Chapter 21

Aerosol Physical and Optical Properties and Processes in the ARM Program

ALLISON MCCOMISKEY

Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, and NOAA Earth System Research Laboratory, Boulder, Colorado

RICHARD A. FERRARE

NASA Langley Research Center, Hampton, Virginia

1. Introduction

At the inception of the Atmospheric Radiation Measurement (ARM) Program, anthropogenic aerosols were becoming widely recognized for their influence on radiation transfer in the atmosphere and their role in climate change. The seminal article by Charlson et al. (1992) estimated the global mean aerosol radiative forcing by anthropogenic sulfate to be $-2\,\mathrm{W\,m^{-2}}$. At the same time, Penner et al. (1992) estimated a similar forcing by biomass burning aerosol under the assumption that it is a primarily light-scattering aerosol. Despite the recognition that there would be an attendant effect from absorbing aerosol, no estimate could be made for lack of knowledge of their mass, optical properties, and distribution. While containing substantial uncertainty, these estimates of cooling by aerosol are of a magnitude that could offset the warming caused by increases in greenhouse gases over the industrial period.

The tenet of the ARM Program at the outset was to improve the representation of properties and processes impacting atmospheric radiation in climate models, with an emphasis on clouds and water vapor. The prevailing approach was a hierarchical method of examination of processes that rested at its base on experimental verification in the form of radiative closure at the surface. The structure of ARM measurements was designed to implement this process evaluation and radiative closure. Historically, the inclusion of a uniform background tropospheric aerosol was considered adequate for closure studies. However, improved knowledge of source strengths of anthropogenic aerosols came with the realization that aerosol loading and optical properties are highly variable both seasonally and geographically. It became evident that inclusion of the local aerosol was required for closure studies and for the characterization of regional and global impacts necessary for improved understanding of climate change. Thus, it became necessary for ARM to address more realistic aerosol contributions to atmospheric radiation transfer.

With a complexity rivaling that of clouds, the characterization of local- to regional-scale aerosol properties would be a challenge to ARM and the larger community, requiring a sound scientific plan and experimental resources. Calling on those at the forefront of aerosol research, ARM formed the Aerosol Working Group (AWG) to develop research questions and recommend a suite of measurements that would allow for the characterization of relevant aerosol properties. The scope of this effort was defined in terms of the pertinent processes, properties, and measurable quantities for aerosol radiative forcing outlined in Charlson et al. (1992) and with regard to the major objectives of ARM (Fig. 21-1).

Several research objectives were developed to guide measurement needs, which encompassed aerosol–radiation interactions in both clear and cloudy skies. These included understanding relationships among cloud condensation nuclei concentrations, cloud droplet concentrations, and droplet size; defining relationships between aerosol chemical and physical properties (size

Corresponding author address: Allison McComiskey, NOAA Earth System Research Laboratory, Global Monitoring Division, 325 Broadway, Boulder, CO 80305.
E-mail: allison.mccomiskey@noaa.gov

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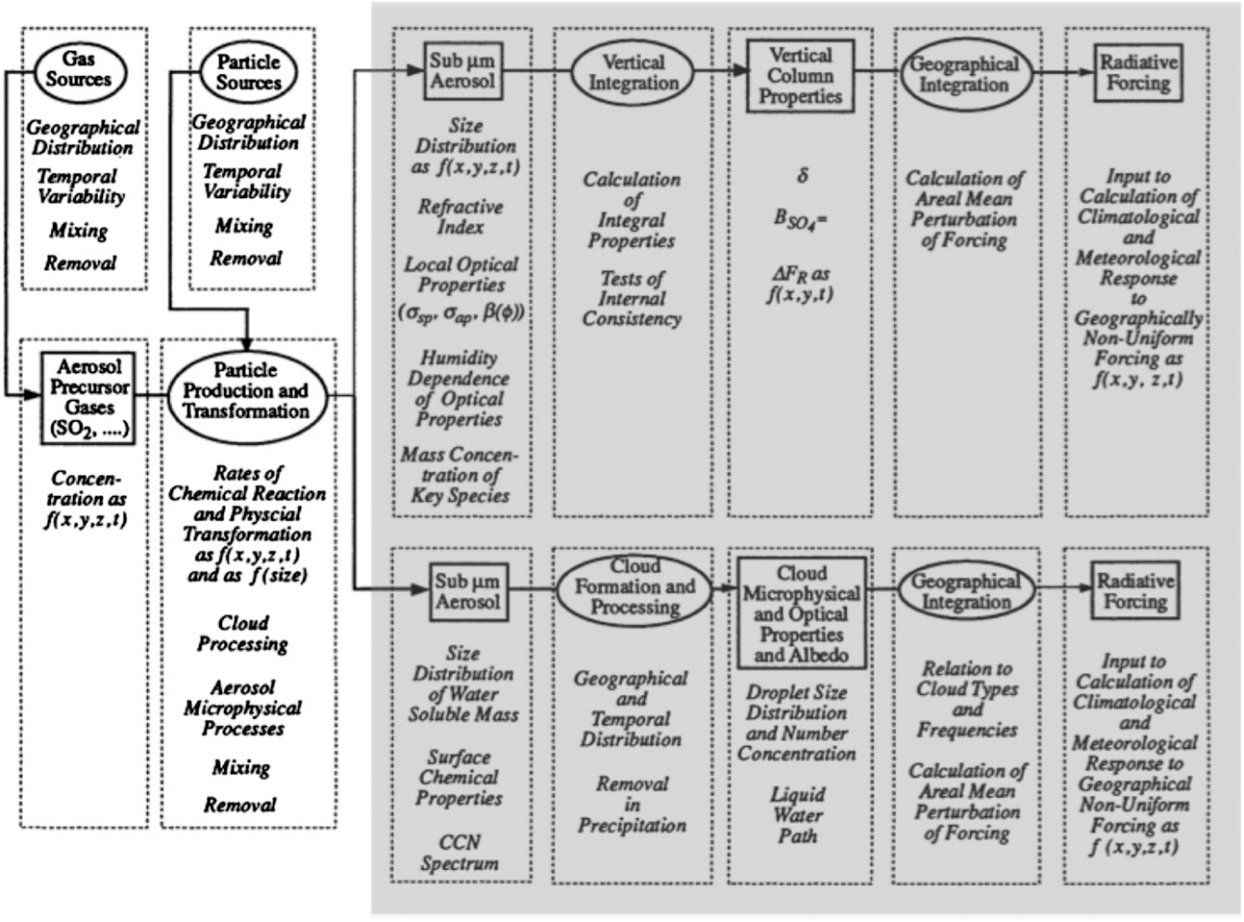

FIG. 21-1. Modified from Charlson et al. (1992, their Fig. 4). The ARM Aerosol Working Group identified the properties and processes in the gray box as related to the larger scope of the ARM Program, which then suggested the priority measurements to be made.

distribution, composition, morphology) and optical properties; defining relationships of aerosol optical and microphysical properties to radiative properties; providing parameterizations for climate models; and defining uncertainties and drivers of variability in these properties and processes and their importance for atmospheric radiation. These objectives were reflected in the suite of recommended measurements that would allow for characterization of instantaneous and temporally averaged aerosol properties within the full column for determination of radiative impacts on a range of scales.

