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Black Carbon Aerosol Deposition Study (BCADS) Field Campaign Report

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Acronyms and Abbreviations

ARM	Atmospheric Radiation Measurement
BC	black carbon
BCADS	Black Carbon Aerosol Deposition Study
CCN	cloud condensation nuclei
DOE	U.S. Department of Energy
ECOR	eddy correlation flux measurement system
LOD	length of day
rBC	refractory black carbon
SGP	Southern Great Plains
SP2	single-particle soot photometer
UHSAS	ultra-high-sensitivity aerosol spectrometer

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1.0 Summary

Black carbon (BC) absorbs incident solar radiation, perturbs temperature gradients in the atmosphere, and indirectly impacts cloud formation and optical properties (Koch and Del Genio 2010). Deposition of BC to snow and ice surfaces alters albedo and enhances snow melt (Hansen and Nazarenko 2004; Flanner et al., 2007). The impact of BC on regional and global climates through these processes depends on its atmospheric concentration, and thus on the relative rates of emission and loss. Combustion of fossil fuels and biofuel, biomass burning, and wildfires are major sources of BC (Bond et al., 2004). Significant sinks include wet and dry deposition. Wet deposition occurs through scavenging by cloud droplets, ice crystals, and precipitation, while dry deposition refers to the direct removal of particles in the atmosphere to planetary surfaces (e.g., plant, soil, ocean, ice surfaces). Bond et al. (2013) and references therein have focused on the source, aging, and optical properties of BC, while deposition components of the BC lifecycle remain poorly constrained. This is predominantly due to the lack of observations of both total aerosol and BC deposition. Removal rates of refractory and non-refractory sub-micron aerosol by wet and dry deposition are one of the most uncertain aspects of modelling cloud condensation nuclei (CCN; Lee et al., [2013]). BC is an ideal tracer for particle deposition because it is non-volatile and effectively chemically inert, though it can become mixed with other species in the atmosphere. Unlike other aerosol species, BC remains 'intact' in water and has no confounding gas-phase contribution to precipitation measurements, and can thus be used to examine the relative importance of wet and dry deposition. Thus, measurements of BC deposition are not only essential for constraining BC sinks and atmospheric lifetime, but also useful for investigating aerosol deposition more broadly.

Figure 1. Instrument set-up for BCADS 2017. The blue boxes housed the pumps, and the air conditioned box housed the SP2) and UHSAS.

The goals of this work were to (1) demonstrate that the single-particle soot photometer (SP2; Droplet Measurements Technology, Boulder, Colorado) instrument is capable of direct flux measurement by the eddy covariance technique, (2) make simultaneous measurements of wet and dry deposition of refractory black carbon over the course of several weeks, and (3) provide an observational constraint on removal rates of BC from the atmosphere. We succeeded in all three goals.

The Black Carbon Aerosol Deposition Study (BCADS 2017) took place at the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facility Southern Great Plains (SGP) site in Lamont, Oklahoma, (36° 36' 18'' N, 97° 29' 6'' W; 312 m above sea level) from 12 June to 23 July, 2017. The general site and measurement environment are described elsewhere (Fischer et al., 2007; Riley et al., 2009; Sisterson et al., 2016). Figure 2 shows relevant meteorological parameters; temperatures varied between 25 °C and 35 °C and the total precipitation recorded during the campaign is 148 mm. The particle inlet and sonic anemometer (Gill Instruments, WindMaster Pro.) were located 2.7 m above ground level on the SGP ECOR 14 (CO_2 flux) tower. The inlet was aligned downward (~45° angle) and located below the center of the sonic anemometer (~40 cm vertical and ~20 cm horizontal displacement). The inlet was 4.5 m of 6.35 mm o.d. stainless steel tubing with a wire mesh screen to exclude insect and debris contamination, and led to the SP2 and the ultra-high-sensitivity aerosol spectrometer (UHSAS), which were housed in an air-conditioned box at the base of the tower. A bypass pump coupled to a mass flow controller maintained turbulent flow (~12 L min-1; Re≈3000; residence time of 0.9 s). Particle losses in the system are estimated to be <5% for the size range measured by the SP2 (70-600 nm).

