

## **Black Carbon at the Mt. Bachelor Observatory Field Campaign Report**

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## Acronyms and Abbreviations

AAE	Angstrom absorption exponent
ARCTAS	Arctic Research of the Composition of the Troposphere from Aircraft and Satellites, a NASA mission
ARM	Atmospheric Radiation Measurement Climate Research Facility
ASL	above sea level
BB	biomass burning
BC	black carbon
BrC	brown carbon
CARB	California Air Resources Board
DOE	U.S. Department of Energy
Dpm	geometric mean diameter
HIAPER	High-performance Instrumented Airborne Platform for Environmental Research
HIPPO	HIAPER Pole-to-Pole Observations, an NSF project
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
MBO	Mt. Bachelor Observatory
MCE	modified combustion efficiency
MODIS	moderate resolution imaging spectroradiometer
nm	nanometer
NASA	National Aeronautics and Space Administration
NOAA	National Oceanic and Atmospheric Administration
NSF	National Science Foundation
PI	principal investigator
PMI	fine particulate matter
RMA	Reduced Major Axis
SMPS	scanning mobility particle sizer spectrometer
SP2	single-particle soot photometer
TAP	tricolor absorption photometer
UV	ultraviolet

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## 1.0 Summary

This campaign was initiated to measure refractory black carbon (rBC, as defined in Schwarz et al. (2010)) at the Mt. Bachelor Observatory (MBO) using the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facility single-particle soot photometer (SP2; unit 54). MBO is a high-elevation site located on the summit of Mt. Bachelor in central Oregon, USA (43.979°N, 121.687°W, 2,763 meters ASL). This site is operated by Professor Dan Jaffe's group at the University of Washington Bothell and has been used continuously as an atmospheric observatory for the past 12 years (Jaffe et al., 2005; Gratz et al., 2014). The location of MBO allows frequent sampling of the free troposphere along with a wide array of plumes from regional and distant sources. MBO is currently supported with funding from the National Science Foundation (NSF) to the Principal Investigator (PI; D. Jaffe) via the project "Influence of Free Tropospheric Ozone and PM on Surface Air Quality in the Western U.S." (#1447832) covering the period 03/15/2015 to 02/28/2018.

The SP2 instrument from Droplet Measurement Technologies provides particle-resolved measurements of rBC mass loading, size and mass distributions, and mixing state. The SP2 was installed at MBO on 6/27/2016 and ran through 9/23/2016. Additional measurements at MBO during this campaign included carbon monoxide (CO), fine particulate matter (PM<sub>1</sub>), aerosol light scattering coefficients ( $\sigma_{\text{scat}}$ ) at three wavelengths using a TSI nephelometer, aerosol absorption coefficients ( $\sigma_{\text{abs}}$ ) with the Brechtel tricolor absorption photometer (TAP), aerosol number size distributions with a scanning mobility particle sizer spectrometer (SMPS), and black carbon (eBC) with an aethalometer. BC data from this campaign have been submitted to the ARM Data Archive.

Black carbon (BC) is the predominant light-absorbing aerosol constituent in the atmosphere, and is estimated to exert a positive radiative forcing second only to CO<sub>2</sub> (Ramanathan and Carmichael, 2008). One of the largest sources of BC globally is biomass burning (BB). (Akagi et al., 2011; Andreae and Merlet, 2001; Bond et al., 2013; Bond et al., 2004; Reid et al., 2005b): a source that is likely to increase in the Western U.S. due to climate change (Dennison et al., 2014; Abatzoglou and Williams, 2016; Westerling et al., 2006). Given the likely increased role of BB aerosol in atmospheric forcing, we need to improve our understanding of the physical and optical properties of aged BB aerosol.

During the SP2 deployment period at MBO we observed seven BB events, all of which originated from the Gap Fire in Northern California, which burned 33,867 acres in the Klamath National Forest (<https://inciweb.nwcg.gov/incident/4997/>). Heavy smoke plumes from this fire were transported to MBO from 8/29/2016 to 8/31/2016. We calculated back-trajectories using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, version 4 (Draxler, 1999; Draxler and Hess, 1997, 1998; Stein et al., 2015) to confirm transport from the fire locations to MBO. We identified the fire locations using moderate resolution imaging spectroradiometer (MODIS) satellite-derived active fire counts (Justice et al., 2002). Transport time for all of the BB events ranged from 8 to 32 hours.