The first ARM measurements were made at the Southern Great Plains (SGP) site in 1992 but, despite the best laid schemes of the AWG, no systematic measurements of aerosols were made until 1996. Given the clear need for inclusion of aerosols in closing the radiation budget and the obvious importance of aerosols on the climate it may seem surprising that ARM did not invest in these measurements at the outset. At this time, focus was placed on the measurement of clouds and clear-sky radiative processes. Understanding cloud–radiation processes was such a challenging effort that, with only so many resources available to the program, tension developed between the clouds and water vapor researchers and those interested in aerosol impacts. The latter—who soon became known as "the dirt guys"—was a group who could potentially absorb valuable program resources. However, in time, the plans from the 1992 AWG became reality and the first Aerosol Observing System (AOS) was built for deployment at the SGP in 1996.

Over the past two decades ARM has provided prodigious support for aerosol measurements and theoretical research contributing to understanding the role of aerosols in climate change in various regimes. While building climatologies was not the original intent of the program, these data are invaluable for characterizing trends and variability in aerosol properties and their radiative forcing. The ARM investment in this discipline continues to grow in step with the desire to address the increasing complexity as climate models become more

TABLE 21-1. Fundamental aerosol properties, measurements, and recommended instrumentation determined by the ARM Aerosol Working Group 1992 for improved understanding of aerosol radiative forcing in clear and cloudy skies. Some measurements were recommended for continuous long-term sampling at the ARM fixed sites while other more complex measurements were recommended for intensive periods only.

	Property	Measurement	Instrument
Local microscale	Size distributed chemical composition	Particle composition in two size classes (0.1–1 μm; 1–10 μm)	Filter samplers; impactor
		Total mass; major ions; elemental composition, organic and elemental carbon	Electrobalance; ion chromatography, particle-induced X-ray emission spectrometry (PIXE), volatilization/oxidation, CO_2 or CH_4 analysis
	Size distributed number concentration	Size distribution 0.01–5 μm (coarse resolution); size distribution 0.001–10 μm (high resolution)	Optical particle counter (OPC); CN counter (CPC) Differential mobility analyzer (DMA), aerodynamic particle sizer (APS)
	Size distributed number concentration as a function of relative humidity	Relative humidity profile Hygroscopic growth	Tandem differential mobility analyzer (TDMA); humidity-controlled nephelometer
	Size distributed cloud drop number concentration		
Local integrated	Scattering coefficient	3 wavelengths 450–850 nm; low reference humidity; as a function of scattering angle	Integrating nephelometer; polar nephelometer
	Backscattering coefficient	3 wavelengths 450–850 nm; low reference humidity	Integrating nephelometer with backscatter shutter
	Absorption coefficient		Filter deposit light attenuation
	Scattering coefficient as a function of relative humidity		
	Size distributed cloud condensation nucleus as a function of supersaturation	CCN at single supersaturation CCN spectrum	Thermal diffusion cloud chamber
Column	Optical depth	Multispectral optical depth Aerosol vertical distribution	Radiometer Lidar
	Absorption optical depth Backscattering optical depth		
	Cloud optical depth	Optical depth at several wavelengths Cloud-base height	Sun photometer, shadowband radiometer Ceilometer
	Cloud albedo		Narrow and broadband radiometers

detailed and we strive to represent climate processes with greater accuracy. This chapter outlines the history of these achievements.

2. Measurements and value-added products

The AWG presented to ARM a list of necessary properties to be measured, associated observational characteristics, and recommended instrumentation to meet the stated research objectives (Penner et al. 1992). These are summarized in Table 21-1. Properties were organized by local integrated, size distributed, and column properties and addressed both clear-sky and cloudy-sky conditions. They included measurements to be made continuously at the fixed ground sites as well as more complex measurements for airborne intensive observation periods (IOPs). The column properties were largely already being addressed by the program in relation to meeting existing objectives through ground-based remote sensing by lidar and shortwave radiometry (Michalsky and Long 2016, chapter 16) and the surface properties would provide further characterization of aerosol properties that cannot be easily gained from remote sensing.

FIG. 21-2. (left) The original Aerosol Observing System (AOS) at the SGP in 1996. (right) Updated AOS racks at the SGP. Courtesy of John A. Ogren.

While the program has invested in developing and improving retrievals of pertinent properties from remote sensing at the surface, airborne in situ measurements in campaign mode are still critical for understanding aerosol properties and processes. Primary properties that are measured routinely at the surface are aerosol light scattering, backscattering, absorption, number concentration, hygroscopicity, size distribution, and chemical composition. Some properties can only be understood in detail through in situ measurements; however, the need for improved remote sensing capabilities is recognized in order to extrapolate knowledge of the finescale nature of aerosol to their regional- to global-scale radiative impacts. Aerosol optical depth at several wavelengths and backscatter and depolarization profiles from active remote sensing are measured routinely at all sites; at the SGP and Tropical Western Pacific (TWP) Darwin sites aerosol extinction profiles also are measured using the Raman technique. Through the years, as discussed below, the aerosol program has focused increasingly on the synergy between in situ and remote sensing techniques for better characterization of aerosols throughout the atmospheric column and representations in models.

a. The Aerosol Observing System

To address a subset of the measurement needs defined by the AWG, an integrated system of instruments for measuring aerosol microphysical and optical properties in situ at the surface was developed by the U.S. Department of Energy (DOE) Environmental Measurement Program (Leifer et al. 1993). The instruments were housed in an exclusive, self-contained shelter with an intake stack that sampled at 10 m above the ground to avoid surface turbidity. A common manifold supplied all instruments with the same sample air. The system included five instruments: a condensation particle counter (CPC), optical particle counter (OPC), single wavelength nephelometer (550 nm), particle soot absorption photometer (PSAP; 565 nm), and three-wavelength nephelometer with backscatter shutter (450, 550, and 700 nm). The system design provided space for expansion in the future. A 10-μm impactor limited the aerosol size sampled by each of the instruments exclusive of the OPC. The first AOS was deployed at the SGP in 1996 and has been making measurements continuously to the present day (Fig. 21-2).

