New Instrumentation for Retrieval of UV-Visible Aerosol Optical Properties - Progress Report

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January 18, 2018

Abstract

Aerosols directly affect Earth's climate by scattering and absorbing solar radiation. Although they are ubiquitous in Earth's atmosphere, direct, *in-situ*, wavelength-resolved measurements of aerosol optical properties remain elusive. As a result, the so-called aerosol direct effects are one of the largest uncertainties in predictions of Earth's future climate, and new instrumentation is needed to provide measurements of the scattering and absorption of sunlight by atmospheric particles, especially in the UV. I have developed three pieces of equipment to address this gap in instrumentation: (1) a UV-visible broadband cavity enhanced spectrometer for the measurement of wavelength-resolved extinction from 375-700 nm; (2) a four-wavelength, single-cell photoacoustic spectrometer for simultaneous measurement of aerosol absorption at 406, 532, 662, and 785 nm; and (3) a three-wavelength, single-cell UV photoacoustic spectrometer for measurements of absorption at 320, 375, and 440 nm. Extra effort has been made to make these instruments compact, robust, and with low power requirements, all while maintaining exceptional detection limits ($<$ 1 Mm-1 of absorption/extinction). Various versions of these instruments have been used in collaborations at other laboratories to make the first measurements of the Ångström exponent (the wavelength dependence to extinction/absorption) of flame-generated soot in the UV and to measure the absorption enhancement effect of organic coatings on soot.

1 Background

Atmospheric aerosols affect Earth's climate by scattering and absorbing incoming solar radiation. Scattering aerosols tend to reflect sunlight back toward space, cooling Earth, while absorbing aerosol transfer the absorbed energy to the atmosphere as heat. Confounding the matter, most aerosols both scatter and absorb UV-visible radiation; the ratio of scattering to total extinction (scattering/(absorption + scattering)) is termed the aerosol single scattering albedo (SSA). This balance remains one of the largest uncertainties in predictions of Earth's future climate, and has been difficult to measure thus far, especially in the UV.[1]

To date, researchers have used various techniques to measure the scattering and absorption by atmospheric aerosols. Filter-based methods are widely used to measure aerosol absorption and include methods whereby particles are collected on a filter, extracted, and the extract analyzed with a UV-visible spectrophotometer, and methods wherein the sample is collected on a filter and the real-time change in light transmission through the filter is measured (e.g. an aethalometers and particle soot absorption photometers). Despite their widespread use, filter-based methods have been shown to suffer from numerous artefacts, including multiple scattering events on the filter and filter-loading effects.[2,3,4] Other scientists have used *in-situ* techniques to measure the optical properties of suspended aerosols. These include photothermal methods (photoacoustic spectroscopy and photothermal interferometric spectroscopy), which are sensitive only to absorption,[5,6,7] and the cavity enhanced spectroscopies (cavity ringdown spectroscopy and broadband cavity enhanced spectroscopy), which are sensitive to extinction (absorption and scattering summed).[8,9,10,11] These techniques are typically limited to one or two wavelengths based on available light sources. To date, few studies have been conducted on the wavelength-resolved optical properties of atmospheric aerosols, especially the UV absorption by aerosols.

The atmospheric aerosols of interest to this work are often defined by their absorption Ångström exponent (AAE). The AAE is defined as:

$$\alpha(\lambda) = \beta\lambda^{-AAE} \tag{1}$$

where α is the absorption due to aerosols (typically in Mm^{-1}), λ is the wavelength of light, β is a scaling factor, and [*AAE*] is the absorption Ångström exponent. (Note that this can also be defined in terms of extinction by reporting α in extinction (same units) and replacing the AAE with the extinction Ångström exponent, EAE.) This work is concerned with black carbon (BC) and brown carbon (BrC). BC, sometimes called soot carbon, originates from high temperature combustion of diesel fuel, coal, and biomass. It absorbs light throughout the UV-visible spectrum, and is defined by have an AAE of 1.0. Alternatively, BrC originates from low temperature combustion of biomass. It absorbs primarily UV light, with an AAE $>>$ 1 (e.g. 3-7).[12,13,14,15]

