Statement of Work

AQRP Project 14-005

Sources and Properties of Atmospheric Aerosol in Texas: DISCOVER-AQ Measurements and Validation

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Abstract

Tropospheric air quality is degraded by local aerosol sources and gas phase precursors as well as aerosol transported over long distances. While the availability of recent satellites such as the Moderate-resolution Imaging Spectroradiometer (MODIS) and the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) offer improved accuracy and global coverage of aerosol, such measurements still rely on broad assumptions in determination of aerosol source and composition. During the fall of 2013, the Houston area was the site of the 2nd field intensive of the National Aeronautics and Space Administration (NASA) Deriving Information on Surface conditions from COlumn and VERtically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) campaign. During DISCOVER-AQ, we operated a new scattering instrument, the Cloud and Aerosol Spectrometer with Polarization (CASPOL) which measures the depolarization ratio of individual particles in the aerosol population. The polarization capabilities of CASPOL facilitate an effective approach to validate spaceborne aerosol retrieval, particularly CALIOP aerosol type classification. The CASPOL was operated on top of the 60 m tall Moody Tower (MT) on the University of Houston campus, a central urban location and site of many complementary measurements during DISCOVER-AQ. We now propose to analyze the CASPOL data set to determine the concentration, size distribution, and optical properties of aerosol from the wide variety of sources including urban pollution sources from downtown Houston and the industrial Ship Channel, and transported aerosol. Combined with additional measurements of organic carbon, black carbon and ozone, the CASPOL data set provides an opportunity to determine the primary aerosol sources and impacts of aging and possible changes in aerosol optical properties due to exposure to ozone. Conversely, highly absorbing black carbon aerosol loadings may also modulate ozone concentrations by reducing photolysis rates. In addition to improving understanding of ozone-aerosols feedbacks, in-situ data will be compared to MODIS and CALIOP aerosol measurements to determine the sensitivity of remote sensing to changes in surface aerosol properties and air quality. The project proposed here will improve the linkage between column observations provided by satellite instruments and near-surface atmospheric composition which is relevant to air quality and human health in the short term and the relationship between future air quality and climate.

This proposal is directly in line with two targeted areas of AQRP research, specifically:
1. Improving the understanding of ozone and particulate matter formation, and quantifying the characteristics of emissions in Texas through analysis of data collected during the DISCOVER-AQ campaign.
2. Analysis of field campaign and other appropriate data to investigate transformations of gas phase pollutants to particulate matter impacting Texas air quality, and to identify the sources of particulate matter (PM) pollution impacting Texas.

Technical Work Plan

Task 1. 1 Analyze CASPOL data collected during DISCOVER-AQ.

We operated the CASPOL instrument continuously from Aug 28 through October 4, 2013 from the top the 60 m tall Moody Tower (MT) on the University of Houston campus as part of DISCOVER-AQ. The CASPOL provides in-situ determination of aerosol concentration, size distribution, and depolarization ratio. Validation of remote sensing measurements of aerosols will be accomplished through in-situ measurements of aerosol size distributions, type, and particle-by-particle determination of depolarization ratio. We call special attention to the fact that in situ measurements by CASPOL provide valuable datasets for validating satellite-based aerosol property retrieval. From this perspective, the proposed research effort will have quite substantial broader impacts.

The coordinated Moody Tower (MT) measurements during DISCOVER-AQ provide time-resolved, 24-hour measurements of key trace gases and aerosols including ozone, carbon monoxide, nitrogen oxide and dioxides, and sulfur dioxide concentrations, aerosol optical depth, boundary layer height and basic meteorological parameters during the diverse conditions encountered in a coastal urban center in close proximity to multiple industrial sources of pollution. The height of MT is sufficient to reduce influence from nearby intermittent sources and yet is be more representative of surface air quality conditions in the Houston area than concurrent aircraft measurements.