Operation and development of the SGP AOS was assumed by the ARM-funded instrument mentors at the National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (now the NOAA Earth System Research Laboratory Global Monitoring Division), and within a year several upgrades were initiated. A switched impactor system was installed in 1997 to sample at two size cuts

(<1 μm and <10 μm) (Sheridan et al. 2001). Sample air is gently heated when necessary to provide measurements at a "dry" (<40%) relative humidity (RH), avoiding evaporation of volatile species while maintaining a reference state that is comparable to similar measurements made in any conditions at other times or locations. A second three-wavelength nephelometer was added in 1998 with an associated humidograph system to measure the change in aerosol scattering with water uptake. The system incrementally increases the RH of the sample over a 30-min period from 40% to ~85% RH. In 2000, monitoring of aerosol chemical composition by filter sample and ion chromatography began and continued until 2008 (Quinn et al. 2002). In 2005 a humidified tandem differential mobility analyzer (HTDMA), designed and deployed by Texas A&M (Santarpia et al. 2004), was installed in the AOS shelter for size distributions and hygroscopic growth. The PSAP was upgraded from a one- to a three-wavelength instrument and in 2006 a cloud condensation nucleus counter (CCNC) was added (Roberts and Nenes 2005).

At the time the AOS was established at the SGP, NOAA CMDL had been operating an aerosol measurement system at the NOAA Barrow, Alaska, observatory since 1976. In 1997, joint contributions from NOAA and DOE ARM were made to upgrade, maintain, and operate this site as the NOAA Barrow/ARM North Slope of Alaska (NSA) AOS system. Recreating the design and instrumentation from the SGP has allowed quantitative comparison of aerosol properties in these two distinct aerosol regimes. The SGP and NSA data records, both longer than 15 years now, constitute the two long-term records of aerosol properties within DOE. These are valuable for their insight into seasonal variability and long-term trends in distinct regimes and for deeper probing into processes that dictate aerosol optical properties and radiative transfer.

The first new fixed-site AOS for long-term monitoring in 15 years has now been established in a remote marine environment on Graciosa Island in the Azores. This system, mentored by the DOE Brookhaven National Laboratory, will mimic previous AOS but with expanded capabilities for measuring aerosol-size distribution, chemical composition, and gases. The Aerosol Chemical Speciation Monitor (ACSM; Ng et al. 2011) is being deployed for understanding episodic influences of long-range transport of dust and urban/industrial aerosol over the northern Atlantic. The site has a scientific focus on aerosol–cloud interactions, and researchers will take advantage of the synergy of surface measurements with remote sensors to understand the relationships of aerosol at the surface, cloud base, and within cloud.

b. *In situ aerosol profiles*

Knowledge of the extent to which aerosol sampled at the surface is representative of properties in the full column above is critical if these long-term surface observations are to be used to determine aerosol radiative forcing. Long-term, continuous observations are generally relegated to the surface where monitoring is logistically feasible. As previously stated, this approach to monitoring is indispensable for characterizing geographical and seasonal or shorter-term variability in aerosol as well as long-term trends. However, full column properties that dictate radiative fluxes at the surface and top of the atmosphere are often not well correlated to properties at the surface (Andrews et al. 2004). Intensive observations are typically made over short durations providing highly comprehensive characterization of aerosol properties in the column but do not resolve geographical and temporal variability. In 1998 the AWG met in Oak Ridge, Tennessee, and originated a concept for continuous yet cost-effective monitoring of the vertical profile of aerosol properties in clear skies. Robust statistical characterization of the vertical profile of aerosol properties comparable to those measured at the surface would be performed over the SGP for several years.

The long-term campaign called the In Situ Aerosol Profiles (IAP; Andrews et al. 2004, 2011) experiment flew a Cessna 172 two to three times a week over the SGP ground site beginning in 2000. Observations were made from an airborne version of AOS, with <1-μm light scattering and backscattering at three wavelengths and light absorption at one wavelength at <40% RH. During each flight, level legs were flown at nine altitudes (465, 610, 915, 1220, 1525, 1830, 2440, 3050, and 3660 m) above mean sea level during daylight hours and in clear-sky conditions. In 2005, an upgrade to a Cessna 206 Turbo was made and the instrument package was updated to measure light absorption at three wavelengths (467, 530, and 660 nm) plus dry scattering at <5 μm. The number of flight levels increased to 12 and the maximum altitude increased to ~4575 m. Figure 21-3 presents the measured light scattering contoured over the full IAP deployment showing the distinct annual cycle but with variability within this cycle among years. The IAP was a first, showing that long-term, routine aircraft sampling could be accomplished in a cost-effective and efficient manner and characterized the vertical structure of aerosol optical properties with greater than seven years of regular data at SGP.

c. *ARM Mobile Facility*

Aerosol chemical and microphysical properties, and thus radiative forcing, exhibit a geographical dependence

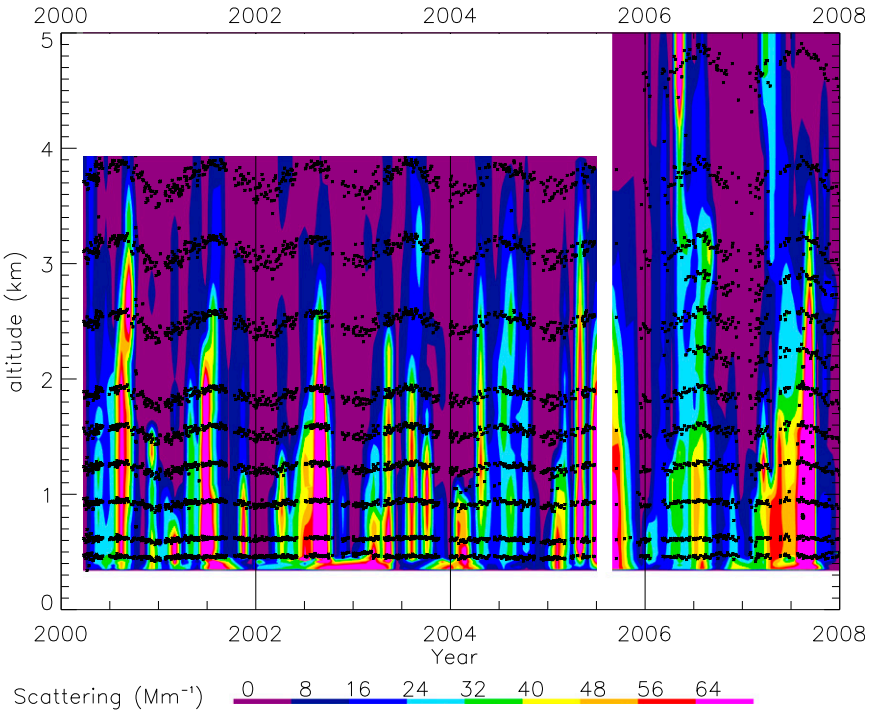

FIG. 21-3. Contoured light scattering from the nephelometer during the IAP campaign. The black dots represent individual flight segments at their altitude above mean sea level. From Andrews et al. (2011, their Fig. 1).

