

# Raman lidar profiling of aerosols over the central U.S.; diurnal variability and comparisons with the GOCART model

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## ABSTRACT

We use profiles of aerosol extinction, water vapor mixing ratio, and relative humidity measured by the ARM SGP Raman lidar in northern Oklahoma to show how the vertical distributions of aerosol extinction and water vapor vary throughout the diurnal cycle. While significant (20-30%) variations in aerosol extinction occurred near the surface as well as aloft, smaller (~10%) variations were observed in the diurnal variability of aerosol optical thickness (AOT). The diurnal variations in aerosol extinction profiles are well correlated with corresponding variations in the average relative humidity profiles. The water vapor mixing ratio profiles and integrated water vapor amounts generally show less diurnal variability. The Raman lidar profiles are also used to evaluate the aerosol optical thickness and aerosol extinction profiles simulated by the GOCART global aerosol model. Initial comparisons show that the AOT simulated by GOCART was in closer agreement with the AOT derived from the Raman lidar and Sun photometer measurements during November 2000 than during September 2000. For both months, the vertical variability in average aerosol extinction profiles simulated by GOCART is less than the variability in the corresponding Raman lidar profiles.

## 1. INTRODUCTION

Global models have been increasingly used to assess climate change scenarios. Since some of the largest uncertainties in model simulations of climate change are associated with aerosols, evaluating how these models portray aerosol characteristics is vital for determining uncertainties in simulations of aerosol radiative forcing and climate change. Assessments of aerosol models have to date focused primarily on comparing estimates of column integrated aerosol optical thickness (AOT) with satellite retrievals and/or ground-based measurements of AOT. However, AOT alone does not provide enough information to resolve several specific model deficiencies. One problem common to all models becomes particularly apparent when comparing the vertical distributions of aerosols. A model intercomparison performed as part of the Third IPCC Assessment of aerosol effects found that the vertical distribution of aerosol concentrations differs by a factor of two or more from one model to the next, especially for components other than sulfate<sup>1</sup>. The lack of a climatological database to characterize the vertical distributions of aerosols has hampered efforts to evaluate and consequently improve such models.

Lidar measurements can provide one means of characterizing the vertical distribution of aerosols. Through its design as a turnkey, automated system for unattended, around-the-clock profiling of water vapor and aerosols, the U.S. Department of Energy Atmospheric Radiation Measurement (ARM) Cloud and Radiation Testbed (CART) Raman lidar (CARL) has begun to provide a climatological database of aerosol and water vapor profiles<sup>2</sup>. In this paper we show how we have used these profiles to characterize the diurnal behavior of the vertical distributions of aerosol and water vapor over the ARM Southern Great Plains (SGP) site. We also discuss how these lidar profiles have been used to evaluate the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) global aerosol model simulations of AOT and aerosol extinction profiles.

## 2. CART RAMAN LIDAR

CARL uses a tripled Nd:YAG laser, operating at 30 Hz with 350-400 millijoule pulses to transmit light at 355 nm. A 61-cm diameter telescope collects the light backscattered by molecules and aerosols at the laser wavelength and the Raman

scattered light from water vapor (408 nm) and nitrogen (387 nm) molecules. A beam expander reduces the laser beam divergence to 0.1 mrad, thereby permitting the use of a narrow (0.3 mrad) as well as a wide (2 mrad) field of view. The narrow field of view, coupled with the use of narrowband (~0.4 nm bandpass) filters, reduces the background skylight and, therefore, increases the maximum range of the aerosol and water vapor profiles measured during daytime operations.