The difficulty in measuring the UV-visible optical properties of atmospheric aerosols results from low concentrations in the atmosphere and low mass absorption and extinction coefficients of many aerosols; typical ambient aerosol absorption for Athens, GA (a "clean" site) is $<$ 10 Mm^{-1}. Likewise, UV measurements are especially hindered by a lack of available instrumentation. New instruments are necessary to provide a complete picture of UV-visible optical properties. I have developed three pieces of equipment to address this gap in instrumentation: (1) a UV-visible broadband cavity enhanced spectrometer for the measurement of wavelength-resolved extinction from 375-700 nm; (2) a four-wavelength, single-cell photoacoustic spectrometer for simultaneous measurement of aerosol absorption at 406, 532, 662, and 785 nm; and (3) a three-wavelength, single-cell UV photoacoustic spectrometer for measurements of absorption at 320, 375, and 440 nm; additional earlier prototypes were also created. Extra effort has been made to make these instruments compact, robust, and with low power requirements, all while maintaining exceptional

detection limits (< 1 Mm^{-1} of absorption/extinction).

2 Goals

The primary goals of this work are to: (1) advance the instrumentation available to the aerosol community for measuring the UV-visible optical properties of atmospheric aerosols and (2) provide insight into the UV-visible optical properties of BC and BrC. This is being achieved through the development of new instrumentation and calibration methods and collaborations with other labs to measure the wavelength-resolved optical properties of BC and BrC.

There has thus far been a lack of instrumentation capable of accurately measuring the UV-visible optical properties of atmospheric aerosols. Developing new instruments is required to provide a complete picture of UV-visible optical properties, and these instruments should be specifically focused on measuring absorption in the UV. Doing so will allow for better measurements of aerosol optical properties (e.g. the SSA) and lead to more robust climate models. Likewise, there is a need for better measurements, including wavelength-resolved measurements, of BC and BrC. The instruments created here will be used in collaborations to make better measurements of these species, and thereby provide data for climate modelers to generate more accurate predictions of Earth's climate.

3 Experimental Approach

The experimental approach consists of two primary facets: (1) developing more sensitive, capable, and portable instrumentation capable of measuring ambient atmospheric aerosols and (2) applying that instrumentation to samples of atmospheric relevance. To the first regard, I have worked to develop three instruments (not including earlier versions and prototypes): (1) a UV-visible broadband cavity enhanced spectrometer for the measurement of wavelength-resolved extinction from

375-700 nm; (2) a four-wavelength, single-cell photoacoustic spectrometer for simultaneous measurement of aerosol absorption at 406, 532, 662, and 785 nm; and (3) a three-wavelength, single-cell UV photoacoustic spectrometer for measurements of absorption at 320, 375, and 440 nm. To the latter regard, I have deployed the instruments in laboratory campaigns to measure both BC and BrC (and will continue to do so). Although this work includes methods of measuring extinction, it has been primarily geared toward the measurements of absorption.

3.1 Advancing instrumentation for UV-visible aerosol optical properties

UV-visible broadband cavity enhanced spectrometer: My initial work at UGA focused on developing a 4-channel broadband cavity enhanced spectrometer (BBCES) for measuring extinction by ambient aerosols, shown in Figure 1. BBCES is similar to cavity ringdown spectroscopy, in that it uses a high-finesse optical cavity composed of two highly reflective mirrors (R $>$ 99.99%) to create an effective path length on the order of kilometers. However, BBCES uses a broadband source (e.g. an LED) and a wavelength-resolved detector (e.g. a grating spectrophotometer) to provide wavelength-resolved measurements of extinction.[16] The initial instrument developed had 4 cavities in parallel and spanned from 350-700 nm. Ultimately, this instrument has been passed on to Mike Pogash, a third-year graduate student, for further development. In its current configuration it contains 3 cavities in parallel and spans from 375-670 nm.

