C_3H_5O_2^+$  ( $m/z=73$ ) to total mass concentration of organics determined by HR-ToF-AMS for August 2, 2011, are shown in Figure 5.4. 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  $\Phi_{ns}$  and biomass burning tracers and lack of correlation of  $\Phi_{ns}$  with rBC loading, strongly suggesting that the rBC-containing particles exhibiting negative lagtimes derive from biomass burning events.

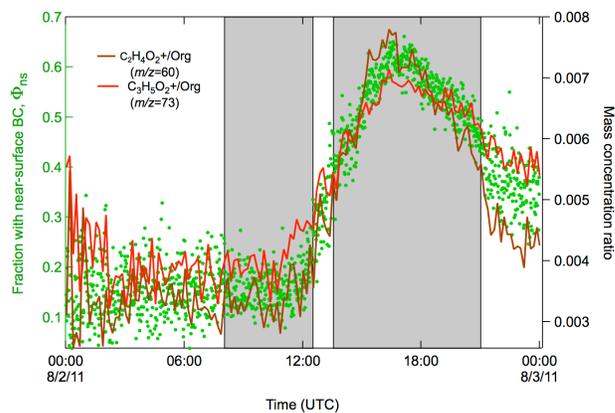


Figure 5.4. Fraction of near-surface rBC containing particles (green dots) and the ratio of the mass concentrations of biomass burning tracers to total mass concentration of organics. Grey boxes delineate episodes when the rBC loading exceeded  $350 \text{ ng/m}^3$ . Adapted from Sedlacek et al. (2012).

Recent analysis of SP2 datasets collected during the DOE-sponsored CARES and 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 the CARES campaign 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 that will be explored by BNL:

- *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-I 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 5.5) will not only enable study of the formation mechanism but also permit a careful examination of the optical properties in a systematic fashion. This interface between field measurements and

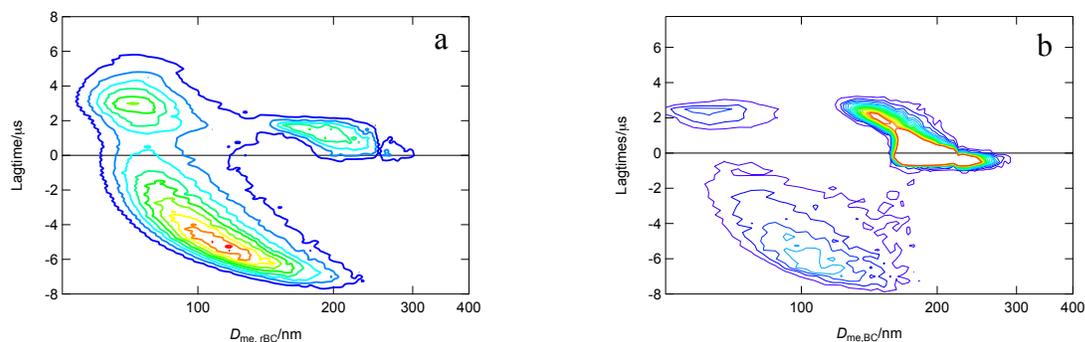


Figure 5.5. Incandescence lagtimes as a function of mass equivalent diameter of rBC,  $D_{me,rBC}$ , observed (a) on August 2 during the summer field campaign held at BNL and (b) through coagulation of regal black with DOS particles in a laboratory experiment conducted at Boston College. Measurement of regal black-DOS particles conducted after 120 minutes of mixing.

laboratory experiments exemplifies one of the strengths that members of this research team will bring to the study of these biomass burn aerosols.

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 is at a frequency commensurate with rapid plume transects, and in the hard to sample diameter range below 60 nm. This dataset 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.

### ***5.2 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; Jacobson 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 5f for flame-generated soot coated with dioctyl sebacate (Cross et al., 2010 and references therein).

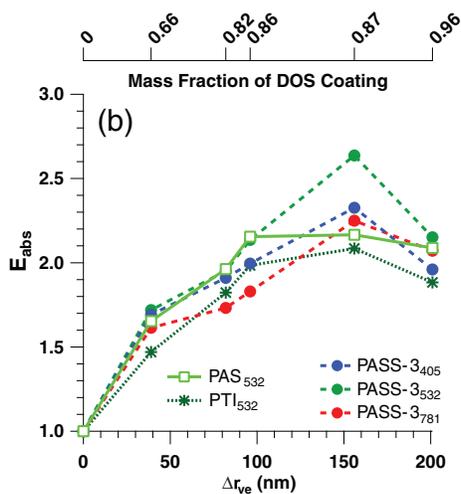


Figure 5.6. Ratio ( $E_{abs}$ ) of mass absorption coefficient for a flame-generated soot particle coated with DOS to uncoated soot versus the change in the volume equivalent radius of the particle,  $\Delta r_{ve}$ , for a particle with a fractal soot core with mobility diameter  $d_m = 168$  nm measured 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 mass absorption coefficient (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 of order 30%, whereas the core-shell structure would exhibit a factor of two increase in light absorption. 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 black carbon mixtures treated as a simple core-shell are higher than that estimated from AeroNet (<http://croc.gsfc.nasa.gov/aeronet/>), with the strongest absorption enhancement being associated with 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 that 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- $\lambda$  nephelometer, the 3- $\lambda$  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 GCM are often restricted to three aerosol/particle configurations: (i) external aerosol mixture, (ii) core-shell configuration, or (iii) 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 GISS (Dr. S.

Bauer), where past work has resulted in the development of the MATRIX model (Bauer et al., 2008).

### 5.3 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  $\text{NO}_x$  to that of  $\text{NO}_y$  [ $-\log_{10}(\text{NO}_x/\text{NO}_y)$ ]. 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 5.7), and photochemical age shows good correlation to O:C ratios in downwind measurements in Mexico City (DeCarlo et al., 2008).

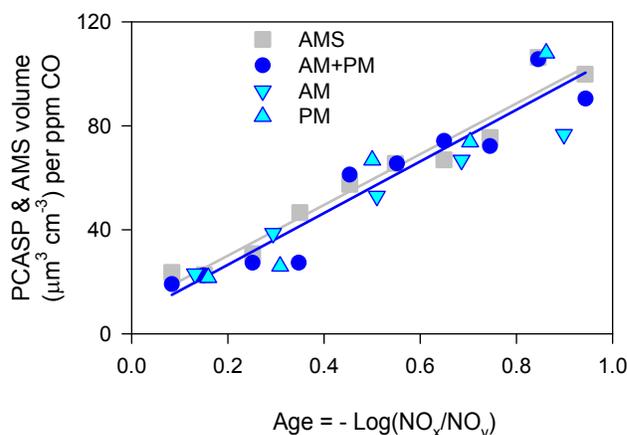


Figure 5.7. Dependence of excess aerosol volume (above background) per excess urban CO (above background) on photochemical age given by  $-\text{Log}(\text{NO}_x/\text{NO}_y)$ ; adapted from Kleinman et al. (2009). Each data point is the reduced major axis slope from a linear regression of aerosol volume vs. 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., 2006), 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., 2006). As with  $\text{NO}_x$  and  $\text{NO}_y$ , the concentration of the organic tracers are 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 ( $C_2H_4O_2^+$ ;  $C_3H_5O_2^+$ ; and potassium) collected by the HR-AMS deployed during the Aerosol Lifecycle IOP from July 23 – July 24, 2011 shown in Figure 5.8, 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,  $C_2H_4O_2^+$ ;  $C_3H_5O_2^+$ ) is observed for July 23<sup>rd</sup>, on July 24<sup>th</sup>, the peak in the potassium mass concentration lags the two other biomass burn tracers as well as the organic mass loading (unpublished, Zhang 2012). Based on three-day HYSPLIT 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.

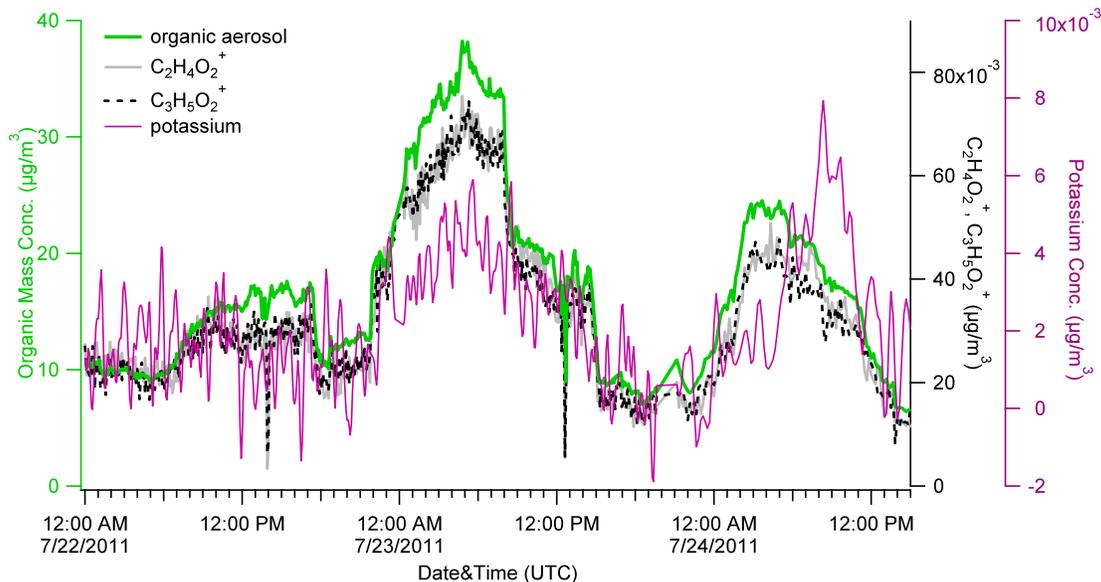


Figure 5.8. Time series of concentrations of organic mass and of  $C_2H_4O_2^+$ ,  $C_3H_5O_2^+$ , and potassium collected by the HR-AMS deployed during the aerosol lifecycle IOP. On July 23, 2011 there is good correlation among the four quantities whereas on July 24<sup>th</sup>, the peak in the potassium mass concentration lags the other three concentrations (unpublished, Zhang 2012).

