### The Atmospheric Radiation Measurement Program May 2003 Intensive Operations Period Examining Aerosol Properties and Radiative Influences: Preface to Special Section

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#### Abstract

Atmospheric aerosols influence climate by scattering and absorbing radiation in clear air (direct effects) and by serving as cloud condensation nuclei, modifying the microphysical properties of clouds, influencing radiation and precipitation development (indirect effects). Much of present uncertainty in forcing of climate change is due to uncertainty in the relations between aerosol microphysical and optical properties and their radiative influences (direct effects) and between microphysical properties and their ability to serve as cloud condensation nuclei at given supersaturations (indirect effects). This paper introduces a special section that reports on a field campaign conducted at the Department of Energy Atmospheric Radiation Measurement site in North Central Oklahoma in May, 2003, examining these relations using in situ airborne measurements and surface-, airborne-, and space-based remote sensing.

#### **1.0 Background and Motivation**

Two key requirements for testing understanding of the influence of radiative processes on climate are: 1) relating observations of radiative fluxes and radiances to the atmospheric composition and, 2) using these relations to develop and test parameterizations to accurately predict the atmospheric radiative properties. These are the primary objectives of the Atmospheric Radiation Measurement (ARM) program supported by the Department of Energy [*Ackerman and Stokes*, 2003]. Among the key uncertainties influencing atmospheric radiation processes in the atmosphere are the influences of atmospheric aerosols. Consequently, ARM has pursued measurement and modeling activities that examine aerosol impacts on atmospheric radiative transfer, both in cloud-free skies (direct effects) and through modification of the microphysical and radiative properties of clouds (indirect effects).

This special issue presents papers reporting results from an intensive field campaign examining the properties and radiative influences of aerosols, the May 2003 Aerosol Intensive Operations Period (AIOP) conducted between May 5-31, 2003 over the ARM Southern Great Plains (SGP) Climate Research Facility (CRF) site (36.606 N, 97.50 W, 315 m). The scientific hypotheses that were investigated during this IOP were posed as "closure experiments" in which an observable quantity is measured in two or more different ways, or is measured as well as calculated (modeled) using other measured quantities. Closure is achieved if the several measures agree within their mutual uncertainties.

The specific closure experiments carried out in this IOP are described below, followed by a brief summary of the measurements acquired during the IOP. Table 1 provides a list of acronyms and symbols.

# 1.1 To what extent can closure between measurements and models of diffuse radiation be achieved, especially under conditions of low aerosol optical thickness?

In prior work using measurements acquired at the SGP CRF, *Halthore and Schwartz* [2000] reported that modeled diffuse downwelling irradiance exceeded measurements by an amount that could not be accounted for by uncertainties in measurements or aerosol-scattering properties that

are input into the radiative transfer models or by errors in multiple-scattering schemes. *Mlawer* et al. [2000] achieved closure between ground-based measurements of direct and diffuse solar irradiance, as measured by the Rotating Shadowband Spectroradiometer (RSS) [Harrison et al., 1999] at the SGP site. That study used well-validated aerosol optical thickness (AOT) [Schmid et al., 1999] and water vapor measurements [Revercomb et al., 2003] as input. However, in order to minimize the residuals between measurements and model, Mlawer et al. [2000] had to assume aerosol single scattering albedos  $\omega_0$  which were "much lower than usually assumed in the aerosol community for this location, and [which] present an intriguing puzzle for this community to consider". For three cases in September/October 1997 Mlawer et al. [2000] found  $\omega_0 = 0.89, 0.90, \text{ and } 0.67$  (assumed spectrally-invariant). These values may be compared with measurements reported by Sheridan et al. [2001] based on a 4-year record (1996-2000) of ground-based aerosol measurements at the SGP site, for which the median value of  $\omega_0$  was 0.95 ( $\lambda$ =550 nm, ambient RH). More specifically, although values of  $\omega_0$  as low as 0.87 occurred on occasion in September/October 1997, such a value is much greater than the value 0.67 needed to achieve radiative closure in a single case by Mlawer et al. [2000]. This discrepancy has raised concerns regarding the accuracy of the  $\omega_0$  measurements at the surface SGP site, and how well  $\omega_0$  derived from the surface measurements represents the effective column value.