as a function of meteorology and proximity to different sources. Characterization of the regional variability in chemical, physical, and radiative properties as well as process understanding related to different aerosol types requires measurements in locales outside of the ARM fixed sites. To explore scientific questions outside those represented at ARM's long-term, fixed sites, an ARM Mobile Facility (AMF) was developed in 2005 for deployment around the world for 6- to 18-month periods (Miller and Slingo 2007). The AMF was equipped with an AOS system that contained the baseline instruments: CPC, nephelometer, humidified nephelometer, PSAP, and CCNC. This system was encompassed in a self-contained, portable container with a collapsible inlet stack and shock-mounted instrument racks for safe transport to any location chosen by ARM for deployment. Since 2005, the AMF has been deployed in distinct aerosol and meteorological regimes including Point Reyes, California; the Black Forest, Germany; Niamey, Niger; Shouxian, China; the Ganges valley, India; Graciosa Island, Azores; and Cape Cod, Massachusetts. As a component of the larger AMF, the AOS is always accompanied by active and passive remote sensors that provide information on the column aerosol, cloud properties, and thermodynamic state of the atmosphere, allowing for comprehensive investigations of aerosol radiative impacts in clear and cloudy conditions.

While the first AMF (AMF-1) has contributed greatly to characterizing aerosol variability over land surfaces, there has long been recognition that marine aerosol–cloud–radiation interactions are critical because of the vast area they represent, but also that these regions are highly undersampled. A second marine-capable AMF (AMF-2) was built mimicking the design of the AMF-1 but with expanded capabilities including an HTDMA. Its first deployment in 2010 was land-based in Steamboat Springs, Colorado, but since then the AMF-2 has spent nearly a year traveling the Pacific Ocean between Los Angeles, California, and Honolulu, Hawaii, sampling a gradient of aerosol between these urban/industrial centers and the remote marine environment as part of the Marine ARM GPCI Investigation of Clouds (MAGIC) Campaign [Lewis and Wiscombe 2012; GPCI is the GEWEX Cloud Systems Study (GCSS) Pacific Cross-section Intercomparison].

In 2009 the American Recovery and Reinvestment Act (ARRA) provided for additional infrastructure that has allowed ARM to delve deeper into processes controlling aerosol radiative properties. Aerosol optical properties are essentially a function of aerosol composition, size, and morphology, which are related to both

sources and sinks and the intervening chemical and physical processing. Knowledge of how composition (source) is related to the processes that determine optical properties is critical for representing radiative forcing with fidelity in climate models. The fixed sites and two mobile facilities provide a baseline set of measurements from which aerosol optical properties have been well characterized. Combined with a greater understanding of how these properties are related to composition, size, and morphology will contribute to improved representation of aerosol processes in climate models. The Mobile Aerosol Observing System (MAOS) was developed by Brookhaven National Laboratory to serve as an accompaniment to either ARM fixed sites during intensive campaigns or mobile facilities with an aerosol focus. The MAOS comprises two separate containers, one containing the baseline aerosol instrumentation (MAOS-A) of the standard AOS with extended measurements of aerosol size distributions, chemical composition, and aerosol morphology while the second container, MAOS-C, contains enhanced chemistry measurements.

d. ARM Aerial Facility

Despite the heart of the ARM Program being located on the ground, the need for airborne measurements was fully recognized from the beginning. For aerosol research, critical applications include characterization of the vertical structure of aerosol properties, support for development and evaluation of remote sensing retrievals from the ground and space, and measurements of properties in situ that can advance understanding of aerosol life cycle processes not always accessible from the ground. In the latter case, understanding aerosol properties at cloud base and interstitial aerosol within cloud are essential for characterizing aerosol–cloud interactions, aerosol convective transport, and aerosol sinks. The ARM Aerial Facility (AAF) supports both routine observations such as the IAP program as well as airborne components of intensive campaigns. As emphasis on studying aerosol–cloud interactions increased in the program, the Routine AAF CLOWD Optical Radiative Observations (RACORO) Campaign (CLOWD = Clouds with Low Optical Water Depths), a second routine airborne program, was conducted over six months in 2009 on the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter to provide statistically robust datasets relevant to the properties of low liquid water boundary layer clouds and their relationship to aerosol (Vogelmann et al. 2012). Primary aerosol measurements made were cloud condensation nuclei (CCN) concentrations and aerosol size distributions above and below cloud.

Campaigns with significant airborne aerosol emphasis include the Indirect and Semi-Direct Aerosol Campaign (ISDAC) from the NRC Convair to better understand cloud and aerosol processes in Arctic mixed-phase clouds (McFarquhar et al. 2011) and the Carbonaceous Aerosols and Radiative Effects Study (CARES), a coordinated multiagency campaign in California to examine the aerosol processes and properties resulting from interactions of urban and biogenic emissions (Zaveri et al. 2012; Shilling et al. 2013). During CARES the DOE G-1 was heavily instrumented with an ARM suite of instrumentation that was expanded through ARRA and also with many guest instruments. The benefits of this comprehensive suite of instruments are evident in many findings from the campaign related to the evolution of secondary organic and black carbon aerosol and their climate impacts near a major urban center (e.g., Cahill et al. 2012; Moffet et al. 2013).

e. Remote sensing of aerosol properties

1) PASSIVE REMOTE SENSING

Broadband and spectral radiometry supported by ARM is described by Michalsky and Long (2016, chapter 16) along with a review of the utility in determining aerosol optical depth. The multifilter rotating shadowband radiometer (MFRSR; Harrison et al. 1994) has long played a key role in providing aerosol properties for applications concerning aerosol radiative impacts. This narrowband radiometer measures the total and diffuse horizontal components of the downwelling solar irradiance in seven wavelength bands and produces the direct normal component by difference. Langley calibrated measurements of the different irradiance components made by the same sensor allow for retrievals of optical depths with reduced uncertainties relative to using several different instruments calibrated by standard lamps. Together with the normal incidence multifilter radiometer (NIMFR), accurate (± 0.01) climatologies of aerosol optical depth have been produced at ARM sites continuously since 1992 (Michalsky et al. 2010). ARM's extensive use of MFRSRs is advantageous for linking with the large global networks of radiometry used to measure aerosol radiative properties such as the National Aeronautics and Space Administration (NASA) Aerosol Robotic Network (AERONET).

Harrison and Michalsky (1994) presented an automated algorithm for determining aerosol optical depth time series from the MFRSR with potential accuracy to 0.003 for averaging periods of 1 to 5 min. Subsequent work has extended and refined calibration and retrieval methods for the MFRSR providing for accurate

retrievals under a larger range of conditions such as partly cloudy skies, which would result in enhanced climatologies, and estimates of additional properties such as effective particle size (Alexandrov et al. 2002a,b; Kassianov et al. 2005; Alexandrov et al. 2007). Kassianov et al. (2007) used an iterative approach with the direct irradiance and direct-to-diffuse ratios to simultaneously retrieve single-scatter albedo and asymmetry parameter. While this technique requires assumptions about the aerosol-size distribution and inputs for surface albedo, it produces the suite of first order optical properties required for calculating aerosol radiative fluxes or forcing integrated over the column. These data are useful for producing climatologies as well as evaluating independent measures of these properties from in situ, and other active and passive space- and ground-based remote sensing approaches. Validation of this procedure at the SGP resulted in radiative closure with direct and diffuse irradiances to within $5\,W\,m^{-2}$. The algorithms have been brought together in the Column Intensive Properties (CIP) Value-Added Product (VAP), which is now operational and available on the ARM Data Archive for the SGP and is planned for other sites.