## 6. Relevancy to Long-Term DOE BER Goals

**DOE Office of Biological and Environmental Research (OBER) Atmospheric Radiation Measurement (ARM) program Mission:** The ARM Climate Research Facility provides “*the climate research community with strategically located in situ and remote sensing observatories designed to improve the understanding and representation in climate and earth system models of clouds and aerosols as well as their interactions and coupling with the Earth’s surface.*”

**DOE Atmospheric Systems Research (ASR) program mission is to** “*quantify the interactions among aerosols, clouds, precipitation, radiation, dynamics, and thermodynamics to improve fundamental process-level understanding.*” Long-term data sets will be supplemented “*with laboratory studies and shorter-duration field campaigns, both ground-based and airborne, to target specific atmospheric processes under a diversity of locations and atmospheric conditions*” with the goal “*to understand and parameterize the processes that govern the atmospheric components.*”

The research described in this proposal meets both the DOE-OBER/ARM mission of conducting *in situ* measurements at strategic locations to improve the understanding and representation in climate and earth system models and the ASR program mission of improving our process-level understanding of aerosols on climate. The proposed study of mid-latitude biomass burning aerosol evolution contains research elements that will contribute to the ASR Aerosol Lifecycle and Aerosol-Cloud Interactions focus groups by providing new process-level data and analysis on biomass burn aerosols in the near-field. Where necessary, field measurements will be augmented by laboratory experiments to target gaps in our understanding of field data. Given the paucity of near-field biomass burn data from the mid-latitudes, the field campaign data and post-campaign analysis described herein are expected to yield in new insights into this important source of primary and secondary aerosols and will lead to improvements in model representations of these aerosols through refinement of parameterizations and improved understanding.

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## 8. Biographical Sketches

### Principal Investigators

Lawrence I. Kleinman  
Arthur J. Sedlacek  
*Brookhaven National Laboratory*

### Co-Investigators

William Patrick Arnott (*University of Nevada*)  
Peter R. Buseck (*Arizona State University*)  
Paul Davidovits (*Boston College*)  
Jeffrey Steven Gaffney (*University of Arkansas*)  
Ernie R. Lewis (*Brookhaven National Laboratory*)  
Wuyin Lin (*Brookhaven National Laboratory*)  
Hans Moosmüller (*Desert Research Institute*)  
Timothy B. Onasch (*Aerodyne Research*)  
John E. Shilling (*Pacific Northwest National Laboratory*)  
Jian Wang (*Brookhaven National Laboratory*)  
Robert J. Yokelson (*University of Montana*)  
Rahul A. Zaveri (*Pacific Northwest National Laboratory*)

## LAWRENCE I. KLEINMAN

[kleinman@bnl.gov](mailto:kleinman@bnl.gov)

Atmospheric Sciences Division  
Brookhaven National Laboratory

### Education

University of California at Irvine (Postdoctoral Fellow, 1971-73); Yale University (Ph.D., Chemistry, 1971); City College of New York (B.S., Chemistry, 1967).

### Research Interests

Atmospheric chemistry related to aerosols, oxidants, and climate change.

### Summary of Experience

PI for aircraft based field campaigns. With the assistance of very capable scientists within my group, field campaigns are planned, executed and the resulting data analyzed and shared with the atmospheric science community.

Research activities, in reverse chronological order:

The time evolution of aerosols and their climate affecting properties

Ozone production rates and efficiencies

Sensitivity of ozone production to NO<sub>x</sub> and VOCs

Theory of low and high NO<sub>x</sub> chemistry

Role of peroxides in atmospheric chemistry

Acid deposition

Evaluation of energy scenarios for DOE

Quantum mechanics

### Selected publications

**Kleinman, L.I.**, P. H. Daum, Y.-N. Lee, E. R. Lewis, A. J. Sedlacek III, G. I. Senum, S. R. Springston, J. Wang, J. Hubbe, J. Jayne, Q. Min, S.S. Yum, and G. Allen, Aerosol concentration and size distribution measured below, in, and above cloud from the DOE G-1 during VOCALS-REx, *Atmos. Chem. Phys.*, 12, 207-223 (2012).

Sedlacek III, A. J., Lewis, E. R., **Kleinman, L.**, Xu, J., and Zhang, Q., Determination of and evidence for non-core-shell structure of particles containing black carbon using the Single-Particle Soot Photometer (SP2), *Geophys. Res. Lett.*, 39, L06802, (2012).

Zaveri, R. A., Berkowitz, C. M., Brechtel, F. J., Gilles, M. K., Hubbe, J. M., Jayne, J. T., **Kleinman, L. I.**, Laskin, A., Madronich, S., Onasch, T. B., Pekour, M. S., Springston, S. R., Thornton, J. A., Tivanski, A. V., and Worsnop, D. R. Nighttime chemical evolution of aerosol and trace gases in a power plant plume: Implications for secondary organic nitrate and organosulfate aerosol formation, NO<sub>3</sub> radical chemistry, and N<sub>2</sub>O<sub>5</sub> heterogeneous hydrolysis. *J. Geophys. Res.* 115, D12304, doi:10.1029/2009JD013250 (2010).

**Kleinman, L. I.**, Springston, S. R., Wang, J., Daum, P. H., Lee, Y.-N., Nunnermacker, L. J., Senum, G. I., Weinstein-Lloyd, J., Alexander, M. L., Hubbe, J., Ortega, J., Zaveri, R. A., Canagaratna, M. R., and Jayne, J. The time evolution of aerosol size distribution over the Mexico City plateau. *Atmos. Chem. Phys.*, 9, 4261-4278 (2009).

- Fast, J., Aiken, A. C., Allan, J., Alexander, L., Campos, T., Canagaratna, M. R., Chapman, E., DeCarlo, P. F., deFoy, B., Gaffney, J., deGouw, J., Doran, J. C., Emmons, L., Hodzic, A., Herndon, S. C., Huey, G., Jayne, J. T., Jimenez, J. L., **Kleinman, L.**, Kuster, W., Marley, N., Russell, L., Ochoa, C., Onasch, T. B., Pekour, M., Song, C., Ulbrich, I. M., Warneke, C., Welsh-Bon, D., Wiedinmyer, C., Worsnop, D. R., Yu, X.-Y., and Zaveri, R. Evaluating simulated primary anthropogenic and biomass burning organic aerosols during MILAGRO: Implications for assessing treatments of secondary organic aerosols. *Atmos. Chem. Phys.*, **9**, 6191-6215 (2009).
- Rogers, R. R., Hair, J. W., Hostetler, C. A., Ferrare, R. A., Obland, M. D., Cook, A. L., Harper, D. B., Burton, S. P., Shinzuka, Y., McNaughton, C. S., Clarke, A. D., Redemann, J., Russell, P. B., Livingston, J. M., and **Kleinman, L. I.** NASA LaRC airborne high spectral resolution lidar aerosol measurements during MILAGRO: Observations and validation. *Atmos. Chem. Phys.*, **9**, 4811-4826 (2009).
- Kleinman, L. I.**, Springston, S. R., Daum, P. H., Lee, Y.-N., Nunnermacker, L. J., Senum, G. I., Wang, J., Weinstein-Lloyd, J., Alexander, M. L., Hubbe, J., Ortega, J., Canagaratna, M. R., and Jayne, J. The time evolution of aerosol composition over the Mexico City plateau. *Atmos. Chem. Phys.* **8**, 1559-1575 (2008).
- Nunnermacker, L. J., Weinstein-Lloyd, J. B., Hillery, B., Giebel, B., **Kleinman, L. I.**, Springston, S. R., Daum, P. H., Gaffney, J., Marley, N., and Huey, G. Aircraft and ground-based measurements of hydroperoxides during the 2006 MILAGRO field campaign. *Atmos. Chem. Phys.* **8**, 7619-7636 (2008).
- Kleinman, L. I.**, Daum, P. H., Lee, Y.-N., Senum, G. I., Springston, S. R., Wang, J., Berkowitz, C., Hubbe, J., Zaveri, R. A., Brechtel, F. J., Jayne, J., Onasch, T. B., and Worsnop, D. Aircraft observations of aerosol composition and ageing in New England and Mid-Atlantic states during the summer 2002 NEAQS Field Campaign. *J. Geophys. Res.* **112**, D09310, doi:10.1029/2006JD007786 (2007).
- Doran, J. C., Barnard, J. C., Arnott, W. P., Cary, R., Coulter, R., Fast, J. D., Kassianov, E. I., **Kleinman, L.**, Laulainen, N. S., Martin, T., Paredes-Miranda, G., Pekour, M. S., Shaw, W. J., Smith, D. F., Springston, S. R., and Yu, X.-Y. The T1-T2 study: Evolution of aerosol properties downwind of Mexico City. *Atmos. Chem. Phys.* **7**, 1585-1598 (2007)
- Wang, J., Daum, P. H., **Kleinman, L. I.**, Lee, Y.-N., Schwartz, S. E., Springston, S. R., Jonsson, H., Covert, D., and Elleman, R. Observation of ambient aerosol particle growth due to in-cloud processes within boundary layers. *J. Geophys. Res.* **112**, D14207, doi:10.1029/2006JD007989 (2007).
- Kleinman, L. I.**, Daum, P. H., Lee, Y.-N., Nunnermacker, L. J., Springston, S. R., Weinstein-Lloyd, J., and Rudolph, J. A comparative study of ozone production in 5 U.S. metropolitan areas. *J. Geophys. Res.* **110**, D02301, doi:10.1029/2004JD005096 (2005).