2.0 Results

This section provides details about our findings about black carbon aerosol deposition during the study.

2.1 Demonstration of SP2 for Eddy Covariance

We demonstrated that the SP2 is fast, sensitive, and selective enough for eddy covariance flux measurements. These requirements are validated by spectral analysis (e.g., Figure 1), in which the co-spectra of vertical wind speed and rBC measurement (mass or counts) are compared to the simultaneous sensible heat flux. Log-binned frequency data shows a (Hz)-4/3 response between 0.01 Hz and 3 Hz, characteristic of the inertial sub-range predicted from Kolmogorov theory and demonstrating that there are no interferences to measurement of turbulent flow (i.e., the measurements were sufficiently high above the ground to capture turbulent energy transfer) (Kaimal and Finnigan 1994). Capture of this inertial sub-range is necessary for accurate eddy covariance fluxes and requires the measurement to be both fast enough and long enough for flux measurement (Baldocchi et al., 1988). The co-spectrum of rBC mass and particle counts follow sensible heat flux, further validating that the inlet was adequately designed to prevent line losses. The lack of spectral attenuation (i.e., steeper slope at high frequencies) indicates no flux underestimation due to high-frequency damping within inlet lines, and that the instrument has a sufficiently fast response for eddy covariance flux measurements.

The cumulative co-spectrum, or ogive, (Figure 1c) demonstrates which frequencies contribute to the total flux. Little increase in cumulative flux at low frequencies suggests that the 30-minute flux averaging periods are sufficiently long to capture the bulk of the flux. The leveling off at high frequencies indicates that the measurement was fast enough to capture flux contributions from the smallest eddies. However,

individual flux periods do not always show this flattening at high frequencies, and we attribute at least some of this loss to sensor separation.

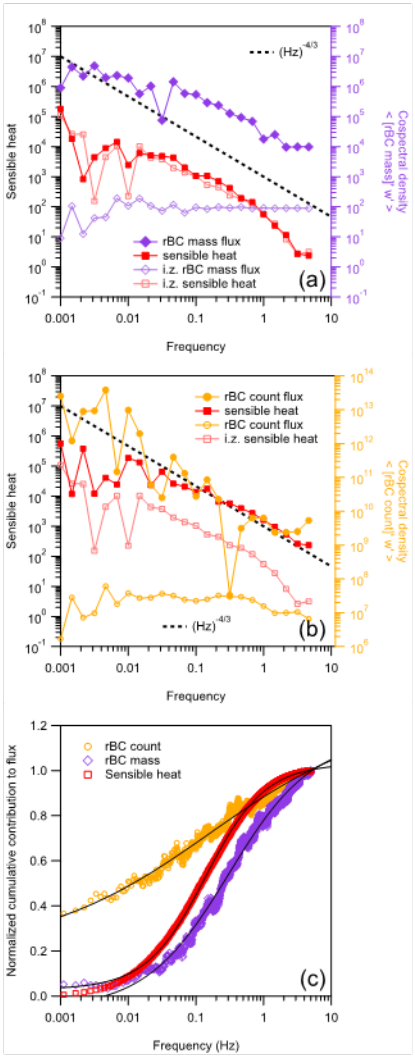

Figure 2. Co-spectral density of (a) refractory black carbon (rBC) mass fluxes, (b) rBC particle count fluxes, and (c) corresponding ogives. Data are shown in comparison to sensible heat for the same time period and an example instrument zero (i.z.). Data presented represent 25 logarithmically spaced bins whose average values are shown; negatives are shown as the absolute value. Ogives presented here are normalized cumulative contributions to the flux based on integrated co-spectral density. Points shown here are medians of all quality-controlled data for each frequency across the displayed range. Curve fits are Hill Functions.

2.2 Refractory Black Carbon Fluxes

Mass and particle concentrations (Figure 2) are representative of a remote North American field site away from major anthropogenic influences (Koch et al., 2009). During BCADS 2017, the median observed rBC mass concentration is 78 ±1 ng m^{-3} and median rBC number concentration is 79±1 # cm^{-3}. rBC mass and number concentrations were consistent throughout the campaign until the last week (11 July to 19 July), when concentrations were suppressed. This decrease occurred over several days, suggesting a larger

meteorological shift or simply a decrease in farming activity in the region. The diel cycle is characteristic of regular boundary-layer expansion observed at the site.

