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Arctic Black Carbon Loading and Profile Using the Single-Particle Soot Photometer (SP2) Field Campaign Report

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ARM CLIMATE RESEARCH FACILITY

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Executive Summary

One of the major issues confronting aerosol climate simulations of the Arctic and Antarctic cryospheres is the lack of detailed data on the vertical and spatial distribution of aerosols with which to test these models. This is due, in part, to the inherent difficulty of conducting such measurements in extreme environments. However given the pronounced sensitivity of the polar regions to radiative balance perturbations, it is incumbent upon our community to better understand and quantify these perturbations, and their unique feedbacks, so that robust model predictions of this region can be realized. One class of under-measured radiative forcing agents in the polar region is the absorbing aerosol—black carbon and brown carbon.

Black carbon (BC; also referred to as light-absorbing carbon [LAC], refractory black carbon [rBC], and soot) is second only to CO_2 as a positive forcing agent. Roughly 60% of BC emissions can be attributed to anthropogenic sources (fossil fuel combustion and open-pit cooking), with the remaining fraction being due to biomass burning. Brown carbon (BrC), a major component of biomass burning, collectively refers to non-BC carbonaceous aerosols that typically possess minimal light absorption at visible wavelengths but exhibit pronounced light absorption in the near-ultraviolet (UV) spectrum. Both species can be sourced locally or be remotely transported to the Arctic region and are expected to perturb the radiative balance.

The work conducted in this field campaign addresses one of the more glaring deficiencies currently limiting improved quantification of the impact of BC radiative forcing in the cryosphere: the paucity of data on the vertical and spatial distributions of BC. By expanding the Gulfstream aircraft (G-1) payload for the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) Climate Research Facility-sponsored ACME-V campaign to include the Single-Particle Soot Photometer (SP2)) and leveraging the ACME-V campaign's deployment within the Arctic Circle during the summer of 2015 (Deadhorse, Alaska [70° 12′ 20″ N, 148° 30′ 42″ W]), the truly unique opportunity presented itself to acquire profile data on BC loading at little additional cost. Since the SP2 is a particle-resolved measurement, the resulting data set provides refractory black carbon (rBC) mass loadings, size and mass distributions, and rBC-containing particle mixing state, all of which are expected to readily find value in the modeling community. As part of the ACME-V (http://www.arm.gov/campaigns/aaf2014armacmev) campaign, CO, CO_2, and CH_4 were also measured, providing the unique opportunity for carbon closure. We will also work closely with modelers who require such data and expect this collaboration will lead directly to a better understanding of the climate impacts of BC in the Arctic.

The primary measurement objective was to acquire airborne data on the vertical and spatial distributions of refractory black carbon (rBC) loading, size and mass distribution, and particle mixing state.

The primary scientific objective was to provide a targeted data set of rBC particle distributions to better understand and constrain the impact of black carbon radiative forcing in the cryosphere.

The SP2-based data set during this campaign is available in the DOE-ARM archive (http://www.arm.gov/campaigns/aaf2015abclp).

Acronyms and Abbreviations

AAF	ARM Aerial Facility
AAOD	aerosol absorption optical depth
ABoVE	Arctic-Boreal Vulnerability Experiment, a NASA project
ARM	Atmospheric Radiation Measurement Climate Research Facility
BB	biomass burning
BC or rBC	black carbon or refractory black carbon
BNL	Brookhaven National Laboratory
BrC	brown carbon
CARVE	Carbon in the Arctic Reservoirs Vulnerability Experiment, a NASA project
DOE	U.S. Department of Energy
G-1	Gulfstream-1 research aircraft
GHG	greenhouse gas
ICARTT	International Consortium for Atmospheric Research on Transport and Transformation
LAC	light-absorbing carbon
m	meter
mbar	millibar
nm	nanometer (10^{-9} m)
NASA	National Aeronautics and Space Administration
NGEE	Next-Generation Ecological Experiment
NOAA	National Oceanic and Atmospheric Administration
OA(s)	organic aerosol(s)
PbP	particle-by-particle
PNNL	Pacific Northwest National Laboratory
PSL	photo-stimulated luminescence
rBC	refractory black carbon
SP	Single-Particle Soot Photometer
SP-AMS	Soot Photometer – Aerosol Mass Spectrometer
UTC	Coordinated Universal Time
UV	ultraviolet

