NOAA SINGLE PARTICLE SOOT PHOTOMETER (SP2)

BC is a near-pure carbon material produced via incomplete combustion. It is refractory (i.e., can survive heating to temperatures of ~3500 K) and an efficient absorber of visible and near-infrared radiation. Atmospheric aerosol BC is typically found mixed with a wide range of other materials within individual particles (i.e. internally mixed) as a result of co-emission, condensation, or coagulation. We refer to these non-BC materials generically as "coatings" although their actual morphology with respect to the BC component is not so simple.

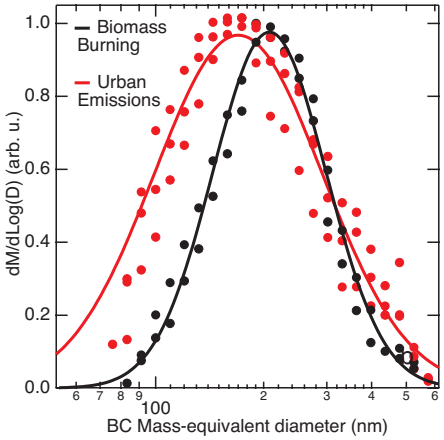

Figure 1: BC size distributions measured from two different types of sources during the TexAQS 2006 campaign. The systematic difference in sizes between the two sources has implications for BC absorption efficiency, CCN impact, and lifetime. Adapted from Schwarz et al., 2008b.

The Single-Particle Soot Photometer (SP2; Droplet Measurement Technologies, Boulder, Colorado) uses laser-induced incandescence to measure the BC mass content of individual particles independently of the total particle morphology and mixing state. It provides high time-resolution information about BC loadings, and provides size distributions, which contain information about the source and history of the aerosol (Figure 1). After years of improvement, the SP2 is a mature and competent instrument for assessing BC loadings even in exceptionally clean air, although correct operation still requires a dedicated attention to detail. Figure 2 shows vertical profiles of BC mass mixing ratio (MMR) compared to model profiles from the AeroCom global model suite [*Schwarz et al.*, 2013]. Obtaining this result required the SP2's high sensitivity, and led to identification of consistent global-scale biases in the models that help inform assessments of BC forcing.

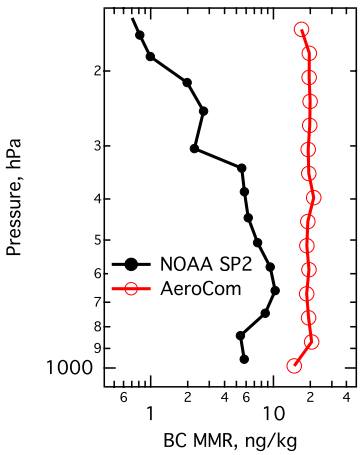

Figure 2: Vertical profiles of BC mass mixing ratio measured in and modeled for the NSF HIPPO campaign (2009 – 2011). The AeroCom model ensemble was biased high throughout the measured column.

The SP2 is shown schematically in Figure 3. Ambient air is drawn through an intense intracavity laser at 1.064 μm wavelength. Aerosol particles enter the laser singly, scattering laser light according to their size and composition. The intensity of the scattered light, and its evolution in time, is recorded. When BC enters the laser, it is heated to vaporization (~4000K), emitting blackbody radiation (incandescent light) in quantities directly related to its mass. The color of this radiation is detected and used to deduce the vaporization temperature of the particle as a constraint on its composition.