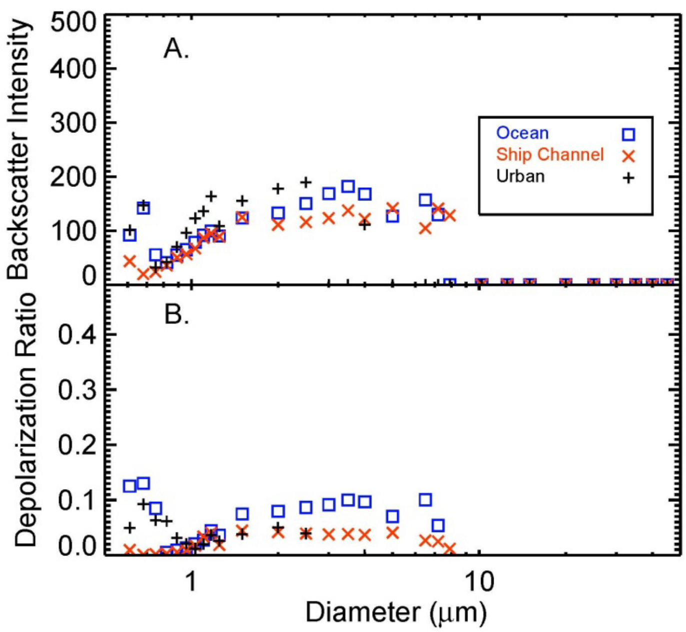
In addition to ongoing measurements at MT, Dr. Rebecca Sheesley of Baylor University operated a seven-channel aethelometer and Particulate Matter with diameter of 2.5 µm and less ($PM_{2.5}$), and Total Suspended Particulates (TSP) filter samplers collocated with the CASPOL throughout the DISCOVER-AQ period. The mass loading of soot in particles and a first order estimate of degree of coating on soot particles will be determined by aethelometer data and the Organic Carbon to Element Carbon ratios (OC/EC) provided by Dr. Sheesley. In addition, the University of Houston operated a sun photometer during DISCOVER-AQ. This instrument, part of the Aerosol RObotic Network (AERONET) [*Schuster et al, 2012*], measures radiances at a set of visible and near-infrared wavelengths to determine column aerosol optical depth. Comparison to this sun photometer data and to the AERONET data in general will provide additional insight on horizontal and vertical variability in aerosol populations.

IB. CASPOL Instrument Details.

The CASPOL instrument determines the concentration, size distribution and optical properties for individual particles in a range of sizes, from 0.6 µm < diameter < 50 µm, using a diode laser at a wavelength of 680 nm. The CASPOL measures light scattered by individual aerosol particles in the forward (4° to 12°) and backward (168° to 176°) directions utilizing three detectors. The forward scattering detector (*f*) provides the size measurement of the sampled particles, the two backward scattering detectors, $\beta_{\parallel}$, and $\beta_{\perp}$, measure the parallel and perpendicularly polarized scattered light respectively. In addition to providing measurements of aerosol number concentration and size distributions in real-time (one minute averages), the CASPOL measurements can be used qualitatively to indicate aerosol type and quantitatively to derive lidar backscattering and depolarization signals at the wavelength of the CASPOL laser (680 nm). Using the two backscattering detectors, the aerosol depolarization ratio (ADR) at 680 nm is calculated for each particle size bin according to:

$$ADR = \frac{\beta_{\perp}}{\beta_{\parallel}}$$

To illustrate CASPOL response to aerosol populations of differing type, the backscatter and aerosol depolarization ratio as a function of particle size (determined by the forward scattering detector), preliminary CASPOL data collected under conditions of downtown Houston (Urban), transported aerosol arriving from the Gulf of Mexico (Ocean), and from an air mass from the ship channel (Ship Channel) are shown in Figure 1. (Source locations are identified by Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) back trajectories as discussed below.)