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Figures

1.0 Background

1.1 Introduction

The polar regions are recognized as exhibiting pronounced sensitivity to changes in radiative forcing. Indeed, the cryosphere is often referred to as the 'canary in the coalmine' for climate change in the popular literature. It is this sensitivity that provides both motivation and need for targeted measurement campaigns that test the behavior and predictive capabilities of current climate models that will lead to an improved understanding of which factors are most important in Arctic climate change. Furthermore, this information is of critical importance if we are to better understand and quantify the impacts of continued commercial development in this region. For example, the economic advantage realized by using heretofore unusable shipping lanes within the Arctic Circle must be balanced by the impact ship emissions will likely exert on the environment. Of specific interest to the present work is how the cryosphere responds to absorbing aerosols such as black carbon and brown carbon.

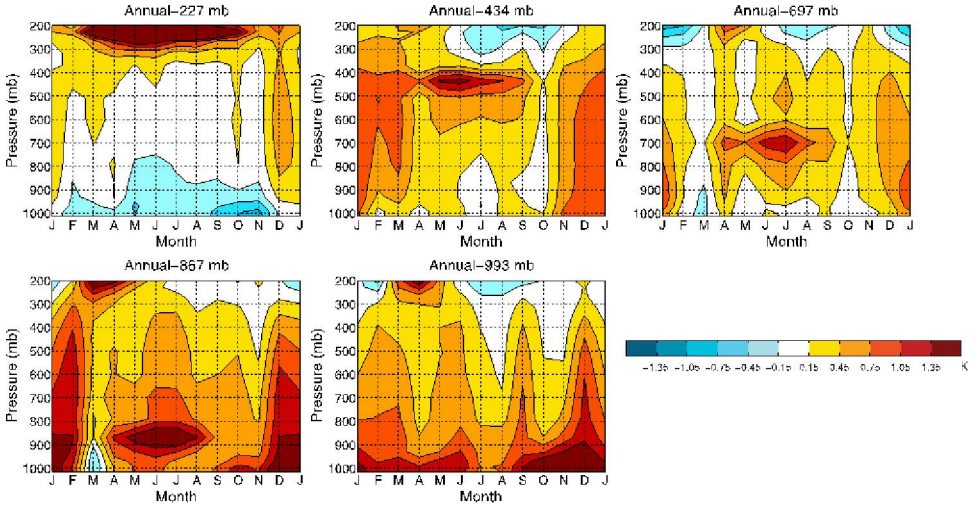

Figure 1. Atmospheric and surface air temperature as a function of month and altitude for five injection heights of BC. Taken from Flanner 2013. Aerosol absorption optical depth (AAOD) is fixed 0.005.

With respect to light absorbing aerosols, the cryosphere is a truly unique region in that it provides these aerosols types two radiative forcing routes: atmospheric and non-atmospheric. In the atmospheric route, light-absorbing aerosols in the Arctic are expected to contribute to radiative forcing much as they do in the mid-latitudes—through atmospheric warming (direct effect) and alteration of cloud properties (semi-direct effect) collectively causing surface dimming and subsequent surface cooling. In fact, such behavior has recently been predicted in a series of equilibrium climate simulations conducted by Flanner (2013) where various case scenarios ('experiments') were examined. In one set of experiments where the impact of atmospheric BC was studied at differing 'injection' heights, Flanner found that BC injected into the upper atmosphere (~ 200 mBar) led to an at altitude warming with increased atmospheric stability and concomitant surface dimming. Taken together, this scenario leads to negative forcing at the surface. However, when BC was injected between 867-993 mBar, a positive forcing condition is created since the lower injection height enables more efficient heat coupling to the surface that triggers a positive feedback causing accelerated snow/ice melt, which reduces of the surface albedo (Hansen and Nazarenko 2004). Indeed, this increased surface light absorption has been found to overcome the surface dimming due to BC aloft, thereby leading to an overall increase in surface temperature. The two scenarios outlined here,