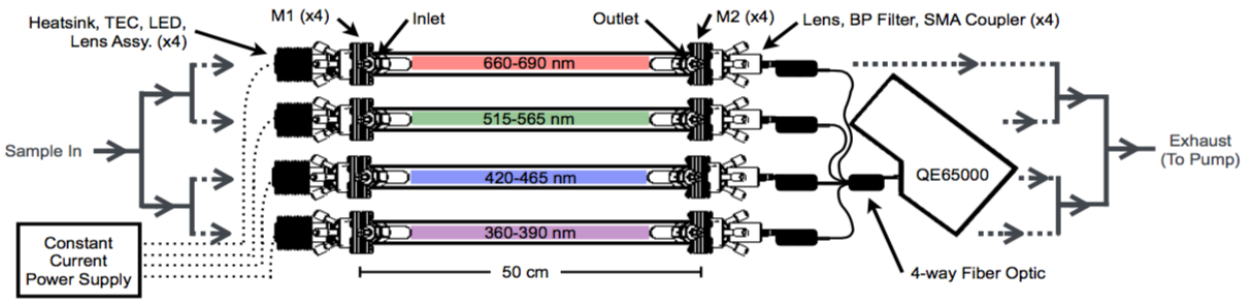

Figure 1: A schematic of the original 4-channel BBCES constructed during the early stages of my thesis work. M = mirror, TEC = thermoelectric cooler, LED = light emitting diode, BP Filter = bandpass filter.

MultiPAS, A multi-wavelength, single-cell photoacoustic spectrometer: The middle and later portions of my time at UGA have been spent developing a modular, portable photoacoustic spectrometer (PAS). The entire instrument is constructed using a commercial cage system (Thor Labs) for stability, and fits in a Pelican case for transport and protection from the elements during field deployments.[17] The first iteration of this was a 3-wavelength, single-cell, multipass photoacoustic spectrometer (termed MultiPAS-III). It employed diode lasers operating at 406, 532, and 662 nm, which were combined with a series of dichroics into a single photoacoustic cell; furthermore, the cell employs a set of highly reflective (>99%) cylindrical mirrors to create a multipass cell for enhancement of the signal.[18] This is the first time multiple lasers have been combined into a single multipass cell (traditional instruments use either multiple single-pass lasers in a single pass cell or multiple multi-pass cells, one for each laser needed). A version of this 3-wavelength PAS was constructed for Rawad Saleh in the UGA College of Engineering.

A next generation PAS, shown in Figure 2 and termed MultiPAS-IV (MP-IV), was developed following the construction of Dr. Saleh's PAS. It consists of the same cage system and PAS cell, although a fourth wavelength was added (780 nm), and the 662 nm beam was split 90% to the PAS and 10% to a cavity ringdown (CRD) spectrometer. Although the CRD provides a real-time measure of aerosol extinction (and thus the SSA at 662 nm), the primary reason for including it in the PAS was as a calibration method. Because CRD is calibration-free (assuming the background ringdown time is known),[19] and the CRD operates at exactly the same wavelength as the 662 nm PAS, and uncertainties on absorption cross sections of calibrants (e.g. NO_2) cancel out; further, it becomes unnecessary to know the concentration of the calibrant because the absorption is measured directly by the CRD and thus any errors from flow meters and dilution flows are become irrelevant.

MultiPAS-UV, A single-cell, 3-wavelength UV photoacoustic spectrometer: My most recent work has been to develop a 3-wavelength UV PAS that operates at 320, 375, and 445 nm. The design

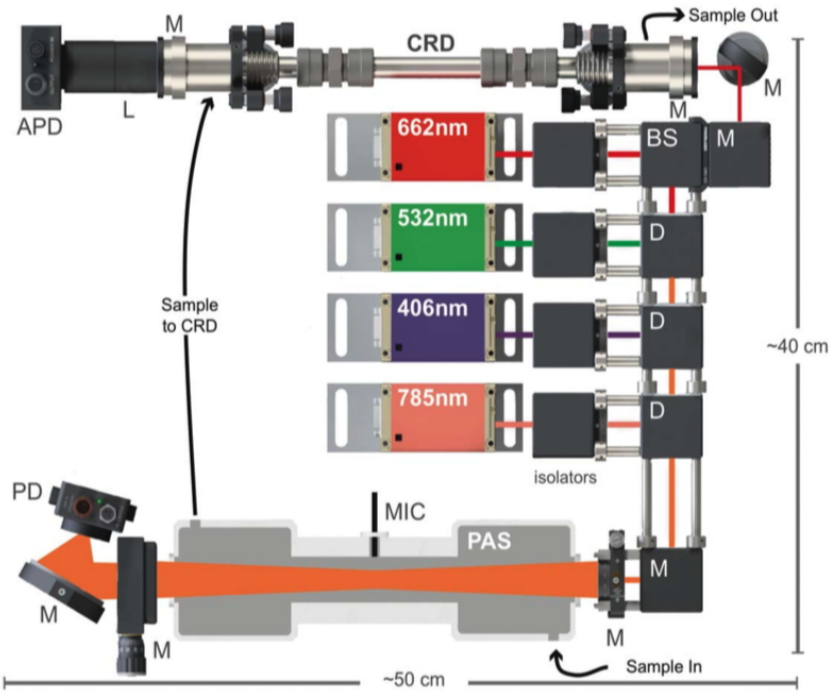

Figure 2: A diagram of the MultiPAS system, showing the 4 channel version. M = mirror, CRD = cavity ringdown, D = dichroic mirror, PAS = photoacoustic cell, mic = microphone, (A)PD = (avalanch) photodiode. Reprinted from Fischer and Smith (2017)[20]