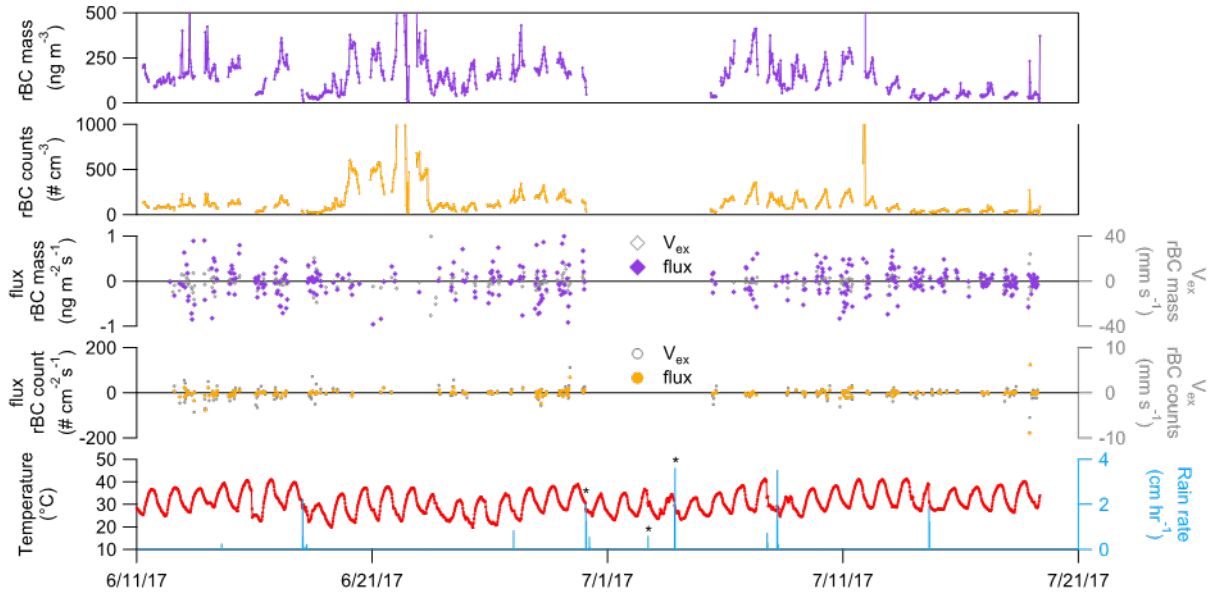

Figure 3. Campaign overview of rBC mass, particle counts, fluxes, vertical exchange, and several meteorological parameters. Flux periods shown are all possible flux periods and have not been quality controlled. Data presented subsequently represents quality-controlled data. Rain periods marked with an (*) refer to periods of collected precipitation. Rain rates were measured using a weighing bucket precipitation gauge (ARM Climate Research Facility, Weighing Bucket Precipitation Gauge).

Average rBC mass and particle number fluxes are negative (i.e., downward) in mid-afternoon (Figure 3) when friction velocity (u*) is at a maximum. During mid-morning, the fluxes have a slight propensity upwards. At night, the fluxes are bi-directional and relatively small, with large uncertainties. Similar trends are observed for exchange velocity. The diel profile in V_{dep}¬ is consistent with several atmospheric patterns. Boundary-layer height increases in the early morning hours, which could cause a decrease in particle number and mass concentration and thus a flux period dominated by downdrafts would have an upward flux. The enhanced afternoon deposition is likely a result of increased atmospheric turbulence and friction velocity. V_{dep} increases with u*, but the correlation is inadequate to establish a parameterization (r2 ≈ 0.25 and 0.15 for mass and counts respectively).

