Demonstration Measurements of Water Vapor, Cirrus Clouds, and Carbon Dioxide Using a High-Performance Raman Lidar

DAVID N. WHITEMAN,* IGOR VESELOVSKII,† MARTIN CADIROLA,# KURT RUSH,† JOSEPH COMER,@ JOHN R. POTTER,† AND REBECCA TOLA&

*NASA GSFC, Greenbelt, Maryland
† University of Maryland, Baltimore County, Baltimore, Maryland
# Ecotronics, Clarksburg, Maryland
@ Science Systems and Applications, Lanham, Maryland
& Barr Associates, Westford, Massachusetts

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ABSTRACT

Profile measurements of atmospheric water vapor, cirrus clouds, and carbon dioxide using the Raman Airborne Spectroscopic lidar (RASL) during ground-based, upward-looking tests are presented here. These measurements improve upon any previously demonstrated using Raman lidar. Daytime boundary layer profiling of water vapor mixing ratio up to an altitude of approximately 4 km under moist, midsummer conditions is performed with less than 5% random error using temporal and spatial resolution of 2 min and 60–210 m, respectively. Daytime cirrus cloud optical depth and extinction-to-backscatter ratio measurements are made using a 1-min average. The potential to simultaneously profile carbon dioxide and water vapor mixing ratio through the boundary layer and extending into the free troposphere during the nighttime is also demonstrated.

1. Introduction

Raman lidar is now regarded as one of the leading technologies for atmospheric profiling of water vapor (Melfi et al. 1989; Whiteman et al. 1992; Goldsmith et al. 1998; Turner et al. 2000), cirrus clouds (Ansmann et al. 1992a; Reichardt et al. 2002; Whiteman et al. 2004), aerosols (Ansmann et al. 1990; Ferrare et al. 2006), temperature (Arshinov et al. 2005; Behrendt et al. 2002; Di Girolamo et al. 2004), and other atmospheric constituents or properties. Experimental measurements using Raman lidar have been made of carbon dioxide (Riebesell 1990; Ansmann et al. 1992b) as well. Traditionally, most Raman lidar measurements based on laser sources in the near UV (approximately 350 nm) were limited to the nighttime. In the 1990s, advances in Raman lidar technology (high-power UV lasers and narrowband interference filters) and techniques (narrow field-of-view detection) resulted in systems operating in the near UV that measure water vapor and aerosols throughout the diurnal cycle (Turner et al. 2000). More recently (Whiteman et al. 2006a; Ferrare et al. 2006) the addition of the data acquisition technique that combines analog-to-digital and photon-counting electronics has permitted a considerable improvement in daytime Raman lidar performance by allowing full-strength signals to be acquired in the presence of elevated solar backgrounds. The combination of a large aperture; narrow field-of-view telescope; high-power UV laser; narrowband, high-transmission filters; and combined analog-to-digital and photon counting data acquisition may be used to optimize the performance of a daytime Raman water vapor lidar (Whiteman et al. 2006a; Ferrare et al. 2006) permitting convective structures in the water vapor field to be studied in the daytime (Whiteman et al. 2006a; Demoz et al. 2006). That same technique is used here in a new, high-performance Raman lidar to acquire profile measurements of atmospheric water vapor, cirrus clouds, and carbon dioxide that improve upon any previously demonstrated using Raman lidar. Scientific motivation will now be provided for the three measurement capabilities to be demonstrated. Following this, the lidar system...
that was used will be briefly described and then the demonstration measurements will be presented.

2. Scientific motivation

In differing ways, the profiles of water vapor, cirrus clouds, and carbon dioxide are important in the atmospheric sciences. We provide here some brief details concerning the importance of each.

a. Water vapor

Water vapor is one of the most important components of the atmosphere from considerations of both weather and climate, yet it is one of the most difficult to quantify due to its high variability on short time and space scales. Advances in water vapor profiling capabilities are sought to improve quantitative precipitation forecasting (Weckwerth et al. 2004) and to improve our ability to quantify and study mesoscale meteorological systems (Demoz et al. 2005, 2006; Wulfmeyer et al. 2006). Raman lidar is a well-established technology for profiling water vapor and other quantities in the troposphere, is used by a large number of groups around the world and is technologically simple enough to permit autonomous operation (Goldsmith et al. 1998). Improvements in Raman lidar technology and techniques that reduce the measurement uncertainty will therefore be significant to the atmospheric sciences.