Because of this uncertainty in the values of aerosol absorption coefficient and single scattering albedo  $\omega_0$  and the resulting difficulty in reconciling measurements and models of diffuse irradiance, a focus of the IOP was the determination aerosol absorption coefficient using multiple in situ and remote-sensing methods. These techniques included conventional filterbased measurements via the Particle Soot Absorption Photometer (PSAP) [*Horvath*, 1993; *Bond et al.*, 1999; Virkkula et al., 2005; Strawa et al., 2005], the recently implemented photoacoustic method [*Moosmüller et al.*, 1998; *Arnott et al.*, 1999; Arnott et al. 2005], and a new method to measure the aerosol absorption coefficient as the difference between aerosol extinction and scattering coefficients measured using Continuous Wave Cavity Ring-Down (CW-CRD) technology [*Strawa et al.*, 2003; *Strawa et al.*, 2005]. As a prelude to this Aerosol IOP, the Reno Aerosol Optics Study (RAOS) [*Sheridan et al.*, 2005] characterized, under controlled conditions, these and other in situ instruments used to measure aerosol light extinction, absorption, and scattering coefficients. After the successful intercomparisons performed during the RAOS, the Aerosol IOP represented the first successful demonstration of an airborne photoacoustic sensor to measure aerosol absorption coefficient and only the second time that an airborne CW-CRD was deployed to measure aerosol optical properties.

In this special section, *Arnott et al.* [2005] and *Strawa et al.* [2005] describe the in situ measurements of aerosol absorption and extinction coefficients, and *Andrews et al.* [2005] describe how values for the Mie-equivalent aerosol asymmetry parameter (g) were derived using both in situ and remote sensing measurements. *Michalsky et al.* [2005] describe comparisons of measured and modeled direct and diffuse irradiance and *Ricchiazzi et al.* [2005] describe the aerosol parameters derived from observations of sky radiance.

## **1.2** What is the agreement among profiles of aerosol scattering and extinction coefficients determined from the ARM SGP Raman lidar, in situ, and remote-sensing measurements?

Extinction closure studies can be viewed as addressing the extent to which in situ measurements of aerosol properties can account for the attenuation of direct normal solar irradiance by an aerosol layer or column. The closure experiment is thus the agreement between aerosol extinction optical thickness  $\tau_{ep}$  at the surface and the vertical integral of the extinction coefficient  $\int \sigma_{ep}(z) dz$ . Aerosol optical thickness is derived at the ARM SGP CRF from routine measurements by several instruments at discrete wavelengths (Cimel Sun photometer, Multifilter Rotating Shadowband Radiometer (MFRSR), Normal Incidence Multifilter Radiometer (NIMFR), CRF Raman lidar (CARL)), and as a continuous function of wavelength using the Rotating Shadowband Radiometer (RSS). Measurements of AOT by the Cimel and the MFRSR have been shown to agree closely, typically to 0.02 [Halthore et al., 1997; Schmid et al., 1999]. Although comparisons of aerosol optical thickness between the Raman lidar and Sun photometer have shown small (<5%) systematic biases, these same comparisons have shown rms differences of typically 20-30% [Turner et al., 2001]. The reasons for the 30% rms differences between the instruments are not known. Possible contributing factors are variations in aerosol extinction/backscatter ratio used for lidar retrievals below 800 meters, and uncertainty in the lidar overlap function correction.