along with two immediate cases, are shown in Figure 1, where the temperature profiles are plotted as a function of month for five different injection heights of BC (Flanner 2013). As is readily seen, the surface temperature response is seen to be sensitive to the BC vertical distribution.

In addition to the injection height, Flanner also reported that extra-Arctic BC (that is, remotely transported BC) is expected to warm the Arctic by increasing the poleward heat flux. One potential consequence of this finding is that extra-Arctic BC could lead to more efficient removal through wet deposition, thereby further contributing to the reduction of the surface albedo. This hypothesis suggests that the modeling of Arctic BC should include the complete aerosol lifecycle to accurately capture the impact of this aerosol type. Unfortunately, data on rBC mixing state is virtually non-existent for this region.

Finally, it is interesting to note that in a model-measurement intercomparison conducted by Koch et al. (2010), it was reported that the average model-to-aircraft BC ratio was 0.4, indicating that the models were under-predicting the observed BC loading in the lower and middle troposphere during the Arctic spring. However, as pointed out by Flanner (2013), it is important to note that such comparisons are severely hampered due to the paucity of available aircraft measurements.

While the Flanner simulations highlight here were restricted to examining the impact of BC, it is reasonable to expect similar sensitivity of the Arctic surface temperature to the vertical and spatial distribution to other light-absorbing aerosols, such as BrC. However, the present work is limited to measurement of the rBC vertical and spatial distributions.

In contrast to the mid-latitudes, the cryosphere provides BC a second route towards influencing surface temperatures—deposition on the surface (Hansen and Nazarenko 2004). Not surprisingly, on-surface BC will result in an accelerated decrease in surface albedo that will activate a positive feedback loop wherein the increased surface light absorption leads to more snow melt, which, in turn, causes further reduction in surface albedo. The impact of BC deposition is expected to be primarily from local sources (e.g., commercial activities, energy extraction, and shipping) and thus most sensitive to commercial development in the area. However, as highlighted above, aged, extra-Arctic BC could contribute to surface albedo reduction through wet deposition during the summer months. Additionally, since BC is insoluble, melt cycles will expose previously deposited BC, increasing the overall on-surface concentration of BC and further reducing the surface albedo. Flanner (2013) predicts that the combined forcings of local atmospheric and cryosphere-deposited BC should warm the Arctic surface with a sensitivity of 0.5 ± 0.4 K (W m 2) -1 while higher-altitude BC will slightly cool the surface.

In addition to anthropogenic sources of BC, wildfires (biomass burning; BB) can inject large quantities of BC and BrC into the atmosphere. Indeed, large biomass burning events originating in Russia have been measured in the Alaskan Arctic (Warneke et al. 2010). In an aircraft study, Warneke et al. (2010) reported that advected plumes from Russian springtime wildfires more than doubled aerosol burden (organic and BC) above the so-called Arctic haze levels. One interesting question is how BB BC will impact the radiative balance in this region? Given that the long-range transport of BB emissions should restrict these species to the upper atmosphere, this additional injection of aerosol would be expected to further enhance cooling through surface dimming, consistent with the simulations by Flanner (2013). However, with long-range transport comes aerosol aging and the increased efficiency of BC removal through wet deposition processes, potentially leading to increase snowmelt and surface warming (Warneke et al. 2010; Flanner 2009, 2013). It is also noted that the dominance of organic aerosols (OA) relative to BC in BB plumes (typically > 80% of the BB burden) would be expected to exert a net cooling effect for plumes aloft, potentially enhancing net surface cooling.

he complexity of radiative forcing exhibited by absorbing aerosols in the cryosphere as highlighted by the model simulations and model-experimental comparisons discussed here underscores the pressing need for additional observations that can be used validate these models and, where possible, provide guidance towards model improvements. It is through this data-model feedback that the necessary insight and quantification of climate change in the Arctic will be realized, and that can, in turn, be used to provide policymakers with critical information on the potential impacts of further commercial development in this hypersensitive region.