b. Cirrus clouds

Cirrus clouds strongly influence the radiation balance of the earth. Some studies have shown that subvisual cirrus clouds may cover as much as 70% of the Tropics (Wang et al. 1996) and yet these are the clouds that are most difficult to detect using passive sensors and that can even go undetected during the daytime by low-pulse-energy lidar systems (Comstock et al. 2002). Space-based lidar systems such as Geosciences Laser Altimetry System (GLAS; Spinhirne et al. 2005) and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO; Liu et al. 2004) have the ability to detect cirrus clouds globally and develop statistics of cirrus clouds not possible with passive sensors. However, to calculate cirrus cloud optical depths the backscatter measured by space-based lidar must be converted to extinction assuming some value for the extinction-to-backscatter ratio, otherwise known as the lidar ratio. Recent work (Whiteman et al. 2004) has shown that this value can vary by a factor of 2 in very cold clouds depending on whether the cloud was hurricane-induced or air-mass-motion-induced. Therefore, it is important to quantify cirrus cloud properties under a range of measurement conditions to assess the natural range of variability of the cirrus cloud lidar ratio. Such measurements can be acquired both by High Spectral Resolution lidar (HSRL; DeSlover et al. 1999) and Raman lidar techniques. Here we concentrate on the simpler Raman lidar approach.

c. Carbon dioxide

The combination of the use of carbon-based fuels and the reduction in photosynthesis due to the clearing of land has caused concentrations of carbon dioxide (CO$_2$) and methane (CH$_4$) to now be higher than they have been for at least 100 000 yr. The challenge of accurately modeling and therefore predicting carbon amounts in the atmosphere is illustrated by the high precision required to study some of the key processes driving carbon flux in the atmosphere. Space-based sensors are challenged to measure changes in the column content of CO$_2$ of less than 1%. However, most of the short-term variation in the column content of CO$_2$ occurs within the atmospheric boundary layer where CO$_2$ concentrations may increase by 5% to 10% overnight particularly closest to the surface (Bakwin et al. 1998). Ground-based and airborne sensors are both closer to the region of maximum variation in CO$_2$ and can be developed more quickly than space-based sensors. Therefore, as space-based systems are developed, it makes sense to pursue attractive ground-based and airborne technologies that can help improve our understanding of the carbon cycle. Raman lidar is an attractive option to consider for these measurements since simultaneous measurements of CO$_2$ and H$_2$O are possible thus permitting these generally anticorrelated quantities to be studied in the same volume of the atmosphere. The instrument used to make the measurements demonstrated here is described below.

3. The Raman Airborne Spectroscopic lidar

The Raman Airborne Spectroscopic lidar (RASL) was developed under the support of the National Aeronautics and Space Administration (NASA) Instrument Incubator Program. RASL consists of a high-power (17.5 W) Nd:YAG laser emitting at the frequency-tripled wavelength of 354.7 nm, a 0.6 Dall–Kirkham telescope operated at 0.25 mrad field-of-view dichroic beamsplitters and interference filters that select a set of spectral measurements, photomultiplier tubes that detect the signal, and combined analog-to-digital and photon counting data acquisition. The specifications of RASL during the tests that are described here are contained in Table 1.
The measurements presented here were acquired during ground-based testing of RASL that occurred from the Earth Sciences Building of NASA/Goddard Space Flight Center (GSFC) in 2004 and 2005. These measurements benefited from newly developed interference filters that were the result of a research effort funded by the NASA Advanced Component Technology Program performed jointly by Barr Associates and NASA/GSFC. The end result of that research effort was the manufacture of several narrowband, high-transmission UV interference filters. The particular filters that were used in the measurements presented here are described in Table 2. Details concerning RASL spectral measurements will now be provided.

### RASL spectral measurements

RASL is designed to measure the Rayleigh–Mie signal at the laser wavelength in unpolarized, parallel, and perpendicular polarizations. It also measures Raman scattering from molecular water vapor, nitrogen, and either oxygen or carbon dioxide. These Raman features have varying spectral locations and widths and thus different filter center wavelengths (CWLs) and bandwidths (BWs) are required to measure the constituents of interest here: water vapor, liquid water, molecular nitrogen, and carbon dioxide.