## **ARTHUR J. SEDLACEK, III**

Brookhaven National Laboratory

Atmospheric Science Division/Department of Environmental Sciences

Phone: (631) 344-2404

e-mail: sedlacek@bnl.gov

### Education

Ph.D., University of Utah, Chemical Dynamics, 1988

Dissertation topic: "*Photochemistry of Amorphous Solids*"

B.S., University of Wyoming, Laramie, Chemistry 1983

### Experience

Research Chemist, Brookhaven National Laboratory; Jan 1992 - present

Postdoctoral Research Associate, Brookhaven National Laboratory; Jan 1989-Sept 1992

### Research Interests

Evolution of carbonaceous aerosol optical and microphysical properties and the impact of these properties to aerosol radiative forcing; development of model aerosol systems to elucidate the effects of encapsulation on aerosol optical properties; development of optical-based instrumentation for aerosol, chemical and biological species detection; measurement of fundamental spectroscopic properties.

### Awards/Honors

DOE Certificate of Appreciation 1998

Distinguished BNL Research and Development Award – 1998

### Professional Activities and Memberships:

American Chemical Society (ACS), Optical Society of America, (OSA) American Geophysical Union (AGU), American Association for Aerosol Research (ARRR).

Served as session chairs for AGU and ARRR conferences.

Have organized several symposia on the application of optical sensors for Homeland Security for the International Society for Optical Engineering (1996, 1997, 1999, 2001, 2001, 2002, 2003, 2004, 2005) as well as serving on the Executive Program committee (2001, 2003, and 2004).

### Publications/Patents

**Sedlacek, III, Arthur**, E. R. Lewis, L. I. Kleinman, J. Xu and Q. Zhang (2012), Determination of and Evidence for Non-core-shell structure of particles containing black carbon using the single particle soot photometer (SP2). *Geophys. Res. Lett.*, 39 L06802, doi:10.1029/2012GL050905

Kleinman, L. I., P. H. Daum, Y.-N. Lee, E. R. Lewis, **A. J. Sedlacek III**, G. I. Senum, S. R. Springston, J. Wang, J. Hubbe, J. Jayne, Q. Mi, S.S. Yum, and G. Allen (2012) Aerosol concentration and size distribution measured below, in, and above cloud from the DOE G-1 during VOCALS-REx , *Atmospheric Chemistry and Physics* 11, 207-223, doi:10.5194/acp-12-207-2012.

Cross, Eben S., T. B. Onasch, A. Ahern, W. Wrobel, J. G. Slowik, J. Olfert, D. A. Lack, P. Massoli, C. D. Cappa, J. P. Schwarz, J. R. Spackman, D W. Fahey, **Arthur J. Sedlacek, III**,

- A. Trimborn, J. T. Jayne, A. Freedman, L. R. Williams, N. L. Ng, C. Mazzoleni, D. Dubey, B. Brem, G. Kok, R. Subramanian, S. Freitag, A. Clarke, D. Thornhill, L. C. Marr, C. E. Kolb, D. R. Worsnop, and P. Davidovits, Soot Particle Studies – Instrument Intercomparison – Project Overview, *Aerosol Sci. Tech.* 44 (8), 592-611 (2010).
- Springston, Stephen R. and **Arthur J. Sedlacek, III**, Noise Characteristics of an Instrumental Particle Absorbance Technique, *Aerosol Sci. Tech.* 41 (12), 1110-1116 (2007).
- Arthur J. Sedlacek, III** and Jeonghoon Lee, Photothermal Interferometric Aerosol Absorption Spectrometry, *Aerosol Sci. Tech.* 41 (12), 1089-1101 (2007).
- Arthur J. Sedlacek, III**, Real-time Detection of Ambient Aerosols using Photothermal Interferometry: Folded Jamin Interferometer, *Rev. Sci. Instrum.* 77 064903 (2006).
- Heiser, John H., and **Arthur J. Sedlacek, III**, Using Lidar to Measure Perfluorocarbon Tracers for the Verification and Monitoring of Cap and Cover Systems, *Water, Air, & Soil Pollution*, 170 345-357 (2006).
- Sedlacek III, Arthur. J.**, M. D. Ray and M. Wu, Application of UV Raman Scattering to Non-Traditional Stand-off Chemical Detection, *Trends in Applied Spectroscopy.* 5, 19-38 (2004).
- Ray, Mark D., and **Arthur J. Sedlacek, III**, Mini-Raman Sensor for the Remote, Standoff Sensing of Chemical/Biological Substances and Method for Sensing Same, *U.S. Patent 6,606,677* (Granted June 22, 2004).
- Sedlacek III, Arthur J.**, M. D. Ray, N. S. Higdon, and D. A. Richter, Short-Range Noncontact Detection of Surface Contamination Using Raman Lidar, *SPIE Proceedings: Vibrational Spectroscopy-based Sensor Systems*, 4577 95 (2001).
- Ray, Mark D., **Arthur J. Sedlacek III**, and M. Wu, Ultraviolet Mini-Raman Lidar for Stand-off, *In situ* Identification of Chemical Surface Contamination, *Rev. of Sci. Instr.* 71 3485 (2000).
- Selected Presentations
- Determination of and Evidence for Non-Core-Shell Structure of Particles Containing Black Carbon* **Sedlacek III, Arthur J.**, E. Lewis, L. Kleinman, Q. Zhang, J. Xu (2011) AGU Fall meeting, Dec. 4-9, 2011.
- Evolution of Black Carbon Mixing-State in an Urban-Biogenic Environment* (2011) **Sedlacek III, Arthur J.**, L. Kleinman, J. E. Shilling, R. Subramanian, S. R. Springston, and R. Zaveri, (2011) -, 10th International Conference on Carbonaceous Particles in the Atmosphere; June 27, 2011.
- Processing of Black Carbon in the Mixed Sacramento Urban-Biogenic Environment* (2011) **Arthur J. Sedlacek, III**, Larry Kleinman, John E. Shilling, Stephen R. Springston, R. Subramanian and Rahul Zaveri, Oral presentation, Annual AGU meeting, San Francisco, Dec 12-17, 2010.
- Aerosol Radiative Forcing: Reducing aerosol absorption uncertainty through Photothermal Interferometry (PTI)*, **Arthur J. Sedlacek, III, Invited Talk**, Pennsylvania State University, April 17, 2008.
- Measurement of Absorption Amplification of Coated Particles using Photothermal Interferometry*, **Arthur J. Sedlacek, III** and Jeonghoon Lee, Oral presentation - Annual American Chemical Society meeting held in New Orleans, April 6-10, 2008.
- Aerosol Radiative Forcing: Reducing aerosol absorption uncertainty through Photothermal Interferometry (PTI)*, **Arthur J. Sedlacek, III, Invited Talk**, National Institute for Standards and Technology (NIST), March 21, 2008.

**WILLIAM PATRICK ARNOTT**

(patarnott@gmail.com)Tel: 775-784-6834

Professor of Physics and Atmospheric Sciences, University of Nevada Reno Fax: 775-784-1398

A. Professional Preparation:

Ph.D.	1988	Washington State University, Pullman	Physics
M.S.	1986	Washington State University, Pullman	Physics
B.S.	1984	University of Southern Colorado, Pueblo	Physics

B. Appointments:

07/11-Present Professor, Physics Department, University of Nevada Reno (UNR).

07/05-06/11 Associate Professor, Physics Department, UNR.

07/05-Present Director, Undergrad Atmospheric Sciences Program, Physics Department, UNR.

2001-Present Research Professor, Desert Research Institute, Atmospheric Sciences, Reno, NV

1996-2001 Associate Research Professor, Atmospheric Sciences Center, DRI

1995-2002 Assistant Director, Atmospheric Sciences Graduate Program, UNR

1992-1996 Assistant Research Professor, Atmospheric Sciences Center, DRI

1991-Present Adjunct Visiting Assistant Professor of Physics, University of Mississippi (UM)

1988-1991 Postdoctoral Scientist, National Center for Physical Acoustics, UM

1984-1988 Research/Teaching Assistant, Washington State University, Pullman

1984 American Physical Society Intern, Xerox Palo Alto Research Center, CA

1983 Research Assistant, University of Southern Colorado, Pueblo

C. Publications: (see [www.patarnott.com/pubs.htm](http://www.patarnott.com/pubs.htm) for further description.)(i) 2 Patents and Five Representative Publications:

**Arnott W. P.**, H. Moosmüller, and J. W. Walker, (2003) Photoacoustic instrument for measuring particles in a gas. U.S. Patent Number 6,662,627.

**Arnott, W.P.**, and H. Moosmüller (2010) Method and Apparatus for Photoacoustic Measurements. U.S. Patent Number 7,710,566.

Gyawali, M., **Arnott, W. P.**, Zaveri, R. A., Song, C., Moosmüller, H., Liu, L., Mishchenko, M. I., Chen, L.-W. A., Green, M. C., Watson, J. G., and Chow, J. C. (2012). Photoacoustic optical properties at UV, VIS, and near IR wavelengths for laboratory generated and winter time ambient urban aerosols, *Atmos. Chem. Phys.*, 12, 2587-2601.

Gyawali, M., **W. P. Arnott**, K. Lewis, and H. Moosmüller (2009). In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of absorbing and non-absorbing coatings on spectral light absorption. *Atmos. Chem. Phys.*, 9, 8007-8015.

Paredes-Miranda, G., **Arnott, W. P.**, Jimenez, J. L., Aiken, A. C., Gaffney, J. S., and Marley, N. A.: (2009) Primary and secondary contributions to aerosol light scattering and absorption in Mexico City during the MILAGRO 2006 campaign, *Atmos. Chem. Phys.* 9, 3721-3730.

**Arnott, W. P.**, H. Moosmüller, and C. F. Rogers, (1999). "Photoacoustic spectrometer for measuring light absorption by aerosols: Instrument description." *Atmos. Env.* **33**, 2845-2852.

Chakrabarty, R.K., H. Moosmüller, **W. P. Arnott**, M. A. Garro, G. Tian, J. G. Slowik, E. S. Cross, J. Han, P. Davidovits, T. B. Onasch, and D. R. Worsnop, (2009), Low Fractal Dimension Cluster-Dilute Soot Aggregates from a Premixed Flame, *Physical Review Letters* 102, 235504.