The incandescence signal intensity is linearly proportional to the BC mass over most of the BC mass in the accumulation mode [*Moteki and Kondo*, 2010]. The NOAA SP2 currently has a BC mass detection range of 0.5 – 350 fg (~0.08 – 0.6 μm mass equivalent diameter assuming 2g/cc density) [*Schwarz et al.*, 2010]. *Schwarz et al.* [2012] have extended SP2 detection to much larger BC particles (up to ~2 μm mass equivalent diameter), and the HD-SP2 system could be configured to this range if scientifically and practically relevant (i.e. if the aircraft inlet/aerosol transport tubing passes large particles with sufficient efficiency). A large uncertainty in some airmasses arises from BC mass outside of the SP2 detection range. In typical aged air and biomass burning emissions the undetected mass fraction can be quite small (~ 5%). However, in urban areas strongly influenced by modern emissions control technology, BC tends to smaller sizes, and up to 40% of BC mass can lie

outside the SP2 detection range (c.f. *Metcalf et al*., 2012). During ACCLIP, we expect to capture the dominant fraction of accumulation-mode BC mass.

The NOAA SP2s are calibrated with fullerene soot (Alfa Aesar, lot# F12S011), a material that has been shown to mimic ambient BC for calibration purposes with 15% uncertainty [*Gysel et al*., 2011; *Moteki and Kondo*, 2010], and which is recommended to the SP2 community for calibration [*Baumgardner et al*. 2012]. The validity of the calibration is the largest uncertainty in determining the accumulation-mode BC MMR.

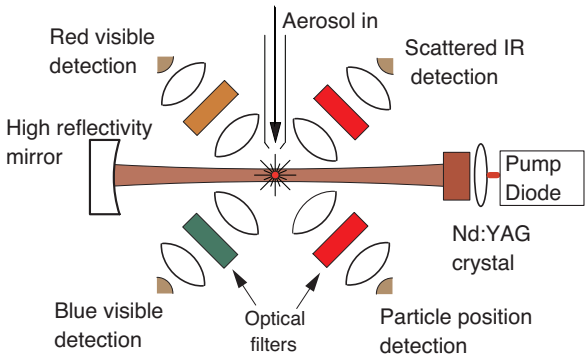

Figure 3: A schematic of the SP2 system, showing four detectors focused on the intersection of the high-intensity laser and aerosol jet.

A detector system developed by NOAA is used to optically size BC-containing particles before they are perturbed by laser heating [*Gao et al*., 2007]. This allows quantification of the amount of non-BC material (often interpreted as coating thickness via shell-core Mie theory) associated with each BC core, and calculation of its impact on the optical properties (including absorption cross-section) of the BC-component [*Schwarz et al*., 2008a]. This technique is typically limited to particles in the size range 200-500 nm optical diameter.

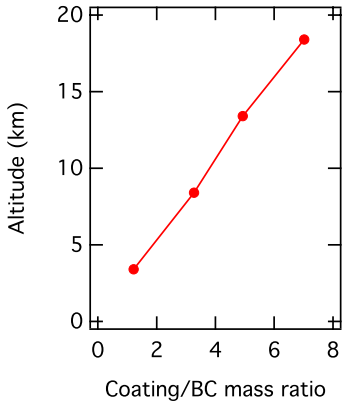

Figure 4: Ratio of coating to BC mass at different altitudes in the tropics. Adapted from Schwarz et al., 2008a.

The coating thickness on a BC core is related to its history and source [*Schwarz et al*., 2008b]; freshly emitted BC tends to be relatively bare, accruing additional coating materials with time. Coatings on a BC core increase its absorption of sunlight, and affect the likelihood of BC's removal through wet deposition [*Lance et al*., 2013].

Coatings on BC are central to uncertainties in modeling BC removal from the atmosphere, which has been identified as a leading cause of model bias over much of the globe [*Schwarz et al*., 2010], and is of specific interest in ACCLIP, with relvance to all primary aerosol injection into the stratosphere by the monsoon. Figure 4 shows a vertical profile of dry coating-mass to BC-mass measured in the tropics from near the ground to 19 km altitude. The increasing fraction of coating mass increases BC forcing efficiency at high altitudes beyond the levels predicted without this trend [*Samset et al*., 2013]. Testing this behavior in ACCLIP will shed light on the processes affecting transport of BC and other tropospheric aerosols in the monsoon.

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