To help address the representativeness of surface aerosol measurements, since March, 2000, ARM has been measuring in situ aerosol profiles (IAP) of aerosol scattering and absorption coefficients by performing systematic flights (typically twice weekly) with a light aircraft (Cessna C-172N) over the SGP site utilizing an aerosol instrument package similar to the one at the SGP ground site. Factors that impact the ability to use IAP measurements to derive aerosol extinction and optical thickness are: the aircraft package measures the aerosol at a relative humidity of 40% rather than at ambient RH, the inlet allows particles to pass only if their aerodynamic diameter is less than1 µm, and the ceiling of the aircraft is limited (~3.5 km). Even after applying (altitude-independent) corrections for all these limitations (using information from ground-based nephelometers and Raman lidar), Andrews et al. [2004] showed that the aircraft measurements do not account for all of the aerosol extinction measured at the surface; specifically, the IAP-derived aerosol optical thicknesses were consistently less (0.05 or  $\sim 30\%$ ) than the aerosol optical thicknesses (AOT) measured on the ground by Sun photometers so that extinction closure was not achieved. A similar discrepancy was found when comparing the IAP extinction with extinction from the ground-based Raman lidar at the SGP site for altitudes between 300 and 3500 m above ground level (i.e. IAP extinction 30% lower than Raman, Ferrare et al., [2002, 2003]. These differences may be due to uncertainties in the humidification factor (correcting the extinction coefficient as measured at low RH to the value appropriate for ambient RH), correction for extinction by supermicrometer particles, and the aerosol Ångström exponent used to scale the lidar measurements from 355 nm to 550 nm.

During the aerosol IOP additional airborne measurements were used to better quantify the errors associated with the IAP measurements and to identify potential reasons for these differences. The NASA Ames Airborne Tracking 14-channel Sunphotometer, AATS-14 [Schmid et al, 2000] was used to measure profiles of aerosol optical thickness  $\tau_{ep}(z)$  and aerosol extinction coefficient (determined as  $d\tau_{ep}(z)/dz$ ) as a function of wavelength at ambient conditions. Papers in this special section compare these profiles to aerosol extinction profiles determined by Raman lidar, airborne in situ, and MicroPulse Lidar (MPL) measurements [Schmid et al., 2005; Ferrare et al., 2005], and report aerosol extinction coefficients as measured by the new Continuous Wave Cavity Ring-Down (CW-CRD) airborne in situ instrument [Strawa et al., 2005] and as derived as the sum of nephelometer measurements of humidified aerosol

scattering and PSAP measurements of aerosol absorption [*Schmid et al.*, 2005; *Ferrare et al.*, 2005; *Strawa et al.*, 2005]. *Hallar et al.* [2005] compare aerosol scattering measured simultaneously by identical commercial nephelometers deployed on two separate aircraft flying in formation, and examine the impacts of coarse mode aerosols on measurements of aerosol scattering.

1.3 To what extent does CCN number concentration (at several supersaturations in the range ~0.1 - 1%) agree with calculations based on aerosol size distribution, at the surface and at cloud base? How well are the cloud nucleating properties of particles just below cloud base represented using surface measurements of cloud nucleating properties of particles along with profiles of relative humidity and aerosol extinction?

The effects of aerosols on cloud properties must be quantified in order to accurately describe the effects of clouds on atmospheric radiative fluxes and radiances. These effects include both the increase in cloud reflectivity due to more and smaller cloud droplets forming on the aerosol, as well as the possible increase in the lifetime of clouds due to reduced precipitation in clouds with more and smaller droplets. Few prior studies have acquired airborne measurements of cloud droplet size distribution and cloud liquid water content together with the CCN spectrum at cloud base. As most of the prior data sets were obtained in maritime areas with relatively low aerosol loadings, the ARM Aerosol IOP examined the aerosol indirect effect over a continental area (i.e. Oklahoma).

One study conducted during the Aerosol IOP examined the feasibility of retrieving the vertical profile of CCN concentration using surface measurements, under conditions of uniform aerosol composition and shape of the aerosol size distribution below cloud base [*Ghan et al.,* 2005]. A combination of aircraft, surface in situ, and surface remote-sensing measurements were used to evaluate the retrieval schemes. Airborne measurements were used to examine CCN concentrations calculated using assumed and inferred aerosol composition and mixing state, and measurements of the aerosol size distribution [*Rissman et al.,* 2005].