2) Active remote sensing

While passive remote sensing from radiometers provides column-integrated aerosol and surface property inputs adequate for many applications, it cannot provide information on the vertical structure of these properties known to have a significant impact on aerosol radiative transfer and atmospheric heating. Short of in situ measurements from aircraft, active remote sensing provides the only method for obtaining this information and is the only method for providing continuous, long-term measurements of vertical profiles. One of ARM's greatest accomplishments has been to produce continuous operational measurements of profiles of aerosol properties and water vapor from the Raman lidar (Turner et al. 2016, chapter 18). Beginning in 1998, the Raman lidar at the SGP site has collected continuous profiles of aerosol backscattering, extinction, and depolarization during daytime and nighttime operations excluding only expected downtimes for instrumental issues and maintenance. These measurements were used to develop and study mean and seasonal water vapor and aerosol extinction profiles over this site (Turner et al. 2001) as well as to study the vertical variability of aerosol above the SGP site (Ferrare et al. 2001). Peppler et al. (2000) demonstrated the utility of the Raman lidar in tracking the vertical distribution of forest and brush fire plumes from Central America over SGP with its ability to provide both aerosol extinction profiles and discrimination of biomass burning from background aerosol through extinction/backscatter ratios.

Several efforts outlined in the following sections have revolved around these measurements to better understand the relationship between lidar retrievals of aerosol properties and those measured by different approaches. Another Raman lidar with similar aerosol and water vapor measurement capabilities was installed at the TWP Darwin site and became operational in 2010. In 2011, a High Spectral Resolution Lidar (HSRL) was deployed at NSA for long-term operation in the Arctic. As with the Raman lidar, the HSRL provides vertical profiles of optical depth, scattering cross sections, and depolarization and is absolutely calibrated, which reduces the need for assumption about aerosol scattering properties for retrievals. Currently, HSRLs reside at the NSA and AMF-2 sites. A very recent addition includes three Doppler lidars enabling the 3D mapping of aerosol properties.

f. Modeling, quality measurement experiments, and value-added products

Early modeling activities associated with ARM provided information where measurements were lacking. Empirically based estimates of large-scale aerosol radiative forcing were impossible due to the dearth of observations of various anthropogenic species. Many studies extrapolated from known properties of sulfate (Charlson et al. 1992; Nemesure et al. 1995; Nemesure et al. 1997), biomass burning (Penner et al. 1992), and other absorbing aerosol (Nemesure and Schwartz 1998) to provide global or hemispheric estimates. The primary modeling objective stated in the original AWG report in 1992 was to examine consistency between measured aerosol size distribution and chemical composition and the associated scattering and absorption coefficients and CCN concentrations—the relationships on which all of these estimates depend. Today, much of the DOE Atmospheric System Research Program (ASR) Aerosol Life Cycle Working Group effort is dedicated to refining our understanding of these relationships. Later, as datasets became more developed and extensive, they could be used for direct evaluation of model performance; for example, the evaluation of the AeroCom models' ability to reproduce aerosol profiles as measured by the Raman lidar at SGP (Ferrare et al. 2005).

Despite the comprehensive instrumentation deployed by ARM, many quantities required in understanding the atmospheric system cannot be measured directly. Rather, these geophysical quantities of interest must be derived from direct measurements of different quantities, often with the benefit of models. Processing and compilation of these data into functional, coherent, and

easily accessible datasets is a routine activity in ARM resulting in numerous VAPs. A special class of VAPs, called Quality Measurement Experiments (QMEs), was developed to identify issues with and improve the quality of existing measurements. QMEs are long time series (multiyear) radiative closure experiments; the first was for spectral longwave radiance using the Atmospheric Emitted Radiance Interferometer (AERI) and led to improvements in specification of model inputs for atmospheric state variables (Mlawer and Turner 2016, chapter 14). This success led to the Broadband Heating Rate Profile (BBHRP) effort that facilitates closure in the longwave and shortwave for all broadband components of radiative fluxes (McFarlane et al. 2016, chapter 20).

Expansion of the QME concept to address shortwave clear-sky fluxes required aerosol and surface albedo inputs, and thus the Aerosol Best Estimate (ABE) VAP was developed. The objective of ABE was to provide vertical profiles of aerosol extinction, single-scatter albedo, and asymmetry parameters (Turner et al. 2005). Climatologies of vertical profiles of extinction are taken from Raman lidar and used to extrapolate properties measured at the surface through the vertical column (Sivaraman et al. 2004; Flynn et al. 2012). Efforts to improve spectral measurements of surface albedo, critical for accuracy in modeling aerosol effects, are discussed by Michalsky and Long (2016, chapter 16). Inclusion of ABE in BBHRP did show improvement in simulating shortwave radiation at the surface; however, the exercise raised questions as to whether aerosol properties were extrapolated adequately in the vertical and across the spectrum to the ultraviolet and near-infrared where measurements were not made (Delamere et al. 2008).

In some cases, measurement–model comparisons pointed to instrumental problems with the radiometers themselves and contributed to improvements and higher-quality measurements at ARM sites over the long term. The bulk of the error, however, derives from the vertically resolved profiles of the aerosol single scatter albedo and asymmetry parameters, which are not currently determined from remote sensing or surface-based in situ measurement approaches. The Raman lidar provides profiles of extinction and backscattering very well, but quantifying the vertical profiles of aerosol absorption, size, and humidification effects required for understanding the effects of aerosol on atmospheric fluxes and heating requires additional measurements. Currently, ABE simply carries values for single-scatter albedo and asymmetry parameters measured at the surface up through the column. Efforts to derive profiles of each of the relevant properties require a compilation of various data sources and significant assumptions, an area of active effort as more sophisticated measurements become available and the sophistication of retrieval algorithms improves concordantly. Further, efforts to retrieve profiles of aerosol properties directly using multiwavelength Raman and HSRL lidars show great promise (e.g., Müller et al. 2001, 2014) and are being pursued actively in the larger community.