## **PETER R. BUSECK**

School of Earth & Space Exploration and Department of Chemistry & Biochemistry, Arizona State University, Tempe, AZ 85287-1404

Office: (480) 965-3945;

[pbuseck@asu.edu](mailto:pbuseck@asu.edu); <http://7starm.asu.edu>

### Professional Experience

Arizona State University, Tempe, AZ; Regents' Professor, 1989-2012; Professor, Depts. Of Geological Sciences and Chemistry/Biochemistry, 1963-1989. Mineralogy using high-resolution transmission electron microscopy; meteoritics; atmospheric chemistry and effects on air quality and climate change

Visiting Professor: Stanford University, Dept. of Geology, 1979-1980; University of Paris, Lab. Mineralogie & Cristallographie, 1986-1987; National Science Foundation, and White House Office of Science and Technology Policy (OSTP), Special Assistant to the Director, NSF, 1994-1995; Harvard University, Dept. of Earth & Planetary Sciences, 2002

Carnegie Institution of Washington, Geophysical Laboratory, Postdoctoral Fellow, 1961-1963

### Education

Columbia University, New York, NY; Ph.D., Economic Geology, 1962

Antioch College, Yellow Springs, OH; A.B., Geology, 1957

### Selected Recent Lectures

Invited Speaker: International Symposium on Polycyclic Aromatic Compounds, 2011; NIST Aerosol Metrology Climate Workshop, 2011; International Mineralogical Assoc., 2010; Goldschmidt Conf., 2010

### Selected Recent Refereed Publications: Atmospheric Chemistry

Tessendorf, S.A., et al., The Queensland seeding research program, cloud *Bull. Amer. Meteor. Soc.* **93**, 75-90, 2012.

Thompson, J.E., et al., Aerosol optical properties at Pasadena, CA during CalNex 2010. *Atmos. Environ.*, in press, available online, doi:10.1016/j.atmosenv.2012.03.01, 2012.

Adachi, K. and **P.R. Buseck**, Atmospheric tar balls from biomass burning in Mexico, *J. Geophys. Res.* **116**, D05204, doi:10.1029/2010JD015102, 2011.

Adachi, K., E.J. Freney, and **P.R. Buseck**, Shapes of internally mixed hygroscopic aerosol particles after deliquescence, and their effect on light scattering, *Geophys. Res. Lett.* **38**, L13804, doi:10.1029/2011GL047540, 2011.

Yokelson, R.J., I.R. Burling, S.P. Urbanski, E.L. Atlas, K. Adachi, **P.R. Buseck**, C. Wiedinmyer, S.K. Akagi, D.W. Toohey, and C.E. Wold Trace gas and particle emissions from open biomass burning in Mexico, *Atmos. Chem. Phys.* **11**, 6787–6808, 2011.

**Buseck, P. R.**, Atmospheric-particle research: Past, present, and future, *Elements* **6**, 208-209, 2010.

Adachi, K. and P.R. Buseck Hosted and free-floating metal-bearing atmospheric nanoparticles in Mexico City, *Environ. Sci. & Tech.* **44**, 2299-2304, 2010.

Pósfai, M., and **P.R. Buseck** Nature and climate effects of individual tropospheric aerosol particles, *Ann. Review Earth & Planet. Sci.* **38**, 17-43, 2010.

## PAUL DAVIDOVITS

### Education

B.S., Columbia University, 1960

M.S., Columbia University, 1961

Ph.D., Columbia University, 1964

### Appointments

1974 - present Boston College - Professor of Chemistry

1994 - 1998 Boston College – Department Chair

1970 - 1974 Yale University - Associate Professor of Applied Science

1965 - 1970 Yale University - Assistant Professor of Applied Science

1964 - 1965 Columbia University - Res. Assoc. in Physics and Lecturer in Engineering

### Awards:

Fellow of the American Physical Society

Fellow of the American Association for Advancement of Science

R.W. Wood Prize for Confocal Microscopy, O.S.A. 2000

Boston College Distinguished Senior Research Award, 2001

Alpha Sigma Nu Book Award, 2003

Research Experience. As part of his earlier work, Paul Davidovits developed the optically pumped rubidium maser and provided the first accurate systematic measurements of cross sections for the gas phase reactions of alkali atoms with halogen molecules. Davidovits, jointly with David Egger, developed the first operational confocal scanning laser microscope and presented the basic theories applicable to the technique. For this work he was a co-recipient in 2000 of the R.W. Wood Prize. He is a co-inventor of an isotope separation technique using seeded beams. In collaboration with the research group at Aerodyne Research, Inc., he developed techniques involving first, entrained monodisperse droplets and, subsequently, entrained bubbles to study gas-liquid interactions. The results of these studies provided basic information about the physical chemistry of gas-liquid interactions at the interface. This work has also provided key parameters needed to understand the role of cloud and aerosol chemistry in atmospheric processes such as acid rain formation, dry acid deposition and ozone depletion. More recently the Aerodyne-Boston College collaborators have developed a novel aerosol mass spectrometer for field and laboratory aerosol studies. Their current work is yielding new information about the morphology dependent chemistry of atmospheric aerosols, including black carbon aerosols. Professor Davidovits has authored or co-authored over 140 scientific publications, has written two textbooks, and has co-edited a text on the alkali-halide vapors. He holds three patents.

### Recent Project-related Publications (three of 12 such publications during the past 3 years)

Lambe, A. T., T. B. Onasch, P. Massoli, D. R. Croasdale, J. P. Wright, A. T. Ahern, L. R. Williams, D. R. Worsnop, W. H. Brune, and **P. Davidovits**, Laboratory studies of the chemical composition and cloud condensation nuclei (CCN) Activity of secondary organic aerosol (SOA) and oxygenated organic aerosol (OPOA), *Atmos. Chem. Phys.* 11, 8913-8928 [www.atmos-chem-phys-discuss.net/11/13617/2011/](http://www.atmos-chem-phys-discuss.net/11/13617/2011/) doi:10.5194/acp-11-8913-8928, 2011

Onasch, T. B., A. Trimborn, E. Fortner, J. T. Jayne, G. L. Kok, L. Williams, D. Worsnop, **P. Davidovits**, Soot Particle Aerosol Mass Spectrometer: Development, Validation, and Initial Application, In press *Aerosol Science & Technology* 2012

Saukko, E., 11 other authors, Humidity Dependent Phase State of SOA from biogenic and anthropogenic precursors, *Atmos. Chem. Phys. Discuss.* 12, 4447-4476, 2012

## JEFFREY STEVEN GAFFNEY

Department of Chemistry, SCLB Room 451  
University of Arkansas at Little Rock  
Little Rock, Phone: 501-569-8840  
Email: jsgaffney@ualr.edu

<u>Degree</u>	<u>Institution</u>	<u>Year</u>	<u>Field</u>
B.S.	University of California, Riverside	1971	Chemistry (Honors)
M.S.	University of California, Riverside	1973	Organic Chemistry
Ph.D.	University of California, Riverside	1975	Physical Organic Chemistry

### Research And Professional Experience

<u>Date</u>	<u>Position</u>
July 2006 – Present	Chair, Department of Chemistry – Professor of Chemistry University of Arkansas at Little Rock (UALR)
August 2005 – June 2006	Program Manager Global Change Research Senior Chemist, Environ. Sci. Division, ANL
April 2000-Aug. 2005	Senior Chemist, Environ. Res. Div., ANL.
March 2003 –February 2004	Chief Scientist, Atmospheric Chemistry Program, DOE/OBER Atmospheric Science Program (ASP)
March 2004 – Present	Lead Scientist, DOE/OBER Atmospheric Science Program, Megacity Aerosol Experiment – Mexico City (MAX-Mex)
December 2002-March 2003	Acting Chief Scientist, Atmospheric Chemistry Program DOE/OBER Atmospheric Science Program (ASP)
June 1998-Present	Chief Scientist, DOE/OBER Global Change Education Prog.
October 2002 -2007	Senior Fellow, Center for Environ. Sci., The Univ. of Chicago
Jan. 1989-March 2000	Chemist, Environmental Res. Div., Argonne National Laboratory
Dec. 2001-Present	Faculty Adj. Prof., Earth and Environ. Sci., U of IL, Chicago
Jan. 1992-2006	Faculty Adj. and Adj. Research Chemist, New Mexico Tech
1985-1988	Staff Scientist, INC-7, Isotope and Nuclear Chem., LANL
1980-1985	Chemist, Dept. of Applied Science BNL

### Synergistic Activities – 5 Activities

Committee Associate of the Joint Board Council Committee on Chemistry and Public Affairs, American Chemical Society (ACS CCPA) (2002-2005); Member, ACS CCPA (2006-), Chair Policy Subcommittee ACS CCPA (2007-2010); American Meteorological Society History Committee (2003-2007); Mexico City – Megacity Research Science Advisory Committee, MIT (2003-2006), American Meteorological Society Commission on Education and Human Resources - Board on Women and Minorities (2006-2012).

### Professional Affiliations

American Chemical Society (Geochemistry and Environmental Chemistry Divisions); American Geophysical Union; American Meteorological Society

### Publications And Presentations:

Dr. Gaffney has authored or co-authored over 200 peer-reviewed journal articles, book chapters, and technical reports and has presented over 400 talks and papers at scientific meetings and public activities.

## **ERNIE R. LEWIS**

Atmospheric Sciences Division  
Brookhaven National Laboratory  
Brookhaven National Laboratory

[elewis@bnl.gov](mailto:elewis@bnl.gov)

631-344-7406

### Current Activities

Principal Investigator (with Warren Wiscombe, NASA) of MAGIC (Marine ARM GPCI Investigation of Clouds), a one-year deployment of the Second ARM Mobile Facility (AMF2) on Horizon Lines cargo container ships *Reliance* & *Spirit* traversing a route between Los Angeles, CA and Honolulu, HI.

### Education

A. B. D., Theoretical Physics, *The University of Texas at Austin*, 1982-1990.

Diploma, *von Karman Institute for Fluid Dynamics, Brussels, Belgium*, 1981-1982.

*University of Washington, Seattle*, 1980-1981.