As can be seen, the transported aerosol population (Ocean) contained particles with much higher depolarization rations than either the Urban or Ship Channel cases. The two local sources are also clearly different than one another. First, the aerosol population from the Ship Channel contains much larger particles, and secondly, submicron particles from the Urban case are highly depolarizing, whereas those from the ship channel have depolarization ratios near zero. In summary, variation in aerosol types present in the Houston environment can be observed and classified by the CASPOL. CASPOL measurements will be used along with other aerosol composition measurements at the Moody Tower to provide an indication of aerosol type present in Houston throughout the DISCOVER-AQ campaign.

Task 1 Deliverable:* *At the end of Task 1, all CASPOL data will be quality controlled and separated by class according to source. Data collected during and after precipitation events will be eliminated, as will any periods during which the CASPOL was operating offline for maintenance, drying, or flow testing. A set of files will be produced, 2 for each day. File

format for the first file will include CASPOL time, total particle number, size distribution. The second file for each day will contain single particle backscattering, and depolarization data, which will be required to generate optical signature plots in Task 2 below.

The due date for Task 1 will be Feb. 27, 2015.

Task 2. Identification of Aerosol Types in the CASPOL DISCOVER-AQ data set

A. Classification of aerosols

First, we will classify sampling periods according to source locations using 5-day HYSPLIT back trajectories [*Draxler and Rolph,* 2003]. Because the CASPOL measures forward and backscattering and depolarization ratio of individual particles, the relationship between aerosol properties (concentration and type) and backscattering intensity and depolarization ratio can be accurately constrained for a variety of aerosol types including those shown in Figure 1 above as well as transported continental background and biogenic aerosols and mineral dust. After initial classification, we will evaluate whether the CASPOL data are unique for each aerosol source type. If unique optical signatures are observed, aerosol classification in future work may rely directly on classification by the CASPOL.

B. Refinement of Classification within urban type

The impacts of pollution on aerosol are complex [*Ma et al,* 2013]. We anticipate that coincident gas phase pollutants will modulate the aerosol properties. One impact of pollution on aerosols is that gas phase ozone interacts with soot aerosol, increasing its hygroscopicity. Hence aged (oxidized) particles will take up water and may be present as solution droplets at ambient relative humidities. (The aerosols may, in turn, reduce ambient ozone concentrations by absorbing light). Ozone data collected at the Moody Tower by researchers from the University of Houston, will be used to classify the CASPOL ozone exposure level. Ozone has been observed to age soot particles when exposed to ozone in concentrations of 30 ppbv or more [*Pöschl et al,* 2001]. Hourly averages of ozone concentration will be identified as low (0 to 33 ppbv), medium (33 to 66 ppbv) or high (66 to 100 ppbv). In addition, OC/EC ratios will be classified as high (>10) or low (<10) CASPOL data collected under each of those classifications. A typical urban value for OC/EC is 6 [Seinfeld and Pandis, 2006]. If the ozone and OC/EC levels are both low, the aerosols will be assumed to be fresh. If the ozone concentration is high and the OC/EC ration is above 10, the aerosols will be classified as aged. For any other ozone concentrations and OC/EC ratios, the aerosol will be assumed to be partially aged.

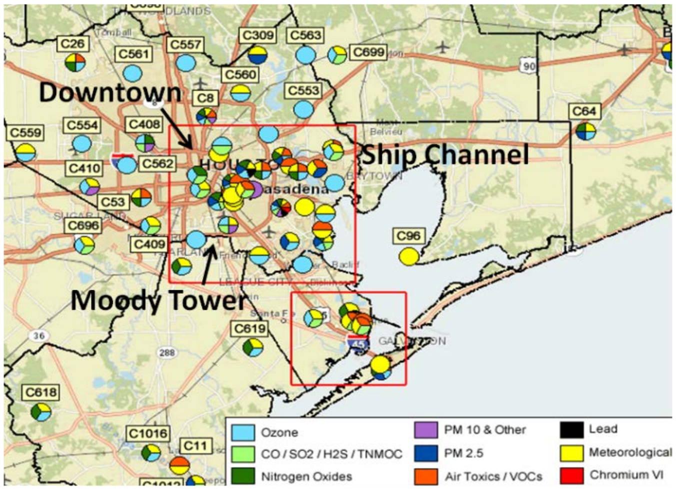


Figure 2. Daily averaged air quality data for October 30, 2013. TCEQ data is available http://www.tceq.texas.gov.