3. Long-term and intensive aerosol characterization at ARM sites and beyond

a. 1994–97 aerosol IOP at SGP

Despite the AOS not being fielded at the SGP until 1996, interest in aerosol measurements was evident from earlier campaigns such as the 1994 Remote Cloud Study IOP. Aerosol backscattering and extinction profiles as well as aerosol intensive properties (real refractive index, single-scattering albedo, and humidification factor) were derived from measurements acquired by the NASA Goddard Space Flight Center scanning Raman lidar and aircraft in situ measurements of size distribution; these represented nighttime aerosol at heights between 0.1 and 5 km (Ferrare et al. 1998). Characterization of the aerosol extinction and backscattering profiles as well as optical depths were examined in comparison to tower-mounted nephelometer measurements, daytime sunphotometer measurements, and airborne size distributions. Similarities and differences seen among these various approaches provided information on the strengths and weaknesses of each that served to drive measurement and retrieval science of aerosol properties into the future.

By 1997, investments had been made in the AOS and a suite of IOPs was conducted at the SGP to address many topics related to aerosol radiative forcing. Much of the work focused on intercomparison of measurement approaches for various aerosol properties to provide a better understanding of these approaches, their utility, and their limitations, in some cases spurring improvements in measurements and retrieval algorithms. Kato et al. (2000) performed a comprehensive comparison of airborne in situ scattering and absorption measurements in the column to ground-based radiometry including the MFRSR, Cimel sunphotometer, and Raman lidar. It was found that, under dry conditions the differences among these approaches were within instrumental uncertainties but became significant for humid conditions, precluding our ability to use measurements to quantify aerosol direct radiative forcing at the top-of-the-atmosphere with sufficient accuracy at this time.

b. *Reno aerosol optics study*

Recognizing the importance of accurate measures of aerosol absorption for quantifying aerosol forcing mechanisms, the Reno Aerosol Optics Study (RAOS) was conducted in June 2002 to compare the performance of existing and new instrumentation for measuring aerosol light absorption at the Desert Research Institute in Reno, Nevada (Sheridan et al. 2005). Instruments of interest included cavity ring-down extinction instruments (Moosmüller et al. 2005; Sheridan et al. 2005; Strawa et al. 2003), a folded-path optical extinction cell (Virkkula et al. 2005), integrating nephelometers, photoacoustic spectrometers, and filter-based instruments (Arnott et al. 2005) including the PSAP, which is deployed at all ARM sites. Because of their wide use, the project focused on determining how well these filter-based measurements represent absorption and found that correction schemes were required to improve their accuracy. Otherwise, good agreement was found among the various methods. While the RAOS experiments were highly valuable in moving our understanding of absorption measurements forward, the conditions under which aerosol were sampled was limited, consisting of mixtures of kerosene soot and ammonium sulfate at low humidity (~15%–25%) only. More recent laboratory and field analyses have revealed that filter-based absorption measurement may be biased under conditions of high organics loadings (Arnott et al. 2003; Cappa et al. 2008; Lack et al. 2008) and the ARM and ASR Programs continue to support improved characterization of these measurements.

c. *2003 ARM aerosol IOP*

The laboratory intercomparisons of aerosol optical properties in RAOS were a preamble to the comprehensive Aerosol Intensive Observation Period of 2003 (AIOP 2003) field-based experiment undertaken in May at the SGP site. At this time is was clear that the largest contributor to uncertainty in forcing of climate change came from the incomplete knowledge of the relationships among aerosol composition, microphysical, and optical properties, leading to uncertainty in their clear-sky radiative effects and influence on the radiative properties of clouds. Comprehensive, redundant measurements of aerosol optical properties as well as broadband and spectral radiation were made from the surface and airborne platforms by in situ and active and passive remote sensing systems and were examined in relation to models to provide a better understanding of the source of these uncertainties.

Measurement verification and validation experiments included flying similar instrument systems in tandem for scattering, absorption, and extinction including nephelometers, PSAPs (Hallar et al. 2006), the Cadenza cavity ring-down extinction plus reciprocal nephelometer measurements (Strawa et al. 2006), and the first airborne photoacoustic measurements of absorption (Arnott et al. 2006). Airborne in situ derived optical properties were also compared to retrievals from ground-based radiance measurements from the Cimel sunphotometer and direct to diffuse ratios of irradiance calculated from these properties were compared to ground-based measurements from the MFRSR (Ricchiazzi et al. 2006). In general, all approaches agreed favorably for determining optical properties during the IOP. When larger differences were found, they were usually attributed to instrument response time, varying size ranges sampled due to differences in aircraft inlets, or conditions of low aerosol loading where sensitivity of some instruments was not adequate.

Other comparisons included those for the asymmetry parameter derived from a variety of methods (Andrews et al. 2006). In this study, high correlations were found between surface and airborne in situ values, but when these were compared to retrievals from both surface and airborne remote sensing measurements the correlations decreased. Pahlow et al. (2006) developed an approach for deriving aerosol hygroscopicity from lidar measurements and compared them to values derived from ground-based humidified nephelometry. While the lidar and nephelometer approaches showed reasonably good correlation, the lidar growth curves were much steeper with better sensitivity at high RH. This provided a possible synergy between these two measurement approaches to improve hygroscopicity estimates at and above the surface.

A large portion of the AIOP 2003 was devoted to better understanding of cloud condensation nuclei measurements. Gasparini et al. (2006) used a (tandem) differential mobility analyzer (DMA/TDMA) to measure aerosol size distribution and size-resolved hygroscopicity and modeled a multicomponent aerosol with these data as constraints. Composition dependent growth factors were determined and closure experiments were performed with measured optical properties and CCN. While reasonable agreement was found with optical properties, CCN were consistently slightly overpredicted. Rissman et al. (2006) also conducted an aerosol/CCN closure study, but in reverse to predict aerosol composition and mixing state. While these studies contributed to our understanding of aerosol properties that promote CCN, Ghan et al. (2006) developed a method for long-term, continuous estimation

of CCN at cloud base, essential for understanding aerosol–cloud interactions from surface in situ measurements. This technique is now an operational VAP available in the ARM Data Archive. New methods for quantifying aerosol–cloud interactions from the ARM suite of surface in situ and remote sensing measurements was presented in Feingold et al. (2006), and the topic is further addressed by Feingold and McComiskey (2016, chapter 22).

Two studies served as compendia of sorts of the AIOP 2003. Schmid et al. (2006) provided a complete comparison of aerosol extinction profiles from all available instrumentation at the SGP site. Airborne instrumentation included the NASA Ames Airborne Tracking 14-channel sunphotometer (AATS-14), the IAP nephelometer and PSAP, and the Cadenza cavity ring-down instrument; the Raman lidar and two micropulse lidars were deployed on the ground. Relative to the AATS-14, which was used as the benchmark measurement, airborne in situ measurements were biased low (11%–17%) and ground-based lidar measurements were biased high (13%–54%). The high 54% bias occurring for the Raman lidar identified a previously undetected slow loss in sensitivity of the instrument leading up to the AIOP which subsequently underwent a full refurbishment and upgrade accompanied by an improved processing algorithm.