B. S. (Honors), *California Institute of Technology, Pasadena, California*, 1975-1979.

### Employment History

1998-present: Atmospheric Sciences Division, *Brookhaven National Laboratory, Upton, NY*

1994-1998: Department of Oceanography, *Brookhaven National Laboratory, Upton, NY*

### Selected Publications

Sedlacek, A. J., III, **E. R. Lewis**, L. Kleinman, J. Xu, & Q. Zhang, "Determination of and evidence for non-core-shell structure of particles containing black carbon using the single particle soot photometer (SP2)," *Geophys. Res. Letts.*, 2012. doi:10.1029/2012GL050905

de Leeuw, G., E. L. Andreas, M. D. Anguelova, C. W. Fairall, **E. R. Lewis**, C. O'Dowd, M. Schulz, & S. E. Schwartz, "Production flux of sea-spray aerosol," *Rev. Geophys.*, 49, RG2001, 2011. doi:10.1029/2010RG000349

McGraw, R. & **E. R. Lewis**, "Deliquescence and efflorescence of small particles," *J. Chem. Phys.*, 131: 194705, 2009. doi:10.1063/1.3251056

Chang, L.-S., S. E. Schwartz, R. McGraw, & **E. R. Lewis**, "Sensitivity of aerosol properties to new particle formation mechanism and to primary emissions in a continental-scale chemical transport model," *J. Geophys. Res. – Atmos.*, 114: D07203, 2009. doi:10.1029/2008JD011019

McComiskey, A., S. E. Schwartz, B. Schmid, H. Guan, **E. R. Lewis**, P. Ricchiuzzi, & J. A. Ogren, "Direct aerosol forcing: Calculation from observables and sensitivities to inputs," *J. Geophys. Res. – Atmos.*, 113, D09202, 2008. doi:10.1029/2007JD009170

Lewis, E. R. & S. E. Schwartz, Sea Salt Aerosol Production: Mechanisms, Methods, Measurements, and Models, Geophysical Monograph Series v. 152, American Geophysical Union, Washington, DC, 2004. 413 pp. Available for purchase at <https://www.agu.org/cgi-bin/agubookstore?book=ASGM1524173>

## WUYIN LIN

Environmental Sciences Department  
Brookhaven National Laboratory  
Tel: 631-3447249  
Email: wlin@bnl.gov.

### Education

Stony Brook University, NY, Ph. D, 2002, Atmospheric Sciences

### Research and Professional Experiences

Associate Scientist: 10/2011 – present, Brookhaven National Laboratory  
Assistant Scientist: 09/2009 – 09/2011, Brookhaven National Laboratory  
Research Scientist: 07/2006 – 09/2009, Stony Brook University  
Postdoc Research Associate: 08/2002 – 06/2006, Stony Brook University

### Selected Publications

- Liu, H., M. Zhang, and **W. Lin**, 2012: An investigation of the initial development of the double ITCZ warm biases in the CCSM. *J. Climate*, **25**, 140–155.
- Liu, H., X. Liu, M. Zhang, and **W. Lin**, 2011: A critical evaluation of the upper ocean heat budget in the Climate Forecast System reanalysis data for the south central equatorial Pacific. *Environ. Res. Lett.*. doi:10.1088/1748-9326/6/3/034022.
- Liu, Hailong, **Wuyin Lin**, and Minghua Zhang, 2010: Heat budget of the upper ocean in the south central equatorial Pacific, *J. Climate*, **23**, 1779-1792.
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- Lin, W.**, and Zhang, M., 2004: Evaluation of clouds and their radiative effects simulated by the NCAR Community Atmospheric Model CAM2 against satellite observations, *J. Climate*, **17**, 3302-3318.

## HANS MOOSMÜLLER

2000 – Present: Research Professor, Atmospheric Sciences Division, Desert Research Institute

Ph.D. Physics 1988 Colorado State University, Fort Collins, CO

DIPLOM (M.S.) Physics 1982 Ludwigs-Maximilians Universität, München

Research Experience: Dr. Moosmüller's interests include experimental and theoretical research in optical spectroscopy and its applications to atmospheric, aerosol, and climate physics. His research focuses on development and application of real time, *in situ* measurement methods for aerosol light absorption, scattering, and extinction, and new optical remote sensing techniques. These measurement methods are being used for ambient air monitoring and vehicle, fugitive dust, and biomass burning emission studies. His latest research interests are fast, ultra-sensitive measurements of elementary mercury concentrations and fluxes and aerosol morphology and its influence on aerosol optical properties with a focus on fractal-like chain aggregates found in combustion particles.

Editor: 2010 special issue of "Atmospheric Chemistry and Physics" entitled "Measurement and Modeling of Aerosol Emissions from Biomass Burning"

### Relevant Peer Reviewed Publications

Chakrabarty, R. K., M. A. Garro, E. M. Wilcox, and **H. Moosmüller** (2012). Strong Radiative Heating due to Wintertime Black Carbon Aerosols in the Brahmaputra River Valley. *Geophys. Res. Lett.*, in press.

**Moosmüller, H.** and R. K. Chakrabarty (2011). Technical Note: Simple Analytical Relationships between Ångström Coefficients of Aerosol Extinction, Scattering, Absorption, and Single Scattering Albedo. *Atm. Chem. Phys.*, **11**, 10677-10680.

Watson, J. G., J. C. Chow, L.-W. A. Chen, D. H. Lowenthal, E. M. Fujita, H. D. Kuhns, D. A. Sodeman, D. E. Campbell, **H. Moosmüller**, D. Zhu, and N. Motallebi (2011). Particulate Emission Factors for Mobile Fossil Fuel and Biomass Combustion Sources. *Sci. Total Environ.*, **409**, 2384-2396.

**Moosmüller, H.**, R. K. Chakrabarty, K. M. Ehlers, and W. P. Arnott (2011). Absorption Ångström Coefficient, Brown Carbon, and Aerosols: Basic Concepts, Bulk Matter, and Spherical Particles. *Atm. Chem. Phys.*, **11**, 1217-1225.

Mack, L. A., E. J. T. Levin, S. M. Kreidenweis, D. Obrist, **H. Moosmüller**, K. A. Lewis, W. P. Arnott, G. R. McMeeking, A. P. Sullivan, C. E. Wold, W. M. Hao, J. L. Collett, Jr., and W. C. Malm (2010). Optical Closure Experiments for Biomass Smoke Aerosols. *Atm. Chem. Phys.*, **10**, 9017-9026.

Levin, E. J. T., G. R. McMeeking, C. M. Carrico, L. E. Mack, S. M. Kreidenweis, C. E. Wold, **H. Moosmüller**, W. P. Arnott, W. M. Hao, J. L. Collett, Jr., and W. C. Malm (2010). Biomass Burning Smoke Aerosol Properties Measured during FLAME. *J. Geophys. Res.*, **115**, doi:10.1029/2009JD013601.

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Lewis, K. A., W. P. Arnott, **H. Moosmüller**, R. K. Chakrabarty, C. M. Carrico, S. M. Kreidenweis, D. E. Day, W. C. Malm, A. Laskin, J. L. Jimenez, I. M. Ulbrich, J. A. Huffman, T. B. Onasch, A. Trimborn, L. Lui, and M. I. Mishchenko (2009). Reduction in Biomass Burning Aerosol Light Absorption upon Humidification: Roles of Inorganically-Induced Hygroscopicity, Particle Collapse, and Photoacoustic Heat and Mass Transfer. *Atm. Chem. Phys.*, **9**, 8949-8966.

## TIMOTHY B. ONASCH

Center for Aerosol & Cloud Chemistry,  
Aerodyne Research, Inc., Billerica, MA  
Tel: Office, (978) 663-9500; [onasch@aerodyne.com](mailto:onasch@aerodyne.com)

### Professional Experience.

2006-present Aerodyne Research, Inc., Principal Research Scientist  
2006-present Boston College, Chemistry Department, Research Associate Professor  
2003-2006 Aerodyne Research, Inc., Senior Research Scientist  
2002-2003 Aerodyne Research, Inc., Research Scientist

### Education.

1999-2002 Alexander Holländer Postdoctoral Fellow, Brookhaven National Laboratory, NY  
1999 University of Colorado at Boulder (Physical Chemistry), Ph.D.  
1993 Carleton College (Chemistry), B.A.

### Selected Recent Honors/Service/Funding/Experience.

Co-organizer and facilitator of the 4th-10th annual Aerodyne Aerosol Mass Spectrometer Users' Meetings, 2003-2009; Member of Ph.D. thesis defense committees at Boston College, 2006 and 2008.

### Selected Recent Refereed Publications.

Cross, E. S., **T. B. Onasch**, A. Ahern, W. Wrobel, J. G. Slowik, J. Olfert, D. A. Lack, et al. (2010) "Soot Particle Studies-Instrument Inter-Comparison-Project Overview." Aerosol Sci. Tech. 44 (8): 592-611.

Jimenez, J. L., M. R. Canagaratna, et al. (2009). "Evolution of Organic Aerosols in the Atmosphere." Science 326(5959): 1525.

Lack, D. A., C. D. Cappa, et al. (2009). "Absorption Enhancement of Coated Absorbing Aerosols: Validation of the Photo-Acoustic Technique for Measuring the Enhancement." Aerosol Sci. Tech. 43(10): 1006-1012.

Lack, D. A., J. J. Corbett, et al. (2009). "Particulate emissions from commercial shipping: Chemical, physical, and optical properties." J. of Geophys. Res. 114: D00F04.

Lambe, A. T., A. T. Ahern, et al. (2011) "Characterization of aerosol photooxidation flow reactors: heterogeneous oxidation, secondary organic aerosol formation and cloud condensation nuclei activity measurements." Atmos. Meas. Tech. 4 (3): 445-461.

Lambe, A. T., **T. B. Onasch**, et al. (2011) "Laboratory studies of the chemical composition and cloud condensation nuclei (CCN) activity of secondary organic aerosol (SOA) and oxidized primary organic aerosol (OPOA)." Atmos. Chem. Phys. 11 (17): 8913-8928.