As can be seen in Figure 2, ozone and $PM_{2.5}$ are monitored at a large number of Continuous Air Monitoring Stations (CAMS) within the Greater Houston Area. PM_{10} is also monitored at a few stations in the region. Hourly averages of TCEQ mass measurements of $PM_{2.5}$ and PM_{10} (blue and purple slices of the pie charts in Figure 2, respectively) will be compared to the CASPOL data throughout the project to evaluate how representative the aerosol sampled by CASPOL at Moody is of the ambient aerosol in the Greater Houston Area.

Task 2 Deliverable: HYSPLIT backtrajectories will be run for all quality controlled CASPOL DATA.

All CASPOL data will be sorted into categories, i.e. urban pollution, industrial pollution from the Ship Channel, or transported aerosol, according to the source regions identified by HYSPLIT. Further categorization will be conducted based on data from additional in-situ measurements. A summary table designating in which category data from all time periods belongs will be produced. Data from each time period will be collected in a single file for each category. From these files, CASPOL data from will be used to generate optical signature plots (backscattering vs. depolarization) for the specified category. Plots will be inspected for characteristic differences which may allow source region identification directly from CASPOL in future studies.

The due date for Task 2 will be Feb. 27, 2015.

Task 3. Comparison of in-situ aerosol properties to satellite-retrieved properties during DISCOVER-AQ

The main objective of the NASA DISCOVER-AQ mission is to improve pollution monitoring from satellites for better air quality forecasting and warning systems and aerosol source identification. While aerosol retrievals were performed for a number of different satellite instruments during DISCOVER-AQ, we will build on the strengths and experiences of the proposal team and focus on the MODIS instrument operated onboard the Terra and Aqua satellites and CALIOP on the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite, which flies in formation with Aqua.

The MODIS instrument is a radiometer that measures reflected solar and emitted terrestrial radiation in 36 bands between 0.4 and 14.4 µm. MODIS is a scanning instrument with a 2030 km swath which means that each MODIS instrument gets a daytime view of Houston with a favorable viewing angle at least every 3 out of 4 days. Because MODIS retrievals are based on reflected solar radiation, they are only made during the daytime. MODIS retrieves aerosol properties by comparing measured reflectances to modeled reflectances from a wide range of aerosol simulations. The modeled reflectances depend on a number of assumptions about aerosol composition, size distribution, and absorption and scattering properties, as well as environmental conditions including surface properties, atmospheric profile, and viewing geometry. The impacts of these assumptions and their global applicability are not certain. For example, our proposal team has shown that neglecting non-sphericity of dust and certain pollutant particles can lead to underestimations of aerosol optical depth (AOD) [*Feng et al.*, 2009].

Aerosol retrievals from passive sensors such as MODIS are very sensitive to cloud contamination. Essentially, AOD cannot be retrieved by satellites in the presence of cloud cover. This means that although the DISCOVER-AQ campaign took place over many days, there are a limited number of cases with widespread aerosol retrieval coverage over Houston from MODIS on Aqua, and several more days with limited coverage over the Greater Houston area. Because the Houston area is heavily urbanized, the surface is relatively reflective. The "Dark Target" MODIS aerosol retrievals have been shown to have decreased accuracy over large urban areas, but there is uncertainty as to whether this is due to surface reflectivity or the aerosol models used in the retrievals [*Levy et al.*, 2010]. Of particular interest for this project will be the upcoming release of the newest version of MODIS aerosol product, Collection 6 [*Levy, et al., 2013*, 2013], which will include the "Dark Target" retrievals as well as the "Deep Blue" retrievals [*Sayer*, 2013] that were designed to work over bright surfaces. The Collection 6 Dark Target retrievals will be available at standard 10-km resolution, as well as an improved 3-km resolution product, while the Deep Blue retrievals will be at 10-km resolution. The Collection 6 properties retrieved over land will include total aerosol optical depth at 0.47, 0.55, and 0.65 µm and qualitative information about aerosol type and size parameters.