Additional investigations were conducted using surface CCN measurements to investigate the diurnal and day-to-day variability of CCN concentrations, the representativeness of the SGP site of continental conditions [*Gasparini et al.*, 2005a], as well as the extent to which the aerosol could be modeled as a population of multi-component particles, each consisting of organic carbon, elemental carbon, mineral dust, ammonium sulfate, and water, or some subset of these components [*Gasparini et al.*, 2005b].

# 1.4 To what extent are remotely sensed parameters adequate for quantifying the aerosol indirect effect?

The Aerosol IOP also examined the ability of ground-based remote sensors to quantify the first aerosol indirect effect at SGP. The premise is that cloud response to relative changes in aerosol extinction, under conditions of equivalent water path, can be quantified using surface-based measurements that simultaneously address aerosol and cloud parameters in a column of air above the site [*Feingold et al.*, 2003]. Cloud response is represented by changes in cloud-drop effective radius and the amount of aerosol loading is represented by aerosol extinction at a prescribed distance beneath cloud base. This approach avoids assumptions that (a) the surface aerosol is representative of the aerosol affecting the cloud [*Ramanathan* et al., 2001; *Menon et al.*, 2002], or that (b) the column integrated extinction coefficient (i.e., optical thickness) in cloud-free areas is representative of the aerosol affecting the cloud [e.g., *Kaufman and Nakajima*, 1993; *Han et al.*, 1998; *Bréon et al.*, 2002]. The approach does, however, raise the question of representativeness of extinction coefficient as a proxy for CCN [*Feingold* 2003].

A key measure of aerosol influences on cloud microphysical properties, the cloud-drop effective radius  $r_{eff}$ , was determined by several remote-sensing techniques including the radar/microwave radiometer combination, MultiFilter Rotating Shadowband Radiometer (MFRSR)/microwave radiometer combination, both of which used surface-based measurements, airborne measurements from the Solar Spectral Flux Radiometer (SSFR), and space-based measurements from the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument. Using measurements acquired during the Aerosol IOP, *Feingold et al.* [2005] developed a methodology for representing the best-estimate of  $r_{eff}$  based on these various retrievals, each with their distinct sampling volumes and vertical weighting. An Aerosol IOP investigation also examined the consistency between retrievals of the aerosol hygroscopic growth factor

determined using ground based lidar measurements and derived from airborne in situ measurements of aerosol scattering as function of RH [*Pahlow et al.*, 2005]. *Gasparini et al.* [2005b] used surface DMA/TDMA measurements of aerosol hygroscopic growth factors, in conjunction with backtrajectory clustering, to infer aerosol composition and to gain insight into the processes responsible for evolution.

#### 2.0 Measurement Summary

During the mission, an extensive suite of instruments were deployed on board the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft [*Bluth et al.*, 1996; *Bane et al.*, 2004]. There were a total of 16 science flights, for a total of 60.6 flight hours, conducted by the CIRPAS Twin Otter aircraft on 15 days during this period. Most of the flights were conducted under clear or partly cloudy skies to assess aerosol impacts on solar radiation. Additional flights were used to target mostly cloudy skies to assess aerosol/cloud interactions, test our understanding and model representation of aerosol activation, and to test how well surface remote sensing of the indirect effect works.

A wide range of aerosol and water vapor conditions were observed over the ARM SGP site during the IOP. Figure 1 shows the water vapor mixing ratio and the aerosol extinction coefficient derived from the Raman lidar measurements during this period as well as AOT (340 nm) and aerosol Ångström exponent (340-870 nm). AOT was derived using measurements from a Cimel Sun photometer deployed as part of the AERONET project [*Holben et al.*, 1998] at the SGP site. Ångström exponents were derived using Cimel AOT measurements at 340, 380, 440, 500, 670, and 870 nm. Note the large variations in the water vapor and aerosol extinction and the correlation of water vapor with aerosol extinction. Such correlation might be attributed to RH-dependent increase of AOT and/or to the two quantities being similarly influenced by atmospheric transport and precipitation removal. High AOT was observed during both early (May 8-9) and late (May 28-29) in the IOP. Low values of the Ångström exponent observed during early May suggests that aerosol extinction was due to large, coarse mode aerosols (e.g. dust), whereas higher values of the Angstrom exponent measured during the latter part of May suggest that these were smaller, accumulation-mode particles.