is the same as the prior instruments, although the optics have all been replaced to access UV-wavlengths. The 445 nm point serves to provide overlapping coverage with MultiPAS-IV, which aides in combining the data both instruments to determine the AAE.

3.2 Applications of the MultiPAS Instruments

The applications of the MultiPAS instruments up to now include furthering the PAS method for the atmospheric community at large through intercomparisons and validations and furthering our knowledge of BC and BrC through lab campaigns.

3.2.1 Intercomparisons and Validations

Carbon Black Intercomparison for NIST: During the summer of 2017, we received a sample of carbon black, with optical properties unknown to us, from the National Institute of Standards (NIST). The sample was a carbon black ink manufactured by Cabot Corporation, assumed to have optical properties similar to those of BC. I measured the compound with MP-IV as part of an international blind intercomparison among aerosol PAS groups; the results of this study are forthcoming from NIST, though my preliminary data are shown in Figure 3.

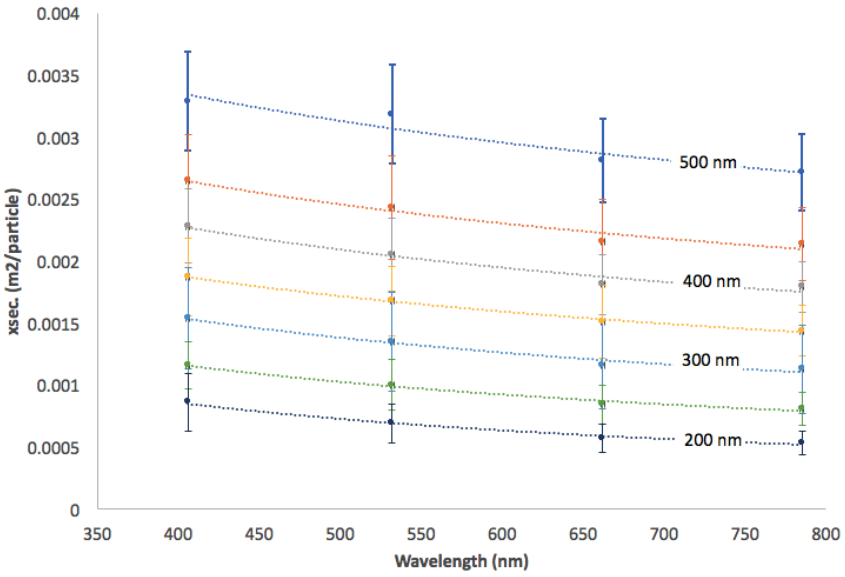

Figure 3: Preliminary carbon black spectra acquired with MP-IV for NIST. Various colors are different sizes selected with a differential mobility analyzer, ranging from 200 to 500 nm in 50 nm increments. Dashed lines are power law fits (Equation 1) to the data.

Calibration with Ozone: There is currently debate in the literature over the use of ozone for calibrating aerosol PASs.[21,22] Our group has long posited there are problems that arise when using ozone as a calibrant, primarily because ozone likely photodissociates at visible wavelengths, which would decrease the measured absorption signal due to the energy that is lost to breaking the bond. Recently, one group showed a discrepancy between ozone calibrations and particle-based calibra-

tions, but provided no explanation as to where the discrepancy came from.[23] Even more recently, another group showed agreement between ozone and particle-based calibrations.[24] My work has consistently shown discrepancies between calibrations done with ozone and nitrogen dioxide, with NO2 showing good agreement to particle measurements, as shown in Figure 4. Exploring this further is of immediate interest, and will soon be published to add to the literature discussion.