1.2 Airborne Carbon Measurements Experiment (ACME)

The Arctic is a climatically sensitive region, and high latitudes have experienced the greatest regional warming in recent decades (Hansen et al. 2010). In fact, this warming trend is projected to increase faster in the Arctic than anywhere else on the globe (Chapman and Walsh 2007). One of the characteristics of the Arctic is permafrost, a layer of permanently frozen subsoil, which stores large amount of carbon (Schuur et al. 2009). Observations suggest that permafrost degradation is happening at a fast pace and linked to increasing air temperature (Jorgenson et al. 2006). Permafrost degradation is expected to affect climate forcing (Callaghan et al. 2011) through biogeochemical (release of CO_2 and/or CH_4) and biophysical feedbacks (inundation, drainage, land cover). The most dramatic changes are expected to occur in the ice-rich permafrost region of the Arctic: the Alaska interior and the North Slope.

The rate at which permafrost degradation is occurring is difficult to quantify and models do not agree on its magnitude (Koven et al., 2013). The goal of the DOE-funded Next-Generation Ecological Experiment in the Arctic (NGEE) is to improve model representations of interactions between vegetation, soils, precipitation, and soil moisture (Koven et al. 2013) that control carbon emissions from Arctic soils. Recent global inverse models and aircraft measurements have found no evidence for increasing CH_4 emissions from Arctic regions in the last 10 years (Bergamaschi et al. 2013, Chang et al. 2014), despite warming, in contrast, to CO_2 emissions (Schaefer et al. 2014).

NGEE-Arctic project and eddy covariance towers observations provide observations at small spatial scale (1-100m) and aircraft-based observations are needed. The National Aeronautics and Space Administration (NASA)-sponsored Carbon in the Arctic Reservoirs Vulnerability Experiment (CARVE) helped linked ground observations to regional scales, but focused on Alaska as a whole. In the summer 2015, ARM Aerial Facility (AAF) will deploy the AAF G-1 research aircraft to fly at the North Slope of Alaska, with occasional vertical profiling to measure background concentrations around Prudhoe Bay, Oliktok Point, Barrow, Atqasuk, Ivotuk, and Toolik Lake. The aircraft payload includes an existing in situ Picarro analyzer for continuous measurements of CO_2, CH_4, and H_2O mixing ratios, a 12-flask sampler for National Oceanic and Atmospheric Administration (NOAA) analysis of carbon cycle gases (CO, CH_4, N_2O, $13CO_2$, $14CO_2$, carbonyl sulfide, and trace hydrocarbon species). This research will build upon the results from the NASA CARVE missions, the DOE NGEE-Arctic project, and the NASA Arctic-Boreal Vulnerability Experiment (ABoVE). The proposed ACME flights focus on the North Slope and are designed to collect essential data needed to address three science questions:

1. Characterize atmospheric mixing ratios across the North Slope region.
2. Evaluate representativeness of ground sites at regional scales.
3. Relate spatial and seasonal differences in greenhouse gas (GHG) sources (e.g., proximity to fossil sources), land-surface properties (e.g., thaw or inundation), and atmospheric transport to variations in CO_2 and CH_4 mixing ratios.

The deployment of the Single-Particle Soot Photometer (SP2) leveraged the ARM-sponsored Airborne Carbon Measurements (ACME V) staged out of Deadhorse Alaska (70° 12′ 20″ N, 148° 30′ 42″ W) where flights took place a couple of times a week during this campaign.