Several times during the IOP, elevated aerosol layers were observed over the ARM SGP site. These layers, which were present 2-5 km above the surface, are often the result of the transport of smoke, dust, or pollution from distant sources. Observations of these layers during the IOP indicate that such layers may be more common than originally thought, and can have a substantial impact on the atmospheric radiation budget. As an example, satellite imagery and back trajectory analyses indicate that the elevated aerosol layers observed by the ground based lidars and the airborne remote-sensing and in situ instruments between May 25-27 were smoke layers produced by Siberian forest fires [*Damoah et al.*, 2004; *Jaffe et al.*, 2004].

The field deployment phase of the Aerosol IOP was successful in several ways. The instruments deployed on the aircraft and on the surface generally worked very well and acquired the data required to address the IOP objectives. These instruments included both well established and newly developed airborne instruments to measure aerosol optical properties (scattering, absorption, and extinction coefficients), aerosol size distribution, and cloud condensation nucleus (CCN) concentrations, as well as surface based instruments to measure aerosol composition, aerosol optical properties, and cloud condensation nucleus concentrations and spectra. Additional airborne and surface instruments acquired the desired measurements of solar direct and diffuse irradiance.

This special issue reports on some of the research conducted during this Aerosol IOP. These several papers present in detail the ground-based and airborne instrumentation, the measurements, as well as the associated analyses.

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### **Figure Caption**

Figure 1. (a) water vapor mixing ratio derived from Raman lidar measurements during the May 2003 Aerosol IOP. (b) same as (a) except for aerosol extinction cofficient (355 nm). (c) Aerosol optical thickness (AOT) derived from ground based Cimel Sun photometer measurements at the SGP site. (d) Aerosol Ångström exponent values (340-870 nm) derived from the Cimel AOT measurements. The white vertical bands in the lidar images represent periods when the Raman lidar did not operate because of malfunctions of the air conditioner system in the lidar enclosure (May 15, 16, 23) and laser cooling system (May 27).

### Table 1. Frequently used acronyms and symbols

AATS-14	Ames Airborne Tracking 14-channel Sun photometer
AERONET	AErosol RObotic NETwork
ARM	Atmospheric Radiation Measurement
AIOP	Aerosol Intensive Operations Period
AOT	Aerosol Optical Thickness
CARL	CRF Raman Lidar
CCN	Cloud Condensation Nuclei
CIRPAS	Center for Interdisciplinary Remotely-Piloted Aircraft Studies
CRD	Cavity Ring-Down
CRF	Climate Research Facility
IAP	In situ Aerosol Profiles
IOP	Intensive Operations Period
MFRSR	Multifilter Rotating Shadowband Radiometer
MODIS	Moderate Resolution Imaging Spectroradiometer
MPL	Micro Pulse Lidar
NIMFR	Normal Incidence Multi-Filter Radiometer
PSAP	Particle/Soot Absorption Photometer
RAOS	Reno Aerosol Optics Study
RH	Relative Humidity
RSS	Rotating Shadowband Spectroradiometer
SGP	Southern Great Plains
SSFR	Solar Spectral Flux Radiometer
g	Aerosol asymmetry parameter
r <sub>eff</sub>	Cloud-drop effective radius
$ au_{ m ep}$	Aerosol extinction optical thickness
$\omega_0$	Aerosol single scatterng albedo



Figure 1. (a) water vapor mixing ratio derived from Raman lidar measurements during the May 2003 Aerosol IOP. (b) same as (a) except for aerosol extinction cofficient (355 nm). (c) Aerosol optical thickness (AOT) derived from ground based Cimel Sun photometer measurements at the SGP site. (d) Aerosol Ångström exponent values (340-870 nm) derived from the Cimel AOT measurements. The white vertical bands in the lidar images represent periods when the Raman lidar did not operate due to malfunctions of the air conditioner system in the lidar enclosure (May 15, 16, 23) and laser cooling system (May 27).