Lee, T., A. P. Sullivan, L. Mack, J. L. Jimenez, S. M. Kreidenweis, **T. B. Onasch**, D. R. Worsnop, et al. (2010). "Chemical Smoke Marker Emissions During Flaming and Smoldering Phases of Laboratory Open Burning of Wildland Fuels." Aerosol Sci. Tech. 44

Massoli, P., P. L. Keabian, et al. (2010) "Aerosol Light Extinction Measurements by Cavity Attenuated Phase Shift (CAPS) Spectroscopy: Laboratory Validation and Field Deployment of a Compact Aerosol Particle Extinction Monitor." Aerosol Sci. Tech. 44 (6): 428-435.

**Onasch, T. B.**, J. T. Jayne, et al. (2009). "Chemical Properties of Aircraft Engine Particulate Exhaust Emissions." J. Propul. Power 25 (5): 1121-1137.

**Onasch, T. B.**, A. Trimborn, E. C. Fortner, J. T. Jayne, G. L. Kok, L. R. Williams, P. Davidovits, and D. R. Worsnop (2012) "Soot Particle Aerosol Mass Spectrometer: Development, Validation, and Initial Application." Aerosol Sci. Tech. 46 (7): 804-817.

## JOHN E. SHILLING

Pacific Northwest National Lab  
Atmospheric Sciences and Global Change Division  
902 Battelle Blvd., Richland, WA 99354  
phone: (509) 375-6874; fax: (509) 372-6168  
[john.shilling@pnl.gov](mailto:john.shilling@pnl.gov)

Research Interests: Secondary Organic Aerosol (SOA), Heterogeneous Kinetics and Chemistry, Phase Transitions, Mass Spectrometry, Field Campaigns

### Education And Research Experience

Pacific Northwest National Laboratory, *2008-present*, Research Scientist, Atmospheric Sciences and Global Change Division, Richland, WA *2008-present*

Research Areas: Secondary Organic Aerosol (SOA), aerosol aging, CCN activity, aerosol mass spectrometry (AMS), Proton Transfer Reaction Mass Spectrometry (PTR-MS), field campaigns

Harvard University, *2005-2008*, Postdoctoral Associate, School of Engineering and Applied Sciences, Cambridge, MA

Research Areas: SOA, aerosol aging, CCN activity, aerosol mass spectrometry

Advisor: Prof. Scot T. Martin

University of Colorado, *1999 – 2005*, Department of Chemistry and Biochemistry, Boulder  
Ph.D. in Analytical/Atmospheric Chemistry

Research Areas: heterogeneous kinetics, cubic ice, nucleation, phase transitions

Advisor: Prof. Margaret Tolbert

Ursinus College, *1995-1999*, Department of Chemistry, Collegeville, PA  
B.S. in Chemistry (Honors, ACS certified)

Research Areas: NMR spectroscopy, optically polarized  $^{129}\text{Xe}$

Advisor: Prof. Holly Gaede

### Selected Publications

Hiranuma, N.; Kohn, M.; Pekour, M.S.; Nelson, D.A.; **Shilling, J.E.**; Cziczo, D.J.; Droplet activation, separation, and compositional analysis: laboratory studies and atmospheric measurements. *Atmos. Meas. Tech.*, **4**, 2333-2343, 2011.

**Shilling, J.E.**; Chen, Q.; King, S.M.; Rosenoern, T.; Kroll, J.H.; Worsnop, D.R.; DeCarlo, P.F.; Aiken, A.C.; Sueper, D.; Jimenez, J.L.; Martin, S.T. Loading-dependent elemental composition  $\alpha$ -pinene SOA particles. *Atmos. Chem. Phys.*, **9**, 771-782, 2009

**Shilling, J.E.**; Chen, Q.; King, S.M.; Rosenoern, T.; Kroll, J.H.; Worsnop, D.R.; McKinney, K.A.; Martin, S.T. Particle mass yield in secondary organic aerosol formed by the dark ozonolysis of  $\alpha$ -pinene. *Atmos. Chem. Phys.*, **8**, 20736-2088, 2008

**Shilling, J.E.**; King, S.M.; Mochida, M.; Worsnop, D.R.; Martin, S.T. Mass spectral evidence that small changes in chemical composition caused by oxidative aging processes alter aerosol CCN properties, *J. Phys. Chem. A.*, **111**, p. 3358-3368, 2007.

**Shilling, J.E.**; Tolbert, M.A.; Toon, O.B.; Jensen, E.J.; Murray, B.J.; Bertram, A.K. Measurements of the vapor pressure of cubic ice and their implications for atmospheric ice clouds, *Geophys. Res. Lett.*, **33**, Article L17801, 2006.

## **JIAN WANG**

Brookhaven National Laboratory

631-344-7920

[Jian@bnl.gov](mailto:Jian@bnl.gov)

### Education and Training

Ph.D. in Chemical Engineering, California Institute of Technology	June 2002
M.S. in Chemical Engineering, California Institute of Technology	June 1998
B.S. in Physical Chemistry, University of Science and Technology of China	June 1996

### Professional Experience

2008-present, Scientist, Atmospheric Science Division, Brookhaven National Laboratory  
2005-2008, Associate Scientist, Atmospheric Science Division, Brookhaven National Laboratory  
2002-2005, Goldhaber Distinguished Fellow, Brookhaven National Laboratory

### Selected Publications

Liu X. H. and **J. Wang** 2011, “How important is organic aerosol hygroscopicity to aerosol indirect forcing?”, *Environmental Research Letters*, **5**, 044010, doi: 10.1088/1748-9326/5/4/044010.

**Wang, J.**, M.J. Cubison, A.C. Aiken, J.L. Jimenez, and D.R. Collins, 2010, “The importance of aerosol mixing state and size-resolved composition on CCN concentration and the variation of the importance with atmospheric aging of aerosols”, *Atmos. Chem. Phys.*, submitted.

**Wang J.** 2009, “A fast integrated mobility spectrometer with wide dynamic size range: Theoretical analysis and numerical simulation”, *J. Aerosol Sci.*, **40**, 890-906, doi:10.1016/j.jaerosci.2009.06.005.

**Wang, J.**, P. H. Daum, S. S. Yum, Y. G. Liu, G. Senum, L. Lu, J. H. Seinfeld, and H. Jonnsson, 2009, “Observations marine stratocumulus microphysics and implications for processes controlling droplet spectra: results from the Marine Stratus/stratocumulus Experiment (MASE)”, *J. Geophys. Res.*, **114**, D18210, doi: 10.1029/2008JD011035.

**Wang, J.**, Y. N. Lee, P. H. Daum, J. Jayne, and M. L. Alexander 2008, “Effects of aerosol organics on cloud condensation nucleus (CCN) concentration and first indirect aerosol effect”, *Atmos. Chem. Phys.*, **8**, 6325-6339.

Kleinman, L. I., S. R. Springston, P. H. Daum, Y.-N. Lee, L. J. Nunnermacker, G. I. Senum, **J. Wang**, J. Weinstein-Lloyd, M. L. Alexander, J. Hubbe, J. Ortega, M. R. Canagaratna, and J. Jayne, 2008, “The time evolution of aerosol composition over the Mexico City plateau”, *Atmos. Chem. Phys.*, **8**, 1559-1575.

**Wang, J.** 2007, “Effects of spatial and temporal variations in aerosol properties on mean cloud albedo”, *J. Geophys. Res.*, **112**, D16201, doi:10.1029/2007JD008565.

Kulkarni P. and **J. Wang**, 2006, “New Fast Integrated Mobility Spectrometer for Real-Time Measurement of Aerosol Size Distribution: II. Design, Calibration, and Performance Characterization”, *J. Aerosol Sci.*, **37**(10), pp 1326-1339.

Kulkarni P. and **J. Wang**, 2006, “New Fast Integrated Mobility Spectrometer for Real-Time Measurement of Aerosol Size Distribution: I. Concept and Theory”, *J. Aerosol Sci.*, **37**(10), pp1303-1325.

## **ROBERT J. YOKELSON**

Dept. of Chemistry, University of Montana, Missoula, MT 59812

Tel: Office, (406) 243-6088; Cell: (406) 546-7881

[bob.yokelson@umontana.edu](mailto:bob.yokelson@umontana.edu);

<http://www.cas.umt.edu/chemistry/facultyDetails.cfm?id=1210>

### Professional Experience.

University of Montana, Missoula, MT; Research Professor, Department of Chemistry. 1993-2012. Air, ground, and lab measurements of fire, ship, and biogenic emissions. Modeling fire emissions.

National Oceanic and Atmospheric Administration, Boulder, CO; Postdoctoral Associate, 1991-1993. Measurements of photochemistry and kinetics relevant to stratospheric ozone depletion.

### Education.

Yale University, New Haven, CT; Ph.D., Chemical Physics. 1986-1991

University of Montana, Missoula, MT; B.S., Chemistry. 1986

### Selected Recent Honors/Service/Funding/Experience.

NASA Leadership Team: SEAC4RS 2011-14. Convener of session on biomass burning; Dec, 2010 AGU. Invited Speaker; Gordon Conference 2009. Session chair; GEOS-CHEM meeting 2009. Current external funding: \$2,368,981. 4 seasons as USFS Wildland Fire Fighter (up to "Hot Shot" level).

### Selected Recent Refereed Publications: Fire Emissions.

H-index: 25 (ResearcherID: C-9971-2011). In progress papers include: Yellowstone emissions, synthesis of lab and field data, photochemical modeling of plume evolution, SP2 measurements of black carbon emissions from fires, multiple papers on 2011 field campaign, etc.)

Akagi, S. K., Craven, J. S., Taylor, J. W., McMeeking, G. R., **Yokelson, R. J.**, Burling, I. R., Urbanski, S. P., Wold, C. E., Seinfeld, J. H., Coe, H., Alvarado, M. J., and Weise, D. R.: Evolution of trace gases and particles emitted by a chaparral fire in California, *Atmos. Chem. Phys.*, 12, 1397-1421, doi:10.5194/acp-12-1397-2012, 2012.