A series of automated algorithms are used to derive water vapor and aerosol profiles<sup>3</sup>. Water vapor mixing ratio profiles are computed using the ratio of the Raman water vapor signal to the Raman nitrogen signal. Relative humidity profiles are computed using these water vapor mixing ratio profiles and the temperature profiles from a physical retrieval algorithm that uses data from a collocated Atmospheric Emitted Radiance Interferometer (AERI). The water vapor mixing ratio profiles are integrated with altitude to derive precipitable water vapor (PWV). Profiles of aerosol scattering ratio, which is the ratio of aerosol+molecular scattering to molecular scattering, are derived using the Raman nitrogen signal and the signal detected at the laser wavelength. Aerosol volume backscattering cross section profiles are then computed using the aerosol scattering ratio and molecular scattering cross section profiles derived from atmospheric density data. These density profiles are computed using coincident pressure and temperature profiles. Aerosol extinction profiles are computed from the derivative of the logarithm of the Raman nitrogen signal with respect to range. AOT is derived by integration of the aerosol extinction profile with altitude.

### 3. MEASUREMENTS OF DIURNAL VARIABILITY

CARL aerosol and water vapor profiles acquired over 946 days between March 1, 1998 and December 31, 2001 were used to characterize diurnal variability. During this period, CARL operated an average of about 55% of the time. Figure 1 shows aerosol extinction, water vapor mixing ratio, and relative humidity profiles averaged over each hour of the day for both the winter (December-February) and summer (June-August) seasons. The average over the summer included CARL measurements from 205 days during these years, and the winter average included CARL measurements over 180 days. Cloudy samples were excluded from these averages. Times of average sunrise and sunset are also shown.

The highest aerosol extinction was generally observed close to the surface during the nighttime just prior to sunrise. The high values of aerosol extinction are most likely associated with increased scattering by hygroscopic aerosols, since the corresponding average relative humidity values were above 70%. After sunrise, relative humidity and aerosol extinction below 500 m decreased with