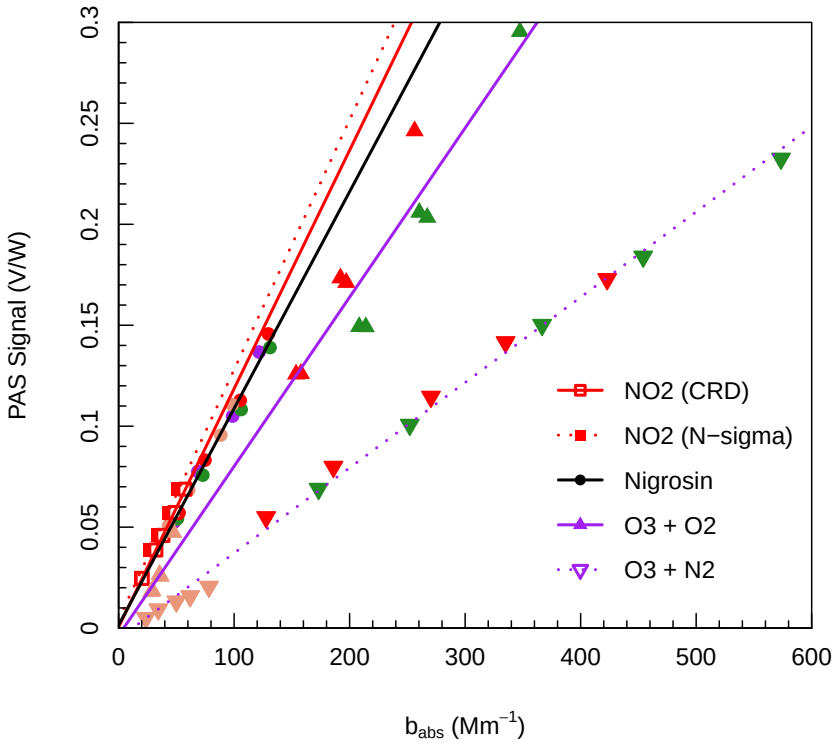

Figure 4: Calibration data collected with the MP-IV. Open red squares and solid red fit represent our preferred method of calibration, with nitrogen dioxide measured simultaneously with the PAS and CRD; this agrees within 5% of theoretical calculations of the expected absorption based on absorption cross sections (solid red squares and dotted red line). The black line and colored circles represent measurements of nigrosin, a particle calibrant, and also agree with NO2. The solid purple line and triangles represent measurements of ozone in a matrix of 100% O2, which is roughly 20% lower than NO2; the dotted purple line and inverted triangles represent measurements of ozone in 100% N2, and are clearly different from other methods of calibration. There is discrepancy about this in the literature.

3.2.2 Lab Campaigns of Atmospheric Relevance

Soot at Boston College, BC4: Prior to the construction of the MultiPAS system, and early, hybrid arc lamp- and laser-based version of the instrument (operating at 301, 314, 364, 406, 436, 546, 578, 662)[25] was sent to Boston College to measure the optical properties of flame-generated BC and the effects of non-absorption coatings on the absorption coefficient of soot. The results of this work are shown in Figure 5.