Prior to the AIOP 2003, efforts to obtain closure for downwelling shortwave diffuse irradiance were met with difficulty (Mlawer et al. 2000) and indicated large uncertainties in aerosol optical properties, specifically single-scattering albedo. Michalsky et al. (2006) performed a shortwave radiative closure using six different radiative transfer models and the well-validated, redundant measurements of aerosol properties from the campaign. In this study, closure was achieved across all models and at a range of solar zenith angles to 1% for direct irradiances and to within 1.9% for diffuse irradiances, a large improvement over former efforts, attributed to more accurate inputs for aerosol optical properties and better irradiance measurements.

These results raised the question: with what accuracy must we be able to measure aerosol optical properties to calculate radiative fluxes or forcing to a desired accuracy? The ARM AWG at the time discussed this issue and developed an approach to provide guidelines for measurement accuracy at the three ARM fixed sites. The sensitivity of direct radiative forcing to inputs of aerosol optical properties (optical depth, single scattering albedo, asymmetry parameter, and Ångström exponents) and environmental variables (solar geometry and surface albedo) were calculated for diurnally averaged and instantaneous quantities (McComiskey et al. 2008). Given typical measurement uncertainties for these inputs, relative uncertainties in radiative forcing were 20%–80%, with higher values at high latitudes where fluxes are lowest and single-scattering albedo as the largest contributor. This work has served as a guide to where efforts should be placed to make the greatest reductions in uncertainty of aerosol radiative forcing.

d. The Aerosol Lidar Validation Experiment

Given the importance and capability of lidar measurements for representing continuous profiles of aerosol properties and the refurbishment and upgrade of the Raman lidar after the AIOP in 2003, a collaborative effort between NASA and ARM in September 2005 at the SGP served as a follow-on validation experiment. The Aerosol Lidar Validation Experiment (ALIVE) focused on validation of the Raman and micropulse lidars, again using the AATS-14 as a benchmark. Flights were made by the IAP Cessna equipped with the suite of in situ aerosol measurement to extend these validation efforts as well as the Research Scanning Polarimeter (Knobelspiesse et al. 2008; Waquet et al. 2009) flown with the AATS-14. During ALIVE, agreement between aerosol extinction profiles derived from AATS-14 and Raman lidar measurements improved to 6% (Schmid et al. 2009).

e. AMF Niamey

A year–long deployment of the AMF in Niamey, Niger, in 2006 provided an excellent opportunity to examine the radiative impacts of dust and biomass burning aerosol. The Sahelian region experiences monsoon conditions throughout the winter, but the dry periods promote large loadings (0.08–2.5 optical depths over the deployment) of both aerosol types (McFarlane et al. 2009). Aerosol optical properties were retrieved from passive remote sensing of the column (MFRSR; Kassianov et al. 2007) and profiles of extinction were determined by the micropulse lidar and MFRSR. Clear-sky, diurnally averaged surface aerosol radiative forcing calculated for the year was $21.1 \pm 14.3\,\mathrm{W\,m^{-2}}$ with surface closure revealing a mean difference between observed and calculated surface fluxes to be $5\,\mathrm{W\,m^{-2}}$. Thus, it was determined that aerosol optical properties retrieved by this method were reasonable and that a 10% variation in these properties would produce closure.

Turner (2008) used data from the AERI, an infrared interferometer measuring downwelling radiation from $530–3050\,\mathrm{cm^{-1}}$ at $1\,\mathrm{cm^{-1}}$ resolution, at the Niamey site to develop an original algorithm for retrieving airborne mineral dust composition. The algorithm is based on differential absorption bands for the different mineralogies. This information can be used to infer aerosol optical properties contributing to improvements in

radiative forcing calculations. Turner found that, during the AMF deployment, kaolinite and gypsum fit most of the data and that the varying amounts of gypsum correlated with air mass origin and trajectory. While this method cannot determine aerosol hematite concentrations due to the lack of an absorption band in the AERI wavelength region, the shortwave radiation signatures examined in McFarlane et al. (2009) suggest a hematite component. Bringing these approaches together will provide more comprehensive information on mineral aerosol and their radiative forcing.

f. Long-term characterizations of aerosol physical and optical properties

Many of the intensive studies described above were dedicated to improved understanding of various approaches to measuring aerosol properties. Observations have improved through better measurement protocols, calibrations, and retrieval methods, and by understanding how these different approaches may complement each other. All of these efforts have contributed to our ability to construct longer-term characterization from continuous measurements at the fixed sites.

1) Southern Great Plains

At the SGP, understanding of the aerosol climatology has come from the combined ground-based in situ, airborne, and remote sensing measurements at the site. A 4-yr (1996–2000) statistical analysis of the early surface AOS data (Sheridan et al. 2001) showed the aerosol at SGP to be a complex mix of aerosol types influenced on a range of scales from local-to-regional scale agricultural activities to synoptic flows. These data show an average annual peak in aerosol scattering in August with a secondary peak in February while absorption was greatest in the summer and fall. Together these patterns resulted in a decrease in single scattering albedo in the fall months. Delene and Ogren (2002) examined the impact of this variability in aerosol optical properties on surface radiative forcing and found less than 10% variability in forcing annually and no significant variation over the diurnal cycle. The profiles from the IAP airborne profile observations reflected these patterns for the column (Andrews et al. 2004, 2011) while showing that the single scattering albedo and Ångström exponent were fairly invariant with altitude. Over the long term, it was found that variability in aerosol properties in the column as measured by IAP was represented by the surface data, but that shorter-term (e.g., daily) variations might not be as well represented. To understand the conditions under which the surface data might be more representative of the column, Delle Monache et al. (2004) examined the IAP data in relation to boundary layer height and mixing and found that a well-mixed boundary layer did not improve correlations between the airborne observations and those at the surface, but that the two were relatively well correlated under a range of conditions.

A full representation of the column aerosol properties can only be made using remote sensing. Michalsky et al. (2010) developed a 12-yr climatology (1992–2008) of aerosol optical depth and Ångström exponent from the MFRSR at SGP (Fig. 21-4) that reflected the summertime peak in scattering from the AOS data. The optical depth time series (top, Fig. 21-4), consisting of over 4000 daily average values, indicated a high degree of variability in the magnitude of the summertime peak whereas the winter minimum was relatively constant and showed a general lack of diurnal variability. The time series of Ångström exponent also showed a robust annual cycle with higher values (smaller particles) in the summer and minima (larger particles) in both December and April, the latter likely due to long-term transport of Asian dust. An outstanding feature in the Ångström exponent is the low values representative of the Pinatubo eruption at the beginning of the time series, and recovery to more typical continental aerosol values within a few years.