1.3 Instrumentation Capabilities, Measurement Strategy, and Data Products

The Single -article Soot Photometer (SP2, Droplet Measurement Technologies) is part of a new generation of aerosol instrumentation capable of providing particle-resolved measurements. It achieves this by measuring the time-dependent scattering and incandescence signals produced by individual BC-containing particles as they travel through a continuous-wave laser beam (Schwarz et al. 2006, Sedlacek et al. 2012). Any particle traversing the laser beam will scatter some of the light while the BC component of a particle will absorb some of the laser energy until its temperature is raised to the point at which it incandesces (within the SP2 community the substance determined by the SP2 to be BC is referred to as refractory black carbon (rBC)). The incandescence signal is used for counting rBC-containing particles and determining the mass of the rBC component within each particle (obtained via calibration). Therefore, the SP2 can provide estimates of the size-resolved number and mass concentrations of rBC-containing particles. In addition to rBC mass loading, when the incandescence signal from individual rBC-containing particles is combined with the signal from the concomitantly acquired scattering channel, information on the rBC particle-population-based mixing states can be probed. Being a particle-resolved measurement, the SP2 is ideally suited for the very low rBC mass loadings expected in the Arctic.

The SP2 has been shown to require minimal setup prior to a research flight and negligible oversight during flight. Therefore expansion of the ACME-V instrument suite to include the SP2 is expected to have a negligible impact on the primary objective of the ACME-V field campaign or on the operation of ACME-V centric instrumentation. A pre-campaign calibration will be conducted in Pasco, Washington prior to the G-1's departure to Deadhorse, Alaska. This will be followed by a mid-campaign calibration on location. Calibration will be conducted using Fullerene soot as the rBC calibration standard and photo-stimulated luminescence (PSL) for the scattering channel.

1.4 Principal Investigator

Arthur J. Sedlacek III (Brookhaven National Laboratory)

1.5 Co-Investigators

Sébastien Biraud (Lawrence Berkeley National Laboratory)

Yan Feng (Argonne National Laboratory)

1.6 Additional Team Members

Special acknowledgement is called out to the AAF pilots, led by Mike Hubble, who safely and skillfully implemented the scientific goal of conducting the vertical profiling transects.

2.0 Notable Events or Highlights

The most notable event was the encounter of a smoke plume on June 30, 2015.

2.1 Instrument Issues

There were no instrumentation issues during the ACME-V campaign.

3.0 Lessons Learned

The campaign was conducted both professionally and safely and met the science and measurement goals of this Arctic BC study. No notable lessons were learned.

4.0 Results

4.1 Calibration plots

Figure 2 shows calibration plots for the SP2 (upper left) and CO (lower left) instruments along with a time series trace of the CO and rBC (refractory BC) measured during the June 30 flight (upper right) and a correlation plot of the CO vs rBC. As can been seen, excellent agreement between the SP2 and the CO is observed.

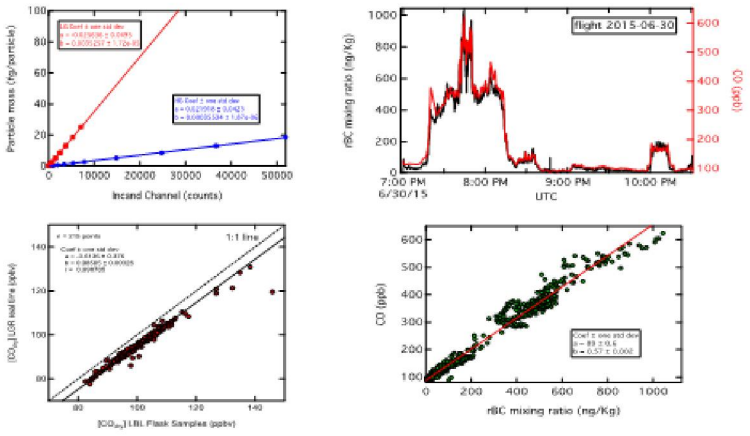

Figure 2. Calibration and correlation plots.