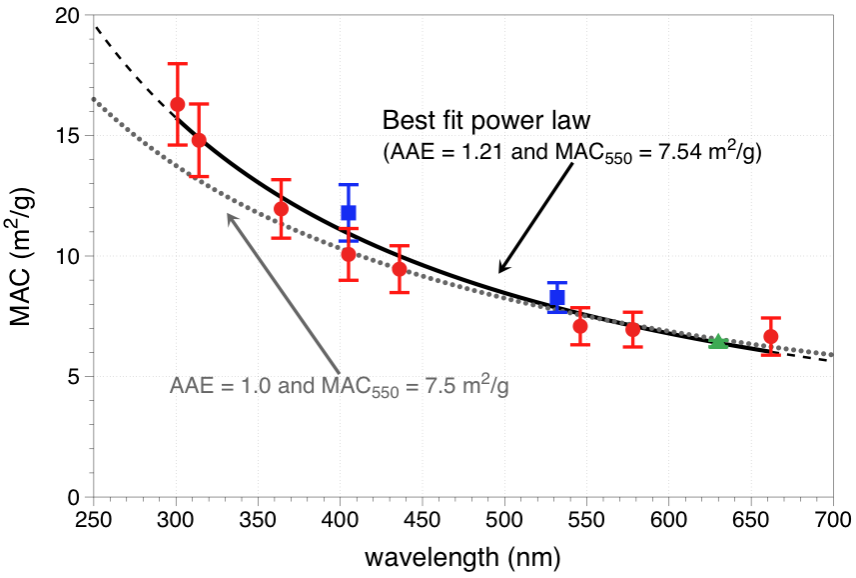

Figure 5: Spectrum of flame-generated soot acquired at Boston College, including data from our lamp-laser PAS (red circles), the UC-Davis PAS (blue squares) and a commercial cavity attenuated phase shift instrument (CAPS-SSA, Aerodyne, Inc., green triangle). The measurements agree well with literature values of the mass absorption coefficient for soot at 550 nm,[26] and an AAE of 1.0 (the assumed value in the literature) fits the visible data well. However, significant deviation is observed in the UV, enough to bring the AAE to 1.21 (solid line). Plot by Dr. Geoff Smith, reproduced from Fischer et al. (2018 - in preparation, see below).

Brown Carbon & the AE-33: We are currently in collaboration with Rawad Saleh in the UGA College of Engineering to compare measurements of lab-generated BrC with those of a commercial 7-wavelength aethalometer (AE-33 by Magee Scientific). The aethalometer is a widely applied and well-accepted "push-button" instrument used to measure the absorption by aerosols at 370,

470, 520, 590, 660, 880, and 950 nm. However, there are many relatively unvalidated assumptions underlying the measurements, and sources have noticed sample- and wavelength-dependent correction factors when the aethalometer is compared to other instruments.[27,28,29] However, no instrument has yet been able to match the wavelength coverage of the aethalomter to fully vet the instrument. Combined with Dr. Saleh's PAS, we will have measurements at 375, 406, 422, 445, 532 (x2), 662, 780, and 782 nm that will allow us to fully characterize the wavelength dependence of aethalometer correction factors. Further, the aerosol generation capabilities in Dr. Saleh's lab will allow us to make measurements over a range of conditions to explore the sample-dependence of these correction factors. Not only will this encourage more accurate measurements by groups using the aethalometer, but it will also provide wavelength-resolved, *in-situ* measurements of BrC. These experiments have been started, and are currently ongoing. Figure 6 shows a summary of preliminary data from these experiments

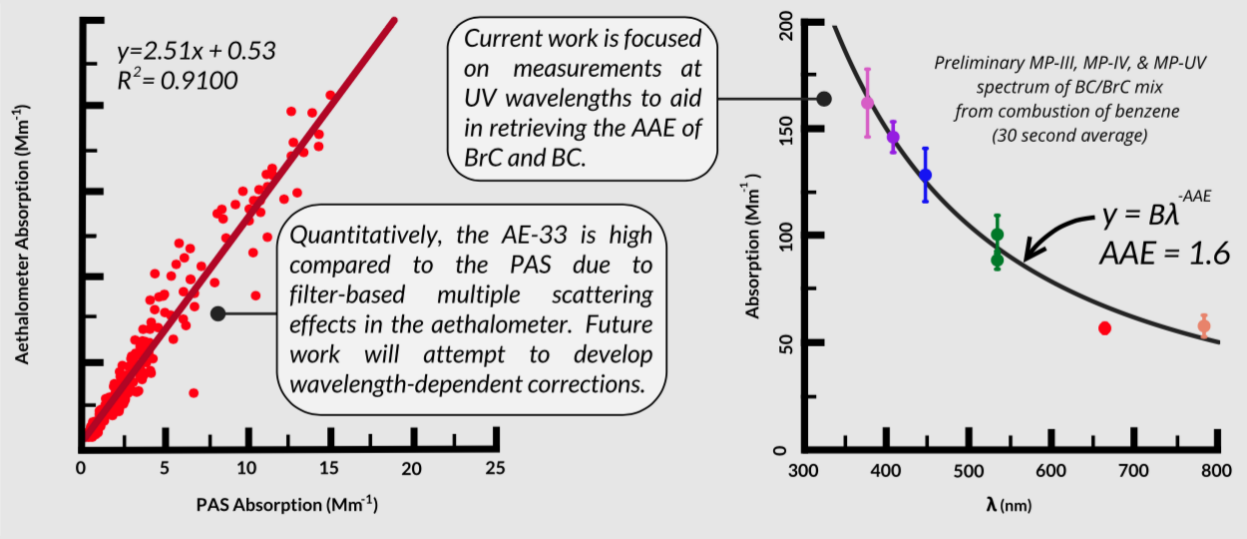

Figure 6: Comparison of the Magee Scientific AE-33 7-wavelength aethalometer to MP-IV at 660/662 nm with ambient aerosols in Athens, GA (left) and the UV-visible spectrum of a mix of black and brown carbon generated in the lab of Dr. Rawad Saleh and obtained with MP-IV, MP-UV, and MP-III simultaneously (right).