2) North Slope of Alaska

The phenomenon of Arctic haze, where aerosol concentration from anthropogenic sources at lower latitudes becomes highly concentrated over the Arctic in the winter and early spring, has been observed and documented for decades. However, quantifying the radiative impacts of these aerosols has been a challenge due to a lack of knowledge of their composition, optical properties, and interaction with radiation in the unique environmental conditions of the Arctic, characterized by high surface albedo and low solar angles. Nowhere have the long-term filter-based aerosol chemistry measurements been as important as at the NSA site in Barrow. Measurements have been made alongside optical properties at the surface since 1998 and have yielded a wealth of information on Arctic haze. Quinn et al. (2002) presented seasonal cycles of aerosol components, suggesting their sources, and their relationship to light scattering and absorption, and indicating the radiative effects of these different aerosol types. Both anthropogenic and natural (fine mode sea salt) aerosol were found to peak in winter and spring, all from long-term transport. In the summer, coarse mode sea salt and marine biogenic aerosols were found in greater concentration. Sea salt contributed most to light scattering in the winter and non–sea salt sulfate in the spring, while both were important over the summer. An analysis of trends at Barrow from 1976 to 2008 (Quinn et al. 2009) revealed decreases in anthropogenic aerosol of ~60% in the Arctic, although it was determined that

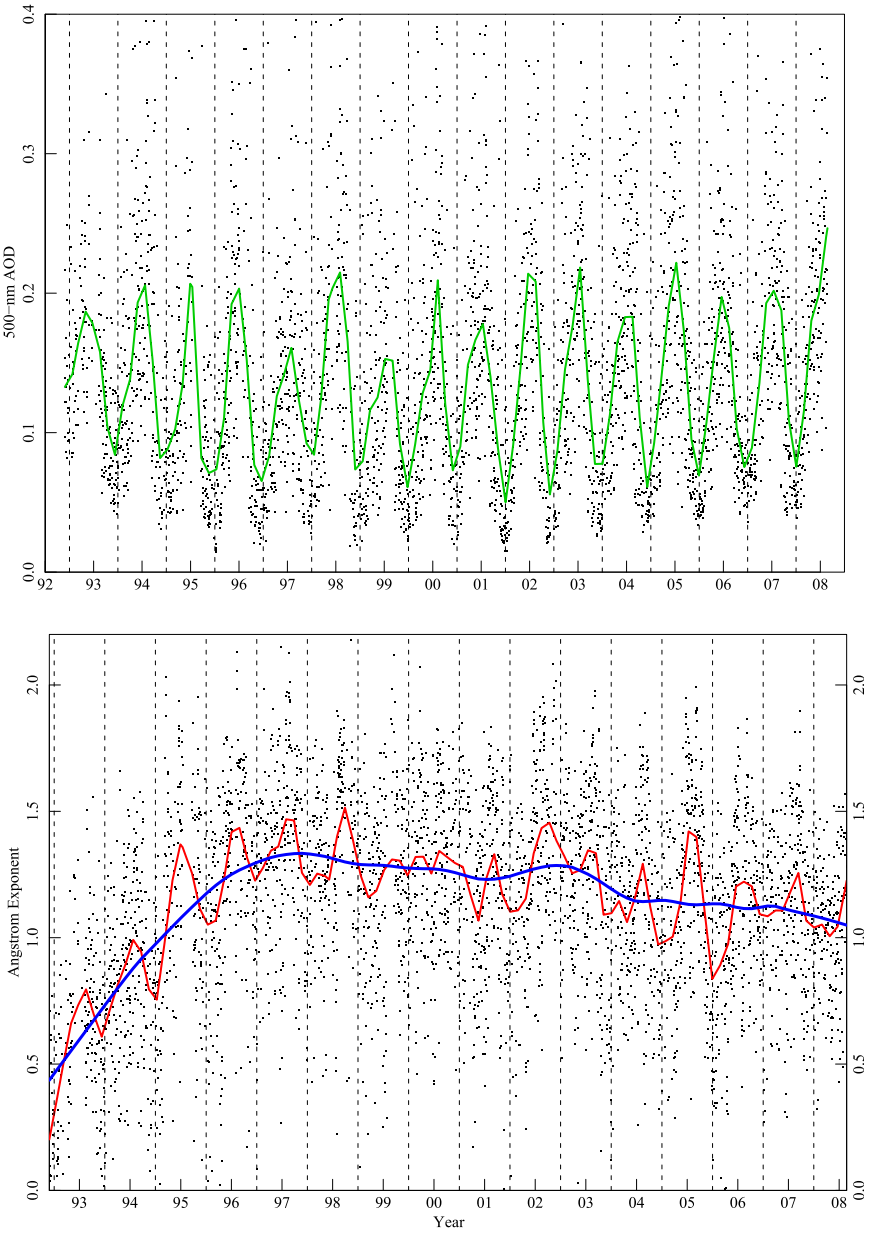

FIG. 21-4. (top) Aerosol optical depth (500 nm) and (bottom) Ångström exponent climatology (1992–2008) from the MFRSR at SGP. Black points represent daily averages and the green and red lines the locally weighted smoothed estimate. From Michalsky et al. (2010, their Figs. 8a and 10, respectively).

the source regions themselves remained similar. At the same time, marine biogenic aerosol has increased in the summertime, which has been found in other studies (O'Dwyer et al. 2000) to be correlated with loss of sea ice extent and increase in sea surface temperature.

4. Looking forward

In the past five years, the ARM Program has achieved a tremendous expansion of its aerosol observational capabilities with the recognition of the role and uncertainty of aerosol in the climate system. The integration of measurements of aerosol chemical, physical, and optical properties for improved model representation and parameterization development has been a focus. Further, ABE is seeing renewed development with the promise of direct retrievals of aerosol optical properties from active remote sensing (e.g., Müller et al. 2014). New foci in the program include understanding aerosol mixing state and

its impact on radiative fluxes (Cappa et al. 2012), new particle formation related to CCN concentrations, secondary organic aerosol (SOA) formation, and improved understanding of measurements of aerosol absorption from various platforms. Obtaining comprehensive geographical coverage of aerosol measurements with the AMFs, with an eye to capturing seasonal cycles by deploying for at least a year, is critical to understanding aerosol processes and remains a priority within the ARM Program.

The seminal article by Charlson et al. (1992) that motivated much of the structure of the aerosol program within the ARM Program enjoys more than 2500 citations 20 years later. The science questions that the ARM AWG outlined that year are relevant today, but the details have changed and the knowledge required to understand processes at the scales represented in models has exploded. Capabilities for detailed, in situ observations at increasingly finer scales and for specific aspects of the aerosol system are continuously being added by the program, for example measurement of black carbon by the Single-Particle Soot Photometer (SP2) in the United States and India (Sedlacek et al. 2012) and new particle formation using the newly acquired nano-SMPS and SO_2 analyzer at SGP. ASR now has an Aerosol Life Cycle Working Group to address some aspect of the problems outlined here and a Cloud–Aerosol–Precipitation Interactions Working Group to address others. Contributions of the ARM Program in the latter category are addressed by Feingold and McComiskey (2016, chapter 22).

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