Going into the campaign we had a number of scientific questions specific to rBC:

1. How do the enhancement ratios of  $\Delta\text{rBC}/\Delta\text{CO}$  and  $\Delta\text{rBC}/\Delta\text{PM}$  in BB plumes depend on plume age, plume origin, modified combustion efficiency (MCE), and fire characteristics?

2. How does the absorption enhancement ( $\Delta\sigma_{\text{abs}}/\Delta\text{CO}$ ) and Ångström absorption exponent (AAE) of BB aerosol scale with rBC mass and size, rBC coating thickness, plume age, or origin (e.g., North America versus Siberia)?
3. What is the size distribution of rBC-containing particles in BB and Asian long-range transport plumes as a function of plume age, origin and/or fire characteristics (e.g., fire radiative power, MCE)? How do these size distributions compare with those reported by the SMPS?
4. What is the mass absorption cross-section of rBC ( $\text{MAC}_{\text{rBC}}$ ) in aged BB plumes and how does this vary with coating thickness, OA/rBC, plume age, or MCE?
5. How does the  $\Delta\text{rBC}/\Delta\text{CO}$  enhancement ratio for Asian long-range transport events compare to other observations (e.g., the HIPPO campaign) and what does this tell us about changes during transport and the rBC lifetime?

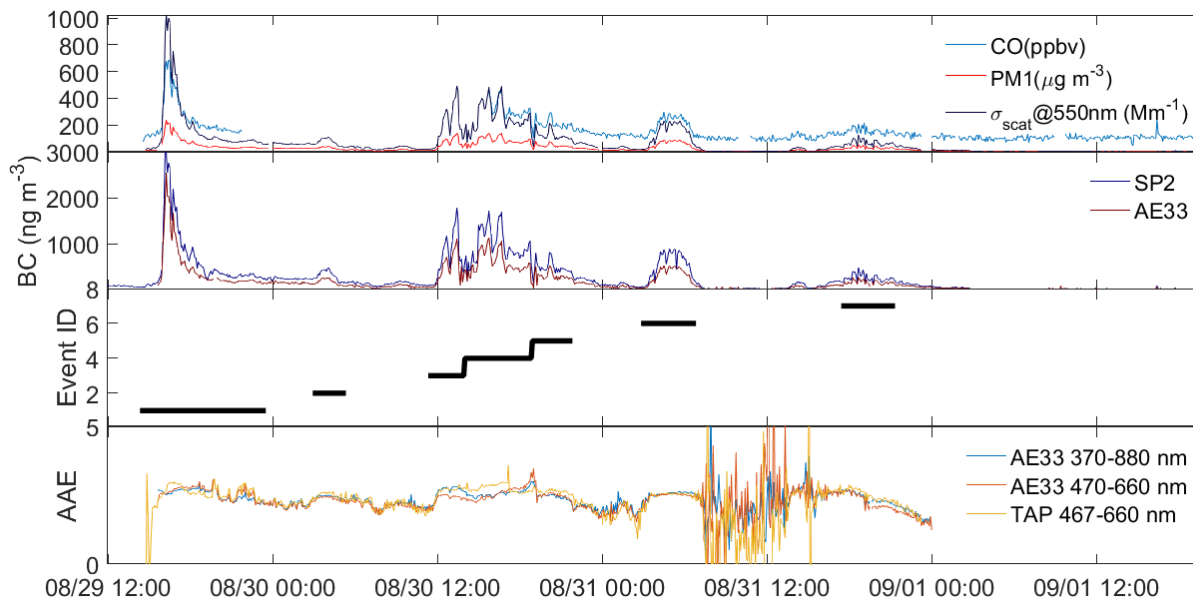
## 2.0 Results

### 2.1 Black Carbon Characteristics of Aged Biomass Burning Plumes

In Figure 1, we show the time series for the Gap Fire BB events. Figure 1a shows CO (blue trace),  $\sigma_{\text{scat}}$  at 550 nm (dark blue trace), and PM1 (red trace); 1b shows BC measurements by the SP2 (rBC; blue trace) and aethalometer (eBC; red trace); 1c shows the events; and 1d shows the Absorption Ångström exponent (AAE) values derived from the TAP (yellow trace) and aethalometer (blue and red traces), respectively. AAE values were calculated for a pair of wavelengths (e.g., for the TAP, 467 and 660 nm) using equation:

$$\text{AAE} = -\log(\sigma_{\text{abs}}^{467}/\sigma_{\text{abs}}^{660})/\log(467/660)$$