3.3 Progress Report

Much of my progress has been described above. In summary, I have:

- Developed three instruments, a broadband cavity enhanced spectrometer, a 4-wavelength visible photoacoustic spectrometer, and a three wavelength UV photoacoustic spectrometer; an additional version of the PAS, a 3-wavelength visible PAS was constructed at the request of Dr. Rawad Saleh, UGA College of Engineering, and delivered to his lab where it is currently in use by his students.
- Used these instruments in atmospherically relevant lab campaigns to measure black carbon and brown carbon, in international PAS intercomparisons, and in studies of calibrants for PAS.

Published and planned peer-reviewed manuscripts related to this work:

- **Fischer, D.A.** and G.D. Smith (2017) A Portable, 4-Wavelength Photoacoustic Instrument for Ambient Aerosol Absorption. *Aerosol Science and Technology.* DOI: 10.1080/02786826.2017.1413231 (*In press*)
- **Fischer, D.A.**, T. Helgestad, L. Renbaum-Wolff, A. Lambe, S. China, C. Mazzoleni, A. Sedlacek, C. Cappa, A. Freedman, T. Onasch, P. Davidovits, and G.D. Smith (2018) UV-visible Absorption Spectrum of Laboratory-generated Soot Particles. *Aerosol Science and Technology.* (*In Preparation*)
- **Fischer, D.A.** and G.D. Smith (2018) *Atmospheric Measurement Techniques*. On the Use of Ozone for Calibrating Photoacoustic Spectrometers. Atmosphere. (*In Preparation*)
- **Fischer, D.A.** and G.D. Smith (2018) *Atmospheric Measurement Techniques*. The Spectral Dependence of the Aethalometer Multiple-scattering Correction Factor, C_s. (*In Preparation*)

Selected presentations and posters related to this work:

*indicates presenting author

- **Fischer, D.A.*** and G.D. Smith. (2017) UV-Visible Photoacoustic Spectroscopy. American Association for Aerosol Research Annual Conference, Raleigh, NC.
- L. Renbaum-Wolff, **D.A. Fischer**, T. Helgestad, A. Lambe, G. Smith, C. Cappa, A. Sedlacek, P. Davidovits, T. Onasch, and A. FREEDMAN*. (2016) Measurements of Soot Mass Absorption Coefficients from 300 to 660 nm. European Geophysical Union General Assembly 2016, Vienna, Austria. EGU2016-9236.
- **Fischer, D.A***. and G.D. Smith. (2015) A UV-Vis Broadband Cavity Enhanced Spectrometer for Ambient Aerosols. Eleventh International User Meeting and Summer School on Cavity Enhanced Spectroscopy, Boulder, CO.
- **Fischer, D.A.*** and G.D. Smith. (2013) Incoherent broadband cavity enhanced spectroscopy for measuring extinction coefficients of atmospheric species throughout the UV-visible spectrum. Southeast Regional Meeting of the American Chemical Society. Atlanta, GA.

3.4 Future Directions

My work on this project is nearing completion. Of immediate concern are:

- Further validating MultiPAS-UV. This is our newest instrument and still has bugs, including some calibration issues related to accurately measuring the power of each of the three wavelengths. This is a current focus of my work.
- Understanding the use of ozone as a calibrant. There is current discussion on this in the literature. I am currently conducting experiments to better understand why discrepancies exist between ozone and other calibrants.
- Fully validate the AE-33 aethalomter. I am currently collaborating with Rawad Saleh's stu-

dents to validate their AE-33 aethalomter, a commercial instrument. Although this work has been started, calibration issues forced is to put it on hold; it will be resumed shortly.

- Although the BBCES is still under development, although the project has been turned over to Mike Pogash, a third-year graduate student. Future directions for that project are dependent on him.
- Make measurements with the PAS and BBCES simultaneously to retrieve an effective refractive index of ambient atmospheric aerosols. (tentative)

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