CALIOP, the lidar instrument on the CALIPSO satellite, provides a vertical profile of attenuated backscattered radiation at two wavelengths, 532 and 1064 nm. In addition, the depolarization ratio (ratio of the perpendicular component to parallel) is profiled at 532 nm. CALIOP backscatter profiles and depolarization signatures make it possible to discriminate between spherical and nonspherical particles, such as dust and liquid aerosol droplets, and water droplets and ice crystals [*Winker*, 2010] and are used in the creation of the CALIPSO cloud and aerosol layer products [*Vaughan*, 2009;]Cho et al, 2008]. Figure 3 shows the CALIOP depolarization ratio to backscatter relationships for each of CALIOP's six retrieved aerosol types: clean marine, dust, polluted continental (urban), clean continental (remote background aerosol), polluted dust

(dust which has travelled away from the source region) and smoke (from biomass burning). Aerosol optical depth retrievals by CALIOP depend on the assumed aerosol type model and comparisons with the ground-based **AE**rosol **RO**botic **NET**work (AERONET) measured optical depths depended on scene type [Schuster, 2012]. AERONET and CALIOP had the greatest AOD differences for the dust scenes.

Because CALIOP has the ability to vertically profile backscatter and depolarization ratio, comparisons between it and the CASPOL are highly desired. However, CALIOP is not a scanning instrument which means that it has limited coverage. During the CASPOL measurement period, CALIOP passed within 35 km of Moody Tower on three daytime occasions (around 17:45 UTC on 8/31, 9/16, and 10/02) and within 83 km on three nighttime occasions (around 8:25 UTC on 9/4, 9/11, 9/20, and 9/27). The nearest overpass to the Moody Tower measurement site was 29.5 km away on 8/31.

Task 3 Deliverable:

A. Identification of time periods in which MODIS and in-situ data are collocated.

B. Identification of time periods in which CALIOP and in-situ data are collocated.

C. Identification of in-situ time periods for representative aerosol types and conditions.

The due date for Task 3 will be May 29, 2015.

Task 4. Comparison of in-situ data and remote sensing products

By inspection of the collaborated data sets, we propose to answer the following questions about MODIS and CALIOP retrievals

1. How sensitive are MODIS retrievals over the Houston Area to aerosol type?

First, we address the question of whether uncertainty in the MODIS aerosols retrievals over urban areas is due to aerosol assumptions or surface reflectivity. We will use aerosol data available across the full region to give each MODIS retrieval (day and pixel) a predominant aerosol type based on assumptions from each of the MODIS algorithms from Collection 5 and Collection 6 and meteorological condition classification. Then, we will compare the MODIS aerosol optical depth and aerosol type assumptions to AERONET optical depths and CASPOL aerosol typing. Next, we will look specifically at MODIS pixels that include Moody Tower to determine if variability detected by the CASPOL is reflected in the MODIS retrievals. If so, this will enable future use of the CASPOL data to improve MODIS aerosol models.