The high AAE values (2-3) for the BB events in Figure 1d confirm the presence of brown carbon (BrC), which preferentially absorbs at shorter wavelengths (near-ultraviolet [UV]). Pure BC, on the other hand, absorbs evenly throughout the visible spectrum and thus tends to exhibit an AAE  $\sim 1$ .

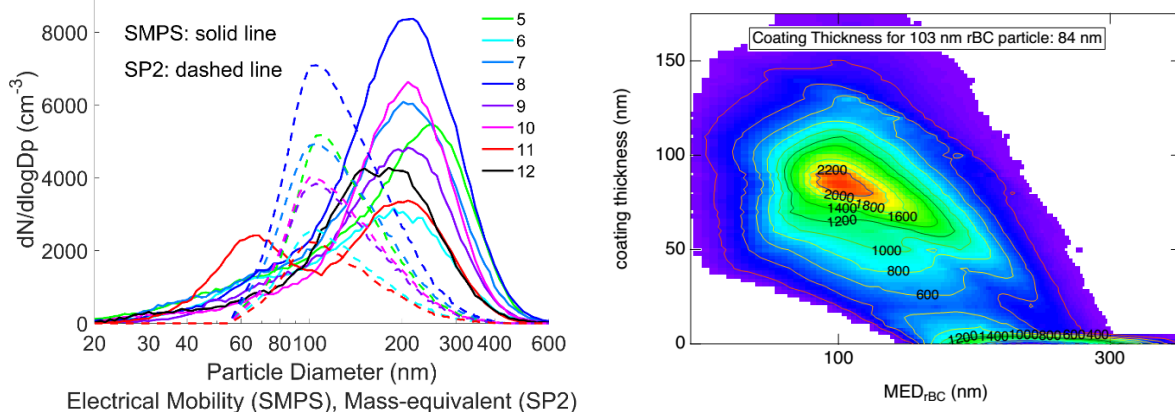


**Figure 1.** Time-series of SP2 data and other tracers during BB events from the Gap Fire in northern California as measured at MBO in summer 2016.

The BB events observed at MBO were well-aged in that the transport time from fire emission to observation at MBO was between 8 and 32 hours and the single scattering albedo ( $\omega$ ) for all of the events was between 0.96 and 0.97. The geometric mean diameter ( $D_{pm}$ ), as determined from the SMPS, for the BB events was  $158 \pm 15$  nm.

For each event, we calculated normalized enhancement ratios as the slope of the Reduced Major Axis (RMA) regression of species relative to carbon monoxide (CO), which is a conserved, inert tracer of combustion. Normalized enhancement ratios reflect the emission ratio of the two species plus any atmospheric processing that takes place between emission and observation. We found the  $\Delta rBC/\Delta CO$  enhancement ratios for the BB events to range from 3.29–4.98 ng m<sup>-3</sup> ppbv<sup>-1</sup>. These values are similar to  $\Delta rBC/\Delta CO$  observed for North American BB plumes during the National Aeronautics and Space Administration (NASA) Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS)-B ( $2.3 \pm 2.2$  ng m<sup>-3</sup> ppbv<sup>-1</sup>) and California Air Resources Board (CARB) ( $3.4 \pm 1.4$  ng m<sup>-3</sup> ppbv<sup>-1</sup>) flight campaigns (Kondo et al., 2011).  $rBC$  mass fraction ( $\Delta rBC/\Delta CO$ ) in the BB events ranged from 0.88 to 1.32%, a similar result to previous studies of BB plumes (Kondo et al., 2011).