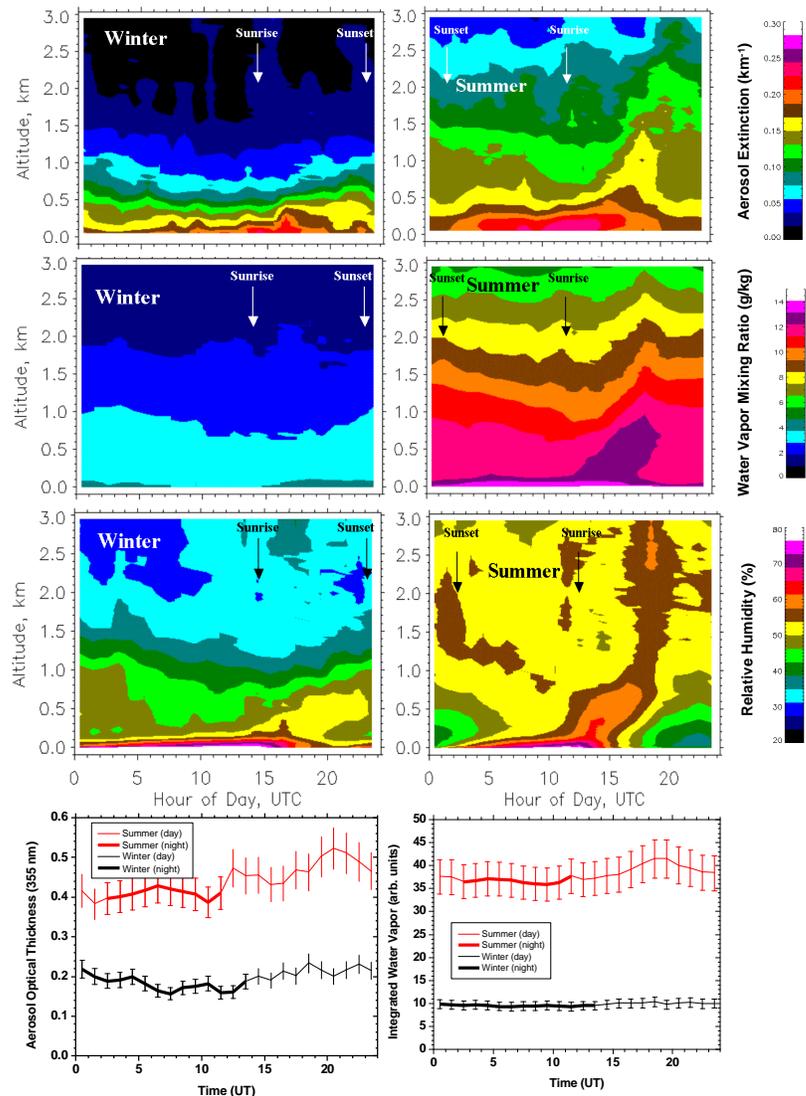


Figure 1. (top) Images showing average diurnal variation of aerosol extinction (top), water vapor mixing ratio (middle), and relative humidity (bottom) profiles measured by CARL for winter (left) and summer (right). (bottom) average AOT (left) and integrated water vapor (right) for summer and winter.

the growth in the daytime convective boundary layer. The largest aerosol extinction for altitudes above 1 km occurred during the early afternoon most likely as a result of the increase in relative humidity. The water vapor mixing ratio profiles generally showed smaller variations with altitude between day and night. The aerosol extinction profiles show that relatively large (10-25%) changes that occur in the average aerosol extinction profiles have a smaller impact on the AOT. Figure 1 also shows the diurnal variability of both AOT and integrated water vapor for winter and summer. The standard deviation of the AOT was about 10% of the daily average AOT during both summer and winter. In contrast, the water vapor profiles showed about half this variability for both the summer and winter cases.

#### 4. COMPARISONS WITH THE GOCART MODEL

The CARL measurements of aerosol extinction and AOT acquired during 2000 were used to evaluate the performance of the GOCART model for this same period. The GOCART model is potentially a suitable tool for linking satellite, surface, and airborne aerosol observations. The model incorporates major tropospheric aerosol types, including sulfate, dust, organic carbon (OC), black carbon (BC), and sea-salt aerosols, and provides global distributions of aerosol concentrations, vertical profiles, and optical thickness of individual components as well as total aerosols. This model is driven by assimilated meteorological fields, which are generated by the Goddard Earth Observing System Data Assimilation System (GEOS DAS). Comparisons of AOT simulated by the model with AOT derived from the AVHRR and TOMS satellite sensors, as well as with ground-based Sun photometers from the Aerosol Robotic Network (AERONET) show that this model reproduces most of the prominent features in the satellite data, including the seasonal shift of the Saharan dust plume from Africa. These comparisons also showed that GOCART reproduces the seasonal variations at most sites, especially those sites where biomass burning and dust dominate, although the magnitudes do not always match the observations<sup>4</sup>.

Figure 2 shows a comparison of monthly average AOT simulated by the model and measured by CARL and a co-located Sun photometer for September and November, 2000. CARL uptimes during these two months were 95% and 85%, respectively. The wavelengths for the AOT and aerosol extinction profiles shown in Figure 2 are 355 nm (CARL), 350 nm (GOCART), and 340 nm (Sun photometer). Note that the GOCART values represent averages over a 2-degree latitude by 2.5-degree longitude box. The comparisons for September 2000 show that the GOCART average AOT was about half that measured by the Raman lidar and Sun photometer. The average GOCART aerosol extinction profile was also significantly less than the corresponding average Raman lidar profile. The excellent agreement between measured and modeled relative humidity profiles indicates that the disagreement between measured and model aerosol extinction and AOT is probably not associated with errors in the simulated relative humidity fields and the resulting humidification of hygroscopic aerosols. These differences may be due to: an underestimate of sulfates caused by an overestimate of the wet removal rate of sulfate and sulfur dioxide<sup>5</sup>, an underestimate of the sulfur dioxide oxidation rate, and/or an underestimate of the amount of dust over the SGP site during the summer. Examination of aerosol Angstrom exponents derived from Sun photometer AOT measurements and dust amounts from model results for previous years suggest that the model may have underestimated the amount of dust amount during the summer of 2000.