4.2 Example of Monthly Profiling Statistics

ACME-V provided the opportunity to collect much-needed statistics on the rBC vertical profile in the North Slope region. These data sets provide constraints of model simulated vertical profiles of BC over the Arctic region. Shown in Figure 3 are box and whisker plots of the monthly median, means and 10, 25, 75, and 90 percentiles are shown for Barrow, Oliktok, PATQ, and Toolik. The dotted line is a CAM5

model based on 2011 inventory. Observation-model agreement varies from very good: PATQ (June/July); Toolik (July); Barrow (July); to very poor (3x difference): Toolik (June and September); Barrow (August); Oliktok (August).

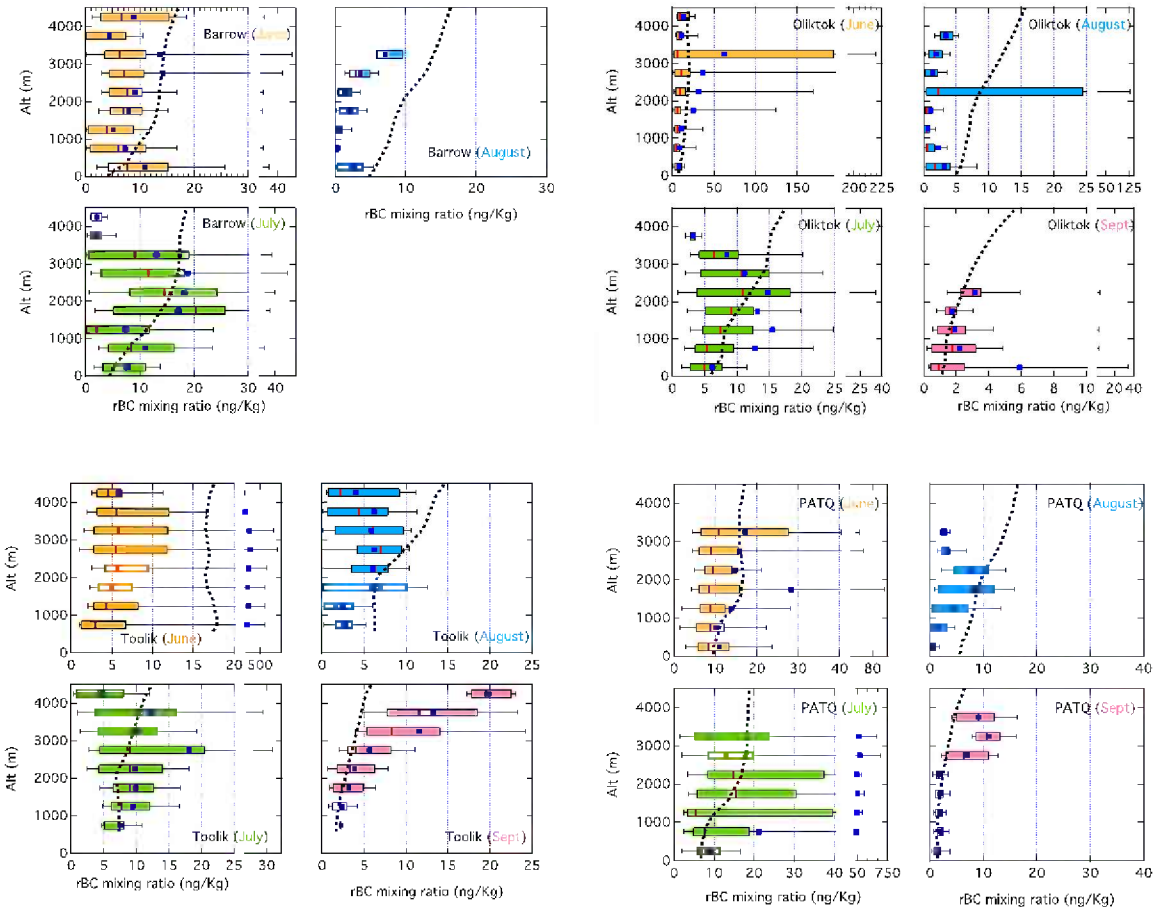

Figure 3. Box and whiskers plots of the derived monthly means, medians, and percentiles of the rBC loading along with the comparison of the CAM5 simulations with the 2011 inventory.