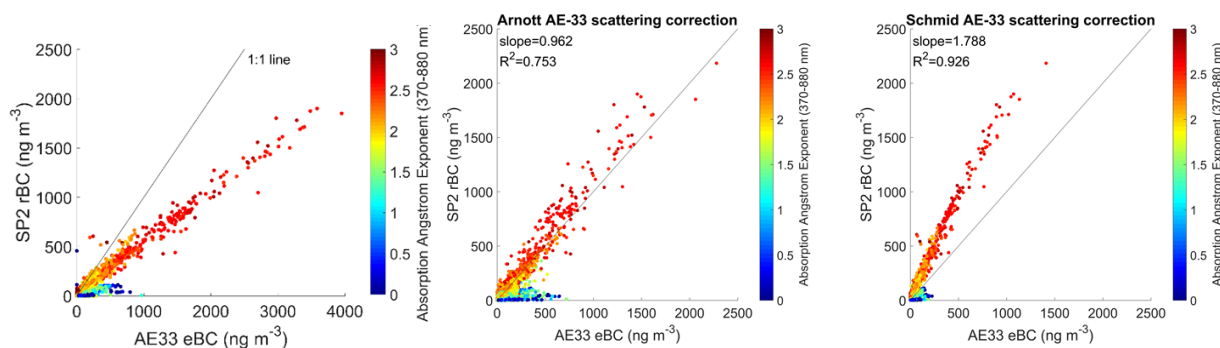


**Figure 2.** (a) Event-averaged aerosol (SMPS – solid line) and rBC (SP2 – dashed line) number size distributions for BB events at MBO. (b) Coating thickness on rBC particles for event 5.

Figure 2a shows the aerosol number size distributions measured by the SMPS and rBC number size distributions for the BB events. rBC particles are the primary particles emitted from fires, and upon atmospheric processing these particles grow due to the condensation of organic material onto the existing particles (Reid et al., 2005a; Seinfeld and Pandis, 2006). If we assume that the rBC particle is the core, and the subsequent organic material is the shell, we can estimate the coating thickness on the rBC particles. The rBC particles during BB events at MBO were thickly coated, which is expected for well-aged BB plumes. Figure 2b shows the coating thickness for one BB event to be 84 nm for a 103 nm particle.

## 2.2 Comparison of SP2 rBC and AE-33 eBC

In addition to the SP2, we operated an aethalometer (model AE-33), which measures “BC” indirectly by measuring the change in light transmission (attenuation) through a filter as it is being actively loaded by particles and assuming a mass absorption cross-section. In order to obtain eBC concentration, the AE-33 data has to be corrected for aerosol loading effects on the filter. BC concentration and ATN do change linearly due to these effects (Arnott et al., 2003; Collaud Coen et al., 2010). The AE-33 employs a dual-spot technology to obtain a real-time loading compensation factor (Drinovec et al., 2015). The AE-33 also uses a multiple-scattering correction factor (Weingartner et al., 2003).



**Figure 3.** Scatter plot between SP2 rBC and aethalometer eBC data (a) without a scattering correction factor, (b) using the Arnott et al. (2005) scattering correction and (c) using Schmid et al. (2006) scattering correction applied to the aethalometer data.

Figure 3a shows that with these internal correction methods the aethalometer overestimates BC relative to the SP2. We applied two scattering correction schemes for comparative purposes. We found the *Arnott et al.* (2003) correction to increase the slope of the scatter plot to close to 1, but decrease the  $R^2$  due to adding additional uncertainty and noise to the measurements. The *Schmid et al.* (2006) correction shows a good  $R^2$ , but leads to an underestimation of the AE33 eBC concentrations relative to rBC. Additional correction schemes will be explored to determine the best correction method for AE33 data for our situation.

## 2.3 Further Research Opportunities

During this summer 2016 campaign we characterized the rBC enhancement ratios, AAE values, and size distributions in seven smoke plumes. We found that these plumes had relatively uniform characteristics. Future research should include additional deployments of the SP2, and other tracers, to examine aerosol in a larger data set of biomass burning plumes as well as pollution plumes transported from Asia.

## 3.0 Presentations and Publications

Laing, JR, DA Jaffe, and AJ Sedlacek. Evaluation of Black Carbon, and Aerosol Physical and Optical Properties in Biomass Burning Plumes at the Mt. Bachelor Observatory in Central Oregon., presented to the American Meteorological Society's 97th Annual Meeting, Seattle, WA, January 24, 2017.

Laing, JR, DA Jaffe, and AJ Sedlacek. 2017. Black Carbon in Aged Biomass Burning Plumes at the Mt. Bachelor Observatory. In Preparation.

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