Burling, I. R., **Yokelson, R. J.**, Akagi, S. K., Urbanski, S. P., Wold, C. E., Griffith, D. W. T., Johnson, T. J., Reardon, J., and Weise, D. R.: Airborne and ground-based measurements of the trace gases and particles emitted by prescribed fires in the United States, *Atmos. Chem. Phys.*, 11, 12197-12216, doi:10.5194/acp-11-12197-2011, 2011.

Simpson, I. J., with 17 authors.: Boreal forest fire emissions in fresh Canadian smoke plumes: C1-C10 volatile organic compounds (VOCs), CO<sub>2</sub>, CO, NO<sub>2</sub>, NO, HCN and CH<sub>3</sub>CN, *Atmos. Chem. Phys.*, 11, 6445-6463, doi:10.5194/acp-11-6445-2011, 2011.

Roberts, J. M., Veres, P. R., Cochran, A. K., Warneke, C., Burling, I. R., **Yokelson, R. J.**, Lerner, B., Holloway, J. S., Fall, R., and de Gouw, J.: Isocyanic acid in the atmosphere: Sources, concentrations and sinks, and potential health effects, *PNAS*, 108, 8966-8971, doi: 10.1073/pnas.1103352108, 2011.

## **RAHUL A. ZAVERI**

Atmospheric Sciences & Global Change Division, Pacific Northwest National Laboratory,  
Richland, WA 99352

Tel: Office, (206) 528-3215;

[rahul.zaveri@pnnl.gov](mailto:rahul.zaveri@pnnl.gov);

[www.pnl.gov/science/staff/staff\\_info.asp?staff\\_num=5722](http://www.pnl.gov/science/staff/staff_info.asp?staff_num=5722)

### Professional Experience.

Pacific Northwest National Laboratory, Richland, WA; Senior Research Scientist, 1998-Present  
Secondary organic and inorganic aerosol formation; aerosol thermodynamics; gas-particle interactions and heterogeneous chemistry; aerosol optical and CCN activation properties; Lagrangian and Eulerian modeling of the transport and chemistry; integrated field, laboratory, and modeling studies on atmospheric aerosol formation and evolution.

### Education.

Virginia Tech, Blacksburg, VA; Ph.D., Chemical Engineering, 1993-1997

University of Kentucky, Lexington, KY; M.S., Chemical Engineering, 1991-1993

University of Bombay, Bombay, India; B.S., Chemical Engineering, 1987-1991

### Selected Recent Honors/Service/Funding/Experience.

Co-convenor of session on Climate Change, Air Quality, and Their Interrelations at the North American West Coast; Dec. 2010 AGU; Co-organizer of special symposium on Recent Field Studies in California and Mexico (CalNex, CARES, Cal-Mex) Oct. 2011, AAAR; Co-organizer of the International Aerosol Modeling Algorithms (IAMA) Conference, University of California, Davis, Nov 30- Dec 2, 2011; Lead PI of the DOE 2010 Carbonaceous Aerosols and Radiative Effects Study (CARES) field campaign.

### Selected Refereed Publications

Gyawali, M., Arnott, W. P., **Zaveri, R. A.**, Song, C., Moosmüller, H., Liu, L., Mishchenko, M. I., Chen, L.-W. A., Green, M. C., Watson, J. G., and Chow, J. C.: Photoacoustic optical properties at UV, VIS, and near IR wavelengths for laboratory generated and winter time ambient urban aerosols, *Atmos. Chem. Phys.*, 12, 2587–2601, doi:10.5194/acp-12-2587-2012, 2012.

Song, C., **R. A. Zaveri**, J. E. Shilling, M. L. Alexander, M. Newburn (2011), Effect of hydrophilic organic seed aerosols on secondary organic aerosol formation from ozonolysis of *a*-pinene, *Environ. Sci. Technol.*, 45 (17), 7323–7329.

**Zaveri, R.A.**, J.C. Barnard, R.C. Easter, N. Riemer, M. West (2010), Particle-resolved simulation of aerosol size, composition, mixing state, and the associated optical and cloud condensation nuclei activation properties in an evolving urban plume, *J. Geophysical Research*, 115, D17210, doi:10.1029/2009JD013616.

Vaden, T.D., C. Song, **R.A. Zaveri**, D. Imre, A. Zelenyuk (2010), Morphology of mixed primary and secondary organic particles and the adsorption of spectator organic gases during aerosol formation, *Proc. Nat. Acad. Sci. U.S.A.*, 107(15), 6658-6663, doi:10.1073/pnas.0911206107.

**Zaveri, R.A.**, 14 authors (2010), Nighttime chemical evolution of aerosol and trace gases in a power plant plume: Implications for secondary organic nitrate and organosulfate aerosol formation, NO<sub>3</sub> radical chemistry, and N<sub>2</sub>O<sub>5</sub> heterogeneous hydrolysis, *J. Geophysical Research*, 115, D12304, doi:10.1029/2009JD013250.

## Description of Affiliated Institutions

### Aerodyne Research, Inc. (ARI)

ARI provides research and development services and advanced sensor and software products to industrial, academic and government customers addressing national and international challenges, including: monitoring and enhancing regional and global environmental quality. ARI scientists and engineers perform fundamental laboratory and theoretical studies; build, run and analyze complex computer models and system simulations; design and execute mobile and fixed-site field measurements; and develop, demonstrate, and deploy advanced measurement instruments and systems.

### Arizona State University

Prof. Buseck's group uses a range of electron-beam instruments, predominately transmission electron microscopes (TEMs) for the chemical and structural analysis of individual aerosol particles. These facilities enable the study of the chemistry and structures of fine-grained aerosol materials, their origin, and how they react to form other compounds. Main analytical techniques used to characterize aerosol particles are X-ray emission spectroscopy by energy analysis (EDS), electron energy-loss spectroscopy (EELS), selected-area electron diffraction (SAED), and high-resolution imaging (HRTEM).

### Boston College

Prof. Davidovits' laboratory is focused on the study of the chemical and optical properties of atmospheric aerosols including soot aerosols. A central component of his research is a particle generation system (flame-generated soot and atomizers) that is coupled to an elaborate particle coating/denuding chamber that enables various particle configurations to be studied in a laboratory setting.

### Desert Research Institute

Research in the Division of Atmospheric Sciences of DRI is focuses on field and laboratory observations, theoretical and laboratory analysis, and computer modeling at many spatial and temporal scales. DAS is home to several major programs, including the [Western Regional Climate Center](#), the [Nevada State Cloud Seeding Program](#), and the [Program for Climate, Ecosystem and Fire Applications](#). DAS also supports various state-of-the-art laboratories including the high-elevation [Storm Peak Laboratory](#).

### Pacific Northwest National Laboratory (PNNL)

PNNL is one among ten U.S. Department of Energy (DOE) national laboratories managed by DOE's Office of Science focusing on research needs within DOE as well as for the U.S. Department of Homeland Security, the National Nuclear Security Administration, other government agencies, universities and industry.

### University of Arkansas

Prof. Gaffney group conducts research in Atmospheric and Environmental Chemistry (Air and Water) with a focused on the role that atmospheric aerosols play in climate - particularly the role of carbonaceous aerosols and black carbon or soot. In addition to light absorption and scattering instrumentation, Prof. Gaffney also uses natural radionuclides to examine the sources and residence times of submicron aerosols.

### University of Montana

Dr. Yokelson group is internationally recognized for their work to elucidate the initial characteristics and subsequent transformations of biomass burning. To better characterize biomass-burning emissions, and their transformations, Dr. Yokelson's group has developed an open-path FTIR (OP-FTIR) system for ground-based and laboratory measurements and closed-cell FTIR systems for airborne measurements (AFTIR) and highly mobile ground-based measurements that capitalizes on the ability of Fourier transform infrared spectroscopy (FTIR) to measure a wide variety of reactive and stable compounds in real time.

### University of Nevada

Prof. Arnott has developed and deployed photoacoustic instruments for measurement of black carbon emission from vehicles in source sampling, and in ambient air quality studies in an effort to establish a detailed knowledge of the conditions giving rise to most of the black carbon and particulate emission to the atmosphere, and their environmental impacts. His photoacoustic spectrometer design as has been commercialized by Droplet Measurement Technologies.

## 9. ARM Resources Required

i) 120 G-1 flight hours are requested and will be divided between Pasco and Little Rock (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 is based on the following projection: 65 flight hours for June-September deployment at Pasco; 35 flight hours for the IOP based out of Little Rock, AR, in October; 15 flight hours to ferry the G1 to and from Little Rock in October; 5 flight hours for pre-campaign installation and testing in June. These estimates represent an upper limit.

ii) Specialized instruments not provided by AAF described in sec. 4.4, that are mission critical in addressing one or more of the scientific objectives listed in Sec.4.5. Equipment requested include (i) an aerosol sampler with offline electron microscopy done 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.

iii) Internet connectivity to allow timely remote access of data (including calibrations) collected on the G-1 after a flight.

iv) Instrument mentors or scientists cross-trained on other equipment to maintain instruments between flights and to operate instruments in-flight.

v) A dilution system maybe 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.

vi) 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 DOE G-1**

Activity	June	July	August	September	October
Instrument Staging & G-1 Test Flights	5-hrs				
Deployment at Pasco (WA)		15-hrs	35-hrs	15-hrs	
3-week IOP at Little Rock (AR)					35-hrs
Round trip Ferry Little Rock to-Pasco					15-hrs

**Table 2: Instrument Resource Request**

<b>Instrument</b>	<b>Measurement</b>
3- $\lambda$ particle soot absorption photometer (PSAP)	Aerosol light abs. @ 450, 550 and 700 nm.
3- $\lambda$ nephelometer	Aerosol light scatt., 450 nm, 550 nm and 700 nm
Cloud Condensation Nuclei counter (Dual-column preferred)	CCN conc @ 2 SS (0.25% and 0.50%)
PTRMS	Trace VOC detection
Trace gas suite	NO, NO <sub>2</sub> , NO <sub>x</sub> , CO, SO <sub>2</sub> , O <sub>3</sub>
Meteorology	Wind direction; wind speed; air temperature; RH and rain fall
Single particle soot photometer (SP2)	BC loading, size distr. mixing-state
UHSAS	Particle size distribution
PCSAP	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 & coating composition (Aerodyne)
Electron microscopy	2-D/3-D Particle morphology & mixing state (Arizona State University)
Photoacoustic Spectrometer (PAS)	Aerosol light absorption @ 405 nm, 870 nm (U. of Nevada)

## **10. Acknowledgements**

We thank Alice Cialella of BNL and Christine Wiedinmyer for providing MODIS fire counts and biomass burn emissions that were used to evaluate experimental sites.