The agreement between the modeled and measured AOT for

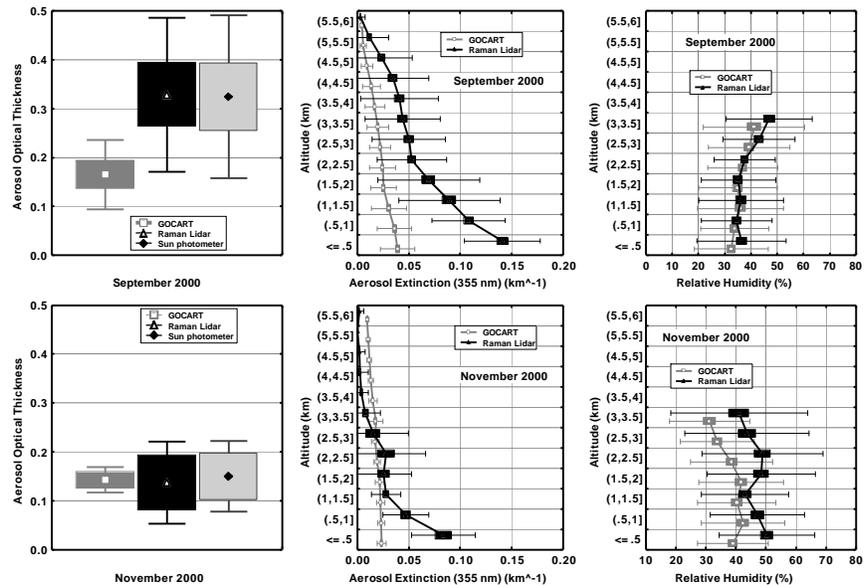


Figure 2. Comparisons of average measured and modeled AOT (left), aerosol extinction profiles (middle), and relative humidity profiles (right). Boxes represent +/- 2 st. error of the average and error bars represent standard deviations of the measurements.

November 2000 was much better. However, note that while the AOT comparison shows good agreement this month, the GOCART and CARL aerosol extinction profiles show significant disagreement throughout the lower troposphere. This case illustrates that good agreement between measured and modeled AOT does not necessarily mean that the model correctly represents the vertical distribution of aerosols. Note that in the case of absorbing aerosols, differences in the vertical distributions of aerosols can have large impacts on calculations of radiative heating rates and radiative forcing. In both September and November 2000 cases, the average GOCART aerosol extinction profiles show much less vertical variability than the corresponding CARL profiles, and considerably smaller values near the surface than the lidar profiles.

## 5. CONCLUSION

Raman lidar profiles of aerosol extinction acquired over nearly four years are used to study the diurnal variability of aerosols and water vapor over the ARM SGP site in northern Oklahoma. These profiles show that significant (20-30%) variability in aerosol extinction occurs between day and night. The largest values of average aerosol extinction occurred generally below about 300 m during the early morning hours prior to sunrise. Aerosol extinction also increased above 1 km during the early afternoon, particularly during the summer. The lidar profiles show that the increase in aerosol extinction is well correlated with similar variations in relative humidity. Water vapor profiles and integrated water vapor amounts generally show smaller variability than aerosol extinction and AOT.

Aerosol optical thickness and aerosol extinction profiles simulated by the GOCART global aerosol model are evaluated using the Raman lidar measurements. GOCART simulations show less AOT during September 2000 than do Raman lidar and Sun photometer measurements. The reasons for this are not known at this time but may be due to the model underestimating sulfates and/or dust during the summer months over the ARM SGP site. AOT comparisons for November 2000 show better agreement. Although the GOCART simulations of AOT may at times agree with AOT derived from Raman lidar and Sun photometer measurements, the aerosol extinction profiles simulated by GOCART for September and November 2000 show less vertical variability than the Raman lidar profiles. These initial comparisons show that the model simulations of aerosol extinction in the lowest 1-2 km are significantly (>50%) smaller than the Raman lidar measurements. Currently, the reasons for these differences are not known and are under investigation.

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