2. How do the CASPOL depolarization ratios and aerosol typing compare with CALIOP?

CALIOP data can be processed to produce remotely sensed optical signature plots, which are nearly identical as the CASPOL signature plots. While CASPOL and CALIOP both provide information about aerosol backscatter and depolarization ratio, there are distinct differences between the measurements. For example, CASPOL provides information about individual particles, while CALIOP retrieves averaged information over 5 km horizontally and approximately 30 m vertically. Additionally, the CALIOP retrievals are based on a number of assumptions about aerosol lidar ratio and depolarization ratio for aerosol types. The CASPOL measurements provide us an exciting and novel opportunity to evaluate the CALIOP aerosol assumptions and propose improved aerosol type models.

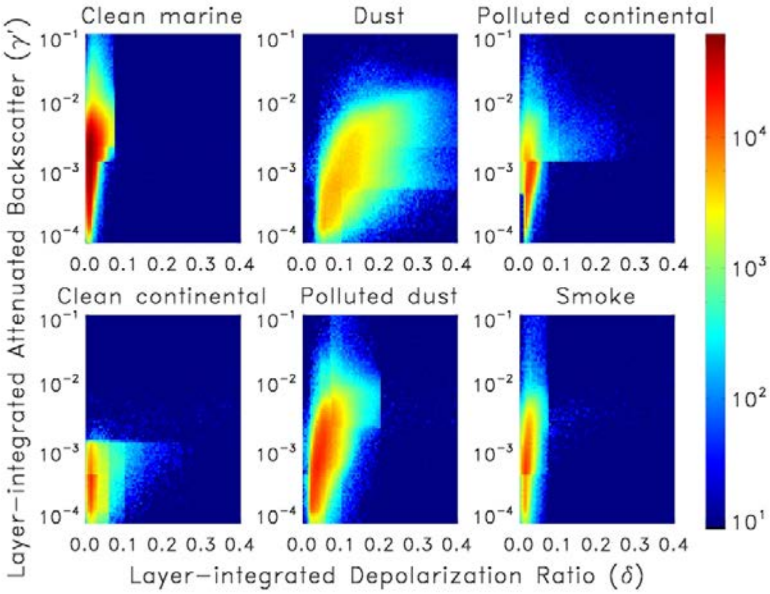


Figure 3. The CALIOP lidar depolarization-backscatter relationships (at 532 nm) for six aerosol types, from the CALIPSO Cloud and Aerosol layer product. ***[H-M Cho, 2011: Studying clouds and aerosols with lidar depolarization ratio and backscatter relationships. Ph.D. thesis, Texas A&M University*****]. Global observations during the entire year of 2008 are included. The color of each pixel presents the frequency of occurrence for a depolarization-backscatter box with 0.01 by 0.002 sr^{-1} interval.**

Task 4 Deliverable: A summary of MODIS aerosol optical depth and aerosol type for each for the cases by the CASPOL. CALIOP remotely sensed optical signature plots for each case identified in the CAPSOL data. A report summarizing which variations observed in-situ data can and which cannot be observed in remotely sensed data sets.

The due date for Task 4 will be Aug. 15, 2015.

Task 5. Final publication and dissemination of results.

Task 5 Deliverable. A presentation of results accomplished during the first two quarters will be made at the June 2015 AQRP. Workshop. Also, a final publication of results will be prepared and submitted to the Journal of Geophysical Research or Aerosol Science and Technology.

The due date for Task 5 will be Nov. 30, 2015.

5. Summary Schedule and Administrative Deliverables:

An **executive summary** describing the project will be submitted by Jan. 9, 2015.

Quarterly reports on the topics below will be submitted prior to the following due dates.

Feb. 27, 2015 Task 1. Post- DISCOVER-AQ CASPOL calibrations and quality control
Task 2. Identification of Aerosol Types in the CASPOL data set

May 29, 2015 Task 3. Comparison of in-situ aerosol properties to satellite-retrieved properties during DISCOVER-AQ

Aug. 15, 2015 Task 4. Comparison of in-situ data and remote sensing products.

Nov. 30, 2015 Task 5. Final publication and dissemination of results.

June, 2015 A presentation of results accomplished during the first two quarters will be made at the June 2015 AQRP Workshop.