## 11 Letters of Collaboration



University of Nevada, Reno

To Whom it May Concern:

18 April 2012

The purpose of this letter is to enthusiastically confirm our participation in the research proposal with Brookhaven National Laboratory Principal Investigators Larry Kleinman and Arthur J. Sedlacek. Measurements from the G-1 aircraft in biomass burn plumes will be used to quantify the time evolution of microphysical, morphological, chemical, hygroscopic, and optical properties of aerosols generated by biomass burning from close to the time of formation to less than one day atmospheric residence time; to use time sequences of observations to constrain processes and parameterizations in a Lagrangian model of aerosol evolution; and to 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.

Our participation will be to provide multispectral measurements of aerosol light absorption and scattering using our photoacoustic instruments. We will also consult with the principal investigators concerning experiment design and interpretation of results.

Sincerely,

A handwritten signature in black ink, appearing to read 'W. Patrick Arnott'.

W. Patrick Arnott  
Professor of Physics and Atmospheric Science  
University of Nevada Reno

April 10, 2012

Arthur Sedlacek, III  
Atmospheric Sciences Division  
Brookhaven National Lab  
Building 815E  
Upton NY 11973-5000

Dear Art:

I am pleased to participate as a Co-Investigator on your proposal "Aerosol Properties Downwind of Biomass Burn" that you are submitting to the Department of Energy. The proposed ARM aircraft-based field campaign provides a wonderful opportunity to resolve some of the uncertainties related to the physical and chemical properties of particles generated through biomass burning. We propose to use transmission electron microscopy (TEM) on samples collected during the campaign. Such measurements provide unique, exquisitely detailed information about the individual aerosol particles and will provide valuable complementary information to the on-line and remote measurements of other groups. We will determine morphological, chemical, hygroscopic, and perhaps optical properties of aerosols generated by biomass burning.

We will supply 3-stage impactor samplers (MPS-3, California Measurements, Inc.) to collect samples for TEM analysis. An interesting outcome of the project is that it should bring more clarity to the relationship between black carbon (BC), which is used for most if not all global climate models, and soot, a well-known product in the combustion community.

Sincerely,



Peter R. Buseck  
Regents' Professor  
<http://7starM.asu.edu>



**BOSTON COLLEGE**  
CHESTNUT HILL, MASSACHUSETTS 02467

Paul Davidovits  
Merkert Chemistry Center  
Department of Chemistry  
(617) 552-3617  
FAX (617)-552-2705  
e-mail [paul.davidovits@bc.edu](mailto:paul.davidovits@bc.edu)

April 12, 2012

Dr. Arthur Sedlacek, III and Dr. Larry Kleinman  
Atmospheric Sciences Division,  
Brookhaven National Lab,  
Upton NY 11973-5000

**Re:** Study of Aerosol Produced in Biomass Burning.

Dear Drs Sedlacek and Kleinman,

I am pleased to support and participate in your proposal to ARM to study biomass burn aerosol. Our aerosol laboratory will host a series of experiments complementing your field campaign. The Boston College laboratory is equipped with a well-characterized soot apparatus consisting of a stable burner that produces, under controlled conditions, soot particles, including particles that simulate those produced in biomass burning. The burner together with associated instrumentation produces particles of known size, black carbon content and morphology. The particles are size selected to yield a monodisperse distribution in the range 30 nm to 500 nm with width  $\pm 10\%$ . Using the existing laboratory set-up, other aerosols including "brown carbon" aerosol can be generated using various techniques including atomizers, nebulizers, condensing reservoirs, and oxidizing flow tubes. The particles can be coated with a variety of atmospherically relevant coatings of controlled composition and thickness. The particles can also be denuded as required by the experimental protocol.

*Paul Davidovits*  
Paul Davidovits  
Professor of Chemistry



Jeffrey S. Gaffney, Ph.D.  
SCLB Room 451  
Office: 501-569-8840  
Fax: 501-569-8838  
Email: [jsgaffney@ualr.edu](mailto:jsgaffney@ualr.edu)

April 26, 2012

Arthur Sedlacek, III Ph.D.  
Atmospheric Sciences Div.  
Brookhaven National Lab  
Building 815E  
Upton NY 11973-5000

Dear Art:

This letter confirms my very strong interest and willingness to collaborate on the proposed future ARM field project entitled: "Aerosol Properties Downwind of Biomass Burns".

As you know, Dr. Nancy Marley and I have been involved for a number of years examining the importance of carbonaceous aerosols particularly with regard to their sources and optical radiative properties. We have taken data here in Arkansas that clearly shows that biomass burning is a very important source of carbonaceous aerosols that have strong light absorption ranging from the UVB into the infrared.

We look forward to collaborative research with you and others from ASR and ARM in working on this project and hope to follow up with conversations we have had regarding improving PSAP data by use of quartz filters and integrating sphere spectroscopy as well as examining the use of XRF low angle fluorescence spectroscopy of these same PSAP media to look at inorganic tracers indicating biomass burning (e.g. halides and potassium).

I am looking forward to hearing that this proposed project is approved and further conversations regarding the project. Please feel free to contact me at 501-569-8840 or by email at [jsgaffney@ualr.edu](mailto:jsgaffney@ualr.edu), if I can be of any further assistance.

Sincerely yours,

A handwritten signature in black ink, appearing to read 'Jeffrey S. Gaffney'.

Jeffrey S. Gaffney, Ph.D.  
Professor and Chair  
Dept. of Chemistry

## ***AERODYNE RESEARCH, Inc.***

45 Manning Road  
Billerica, Massachusetts 01821-3976  
(978) 663-9500 Fax (978) 663-4918

April 11, 2012

Drs. Larry Kleinman and Arthur J. Sedlacek  
Environmental Sciences Department  
Brookhaven National Laboratory  
Mail Stop: 815E  
Upton, NY 11973-5000

Dear Drs. Kleinman and Sedlacek,

Aerodyne Research, Inc. (ARI) is pleased to commit to participating in the biomass burning studies as part of the DOE ASR proposal entitled, "Aerosol Properties Downwind of Biomass Burns". Our task in this project will be to deploy our Soot Particle Aerosol Mass Spectrometer (SP-AMS) on the G1 aircraft to characterize the mass, chemistry, and size of black carbon-containing particles (laser vaporizer) and nonrefractory biomass burning particles (heated tungsten vaporizer) as they are generated in biomass burning events and as they evolve in the atmosphere. Our results will help formulate parameterizations of these emissions for inclusion in Lagrangian and radiative transfer models for assessment of biomass burning impacts on climate.

Sincerely,



Charles Kolb  
President



Department of Chemistry  
The University of Montana  
Missoula, Montana 59812-1656  
Phone: (406) 243-4022  
FAX: (406) 243-4227  
E-mail: chem@selway.umt.edu

Dr. Robert J. Yokelson  
Department of Chemistry, University of Montana, Missoula, MT 59812  
Tel: Office, (406) 243-6088; Labs, (406) 329 4878, (406) 329-4861; Cell: (406) 546-7881  
Fax: (406) 243-5782, Email: bob.yokelson@umontana.edu

April 18, 2012

Dr. Arthur Sedlacek, III and Dr. Larry Kleinman  
Environmental Sciences Department  
Atmospheric Sciences Division,  
Brookhaven National Laboratory  
Mail Stop: 815E  
Upton, NY 11973-5000

Re: Study of Aerosol Produced in Biomass Burning.

Dear Drs Sedlacek and Kleinman,

I am very pleased to contribute to the study described in the DOE ASR proposal entitled, "Aerosol Properties Downwind of Biomass Burns." The proposal features an exceptionally strong group of collaborators that will have the capacity to probe biomass burning (an important, understudied source) with important new instrumental and modeling capabilities. I anticipate that my contribution would be as follows. Since 1997, I have conceived of, financed, and managed campaigns where I sampled well over a hundred different fires as flight scientist in Africa, Brazil, Mexico, and US (7 campaigns in the US alone). If I am supported to participate in the field campaign I anticipate providing assistance with: locating suitable fires via agency contacts and remote sensing; flight-planning; interpreting quick look data post-flight in terms of fire, fuels, and atmospheric characteristics, etc. I would also contribute post-mission to data analysis and quality publications if additionally supported for this purpose.

Sincerely,

Robert J. Yokelson



Department of Environmental Sciences  
School of Environmental and Biological Sciences  
Rutgers, The State University of New Jersey  
14 College Farm Road  
New Brunswick, NJ 08901

carlton@envsci.rutgers.edu  
732-932-9800, Ext. 6230

April 23, 2012

Dr. Larry Kleinman  
30 Bell Avenue  
Bldg. 815E  
Upton, NY 1173  
[kleinman@bnl.gov](mailto:kleinman@bnl.gov)

Dear Larry,

As a Principal Investigator for the Southern Oxidants and Aerosol Study (SOAS) in summer 2013, I am writing to affirm the synergies with a coincidental DOE study proposed for June – October 2013. NOAA, NSF and EPA are planning to sponsor aircraft and ground based measurements, in addition to process-level and photochemical modeling of the Southeastern U.S. Study data, including gas phase species and aerosol measurements, in addition to model emissions will be made publicly available and I affirm to sharing with DOE.

Sincerely,

A handwritten signature in blue ink that reads "Anmarie Carlton".

Anmarie Carlton, Ph.D., P.E.