4.3 Example of Aerosol layering

During ACME-V several rBC profiles revealed the presence of pronounced aerosol stratification likely due to biomass burning (BB) events. The rBC mixing ratios are much higher than those monthly mean statistics shown in Figure 3. These BB observations provide opportunities to examine evolution of aerosol absorption properties and size distribution in aging. Figure 4 shows examples of this stratification.

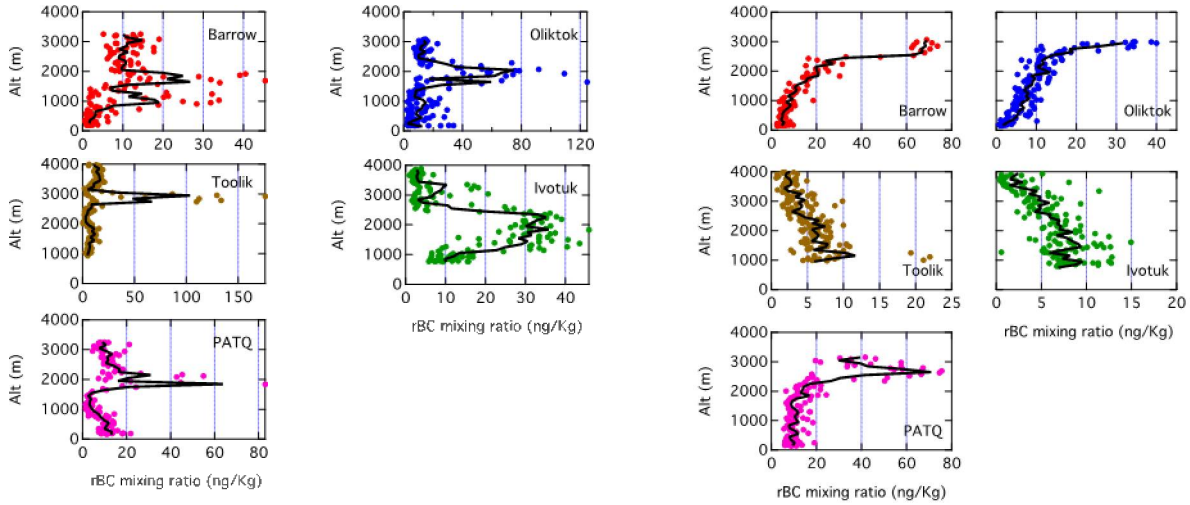

Figure 4. Examples of aerosol stratification: June 7, 2015 (left) and July 7, 2015 (right).

5.0 Public Outreach

Public outreach has thus far been limited to information dissemination through poster presentations at professional society meetings.

6.0 Publications, Presentations

6.1 Journal Articles/Manuscripts

A manuscript highlighting the availability of the ACME-V rBC data set along with a model comparison and the implications of rBC stratification is planned.

6.2 Meeting Abstracts/Presentations/Posters

FY2016

Sedlacek, AJ, Y Feng, S Biraud, and S Springston. 2016. "Vertical and spatial profiling of Arctic black carbon on the North Slope of Alaska 2015: Comparison of Model and Observation." ARM-ASR Spring Meeting, May, 2016.

FY2015

Sedlacek, AJ, Y Feng, S Biraud, and S Springston. 2015. "Vertical and Spatial Profiling of Arctic Black Carbon on the North Slope of Alaska 2015: Comparison of Model and Observation." AGU Fall Meeting, December, 2015.

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