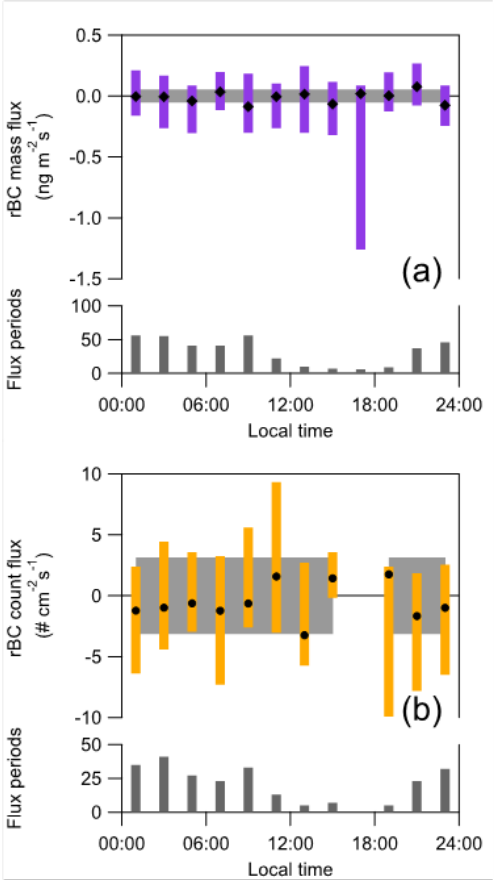

Figure 4. Bi-hourly binned fluxes of rBC counts (a) and rBC mass (b). Symbols are medians; upper and lower ends of the boxes are the 75th and the 25th percentiles respectively. The shaded grey bar represent the length of day (LOD), and bottom vertical bars are the number of flux periods that represent the data above.

Restricting the analysis to periods in which deposition was observed, the ensemble distribution of deposition velocities follow a log-normal distribution. We calculate a V_{dep} for rBC mass of 2.3±1.4 mm s^{-1} and particle number of 0.6±1.5 mm s^{-1}. To our knowledge, this represents the first in situ estimate of rBC deposition velocities. Our observed deposition velocities by particle number are consistent with values used in current global models. Huang et al. (2010) and Reddy and Boucher (2004) employed a global annual mean BC and organic aerosol deposition velocity of 1 mm s^{-1} for particle numbers in the submicron mode. Wesely (1989) used a particle number deposition velocity of 0.8 mm s^{-1} over snow and ice surfaces and Liu et al. (2011) improved their Arctic BC simulations with number deposition velocities of 0.1 to 0.7 mm s^{-1}. The discrepancy in values used in cryosphere/Arctic simulations may be due to surface properties, suggesting a need for further rBC flux measurements over the cryosphere.

2.3 Wet and Dry rBC Deposition

From the three rain events, we estimate an average wet deposition flux ($\bar{F}$) of 0.05 ± 0.1 mg m^{-2} day^{-1}. These data indicate dry deposition accounts for 37 ± 7% of the total rBC mass deposition, which is larger than the typical 5-20% assumed in global climate models (Koch et al., 2009). However, this value is highly dependent on precipitation rates. Deposition velocities describe the efficiency of the loss process,

allowing us to estimate the lifetime of rBC with respect to wet and dry deposition as a function of boundary-layer height. We calculate lifetimes of 1-3 and 3-14 days for wet and dry deposition, respectively (95th % confidence interval).These lifetimes are shorter than the 2-3 weeks typically described in the literature. These observations suggest that either dry deposition is faster than model predictions, or wet deposition is less efficient than expected. While dry deposition rates are assumed to be invariant, wet deposition processes may be first order with respect to rBC burden. However, only one precipitation event (of seven observed) shows a rapid decrease of rBC number and mass concentrations. Below-cloud washout of rBC may not be efficient and the loss mechanism may be dominated by in-cloud scavenging.

3.0 Future Opportunities

This work raises several questions that warrant further investigation. In particular:

1. How do changes in precipitation and rBC mass loading impact the relative amounts of wet and dry deposition?

   That is, how robust is the observation that dry deposition accounts for about a third of the loss processes of rBC at SGP?

2. How do surface properties (Monin-Obukhov length, homogeneity, water content) impact deposition velocity?

   That is, to what extent can measurements from SGP be extrapolated to the cryosphere or other regions?

These questions provide opportunities for future work to better constrain wet and dry deposition of rBC.

4.0 Publications and References

4.1 Presentations

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4.2 Journal Articles

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