Technical reports and **financial status reports** will be submitted monthly to the Project Manager and TCEQ Liaison as a Word doc using the AQRP FY14-15 MTR Template.

Specific dates for the technical reports are as follows:

Report	Period Covered	Due Date
Technical Report #1	Project Start – February 28, 2015	Monday, March 9, 2015
Technical Report #2	March 1 - 31, 2015	Wednesday, April 8, 2015
Technical Report #3	April 1 - 28, 2015	Friday, May 8, 2015
Technical Report #4	May 1 - 31, 2015	Monday, June 8, 2015
Technical Report #5	June 1 - 30, 2015	Wednesday, July 8, 2015
Technical Report #6	July 1 - 31, 2015	Monday, August 10, 2015
Technical Report #7	August 1 - 31, 2015	Tuesday, September 8, 2015

Specific dates for the financial status reports are as follows:

Report	Period Covered	Due Date
FSR #1	Project Start – February 28, 2015	Monday, March 16, 2015
FSR #2	March 1 - 31, 2015	Wednesday, April 15, 2015
FSR #3	April 1 - 28, 2015	Friday, May 15, 2015
FSR #4	May 1 - 31, 2015	Monday, June 15, 2015
FSR #5	June 1 - 30, 2015	Wednesday, July 15, 2015
FSR #6	July 1 - 31, 2015	Monday, August 17, 2015
FSR #7	August 1 - 31, 2015	Tuesday, September 15, 2015
FSR #8	September 1 - 30, 2015	Thursday, October 15, 2015
FSR #9	Final FSR	Monday, November 16, 2015

A **draft final report** and a **final report** will be submitted on Aug. 18 and Sept 30, respectively.

6. Research Experience and Expertise at TAMU Atmospheric Sciences

The Department of Atmospheric Sciences at Texas A&M is home to 20 faculty members with a broad range of research expertise. State of the art computing and laboratory facilities are available for this project. CASPOL calibrations will be conducted in Dr. Brooks' 1040 sq. ft. laboratory in the department using a Topaz Solid Aerosol Generator 410, Vibrating Orifice Aerosol Generator, TSI atomizers, and a light duty torch for soot generation. Additional aerosol equipment includes a TSI Electrostatic Classifier, TSI Condensation nuclei counter, GRIMM SMPS, GRIMM 1.108 Aerosol Spectrometer, and cascade impactors. We can use a supercomputer at the Supercomputer Institute, Texas A&M University, as well as existing workstation computers in our groups to download and analysis MODIS and CALOP data, develop light scattering computational models, to organize/store the scientific results/data, and to write project reports and papers.

Sarah Brooks is an associate professor at the Department of Atmospheric Sciences, Texas A&M University. She has expertise in instrument development and measurements of the physical, chemical, and optical properties of aerosols, and has worked on a large number of field campaigns, including the TEXAS-AQ2 and DISCOVER-AQ measurements conducted at the Moody Tower in Houston, Texas (Wright et al., 2009; Atkinson et al., 2010). Dr. Brooks was honored with the Presidential Early Career Award for Scientists and Engineers (PECASE), the highest honor bestowed on outstanding young scientists by the U.S. government in 2007 and an NSF CAREER award in 2006. For this project, she will serve as the principal investigator and coordinate all the aspects of the proposed research.

Ping Yang is Professor and Head of the Department of Atmospheric Sciences, Texas A&M University. He has expertise in developing and evaluating satellite remote sensing algorithms for aerosols and clouds and in theoretical light scattering and radiative transfer. He has worked extensively with the MODIS and CALIPSO satellites and the relevant data products. He has published 241 peer-reviewed journal articles, 9 book chapters and 2 textbooks. For this project, he will lead the interpretation of satellite retrievals and comparisons with in-situ data and will use his recently developed computational model to calculate the single-scattering properties of realistic aerosol shape models to interpret the CASPOL depolarization data.

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