Water vapor is an asymmetric top molecule where the Q branch of the $v_1$ component of the Raman vibrational spectrum has a band origin located at a shift of $3657 \text{ cm}^{-1}$ (Avila et al. 2004) from the exciting line and spans approximately $20 \text{ cm}^{-1}$, which corresponds to a width of $0.3 \text{ nm}$ when excited at $354.7 \text{ nm}$. The use, therefore, of a 0.25-nm filter, as in the present case, introduces a temperature sensitivity to the measurements that is accounted for using published techniques (Whiteman 2003a,b). The CWLs and BWs were chosen after considerations of signal throughput, background light rejection, and the temperature sensitivity of Raman scattering. The out-of-band rejection specification for the water vapor filter, for example, was determined such that the contamination of the backscatter signal on the water vapor mixing ratio measurement was less than 0.1% under dry upper-tropospheric conditions. This resulted in an optical density of 12 at the laser wavelength.

The filter at $386.7 \text{ nm}$ was specified to transmit the Raman vibrational Q branch of molecular nitrogen and reject the rotational parts of the Raman $N_2$ spectrum. Approximately 85% of the full vibrational Raman $N_2$ cross section is thereby transmitted (Measures 1984), but the temperature dependence associated with transmitting just a portion of the Stokes and anti-Stokes components of the spectrum (Whiteman 2003a) is minimized since the cross section of the Q branch is essentially temperature independent (Avila et al. 2004). The filter passband was centered on the Q branch of $N_2$ by tilt-tuning the filter. High-resolution spectroscopy indicates that the Q branch of $N_2$ consists of closely spaced lines over a spectral interval of approximately $5 \text{ cm}^{-1}$ (Bendtsen and Rasmussen 2000). This translates to approximately $0.075 \text{ nm}$ in wavelength space at the Raman-shifted wavelength of $386.7 \text{ nm}$. Variations in laser output wavelength could cause a varying fraction of the Q-branch intensity to be transmitted by the filter. The Continuum 9050 laser in use in this experiment was not injection seeded, however. We observed changes in the transmitted intensity of the Raman $N_2$ signal when the water flow in the internal cooling loop of the laser cycled on and off. This wavelength variation was accompanied by changes in the polarization purity of the laser. We concluded that the cycling of internal water flow cooling the laser optical components induced thermal stresses that changed the gain and polarization characteristics of the lasing media thus producing changes in output wavelength and polarization purity.

| Laser | Nd:YAG (355 nm), 350 mJ per pulse, 50 Hz (Continuum 9050) |
| Telescope | Custom, athermal, 0.6 m (DFM Engineering) |
| Data acquisition | 250-MHz photon counting and 20-MHz analog detection (Licel) |
| Range resolution Measurements | 7.5 m |
| Wavelength (nm)/bandpass (nm) | Water vapor/407.5/0.25 (Barr Associates) |
| | Liquid water/403.2/6.0 (Barr Associates) |
| | Nitrogen/386.68/0.1 (Barr Associates) |
| | Oxygen/375.4/0.3 or CO$_2$/371.71/0.1 (Barr Associates) |
| | Elastic unpolarized/354.7/0.3 (Barr Associates) |
| | Elastic parallel polarized/354.7/0.3 (Barr Associates) |
| | Elastic perpendicular polarized/354.7/0.3 (Barr Associates) |
| Detectors | Hamamatsu R1924 PMTs. Products for research bases. |
| Field of view | 0.25 mrad |
These changes in the laser output characteristics occurred when the external laser cooling water temperature was kept below 16°C. Raising the external laser cooling water to 18°C eliminated the cycling of the internal water flow and the associated wavelength variations as confirmed by Burleigh pulsed wavemeter measurements. The polarization purity variations were also eliminated as confirmed by RASL depolarization measurements that showed no significant variation in the background value of molecular depolarization.

Carbon dioxide measurements were performed by centering a filter, again using tilt-tuning, on the Q-branch feature of the Raman $2\nu_2$ feature of CO$_2$, which is located at 371.7 nm when excited by 354.7-nm radiation. This is approximately coincident with the twenty-first line in the anti-Stokes component of the ro-vibrational spectrum of oxygen, which is a source of potential contamination for the measurement of CO$_2$ as will be discussed in section 7a.

### 4. Water vapor mixing ratio measurements

On 26 July 2005, RASL was operated from the ground over a period of approximately 14 h from early morning until late in the evening in order to test its upward-looking measurement capability. These measurements were acquired from the NASA/GSFC Earth Sciences building in Greenbelt, Maryland. On this day, the aerosol optical thickness, as reported by a collocated sun photometer, varied between approximately 0.4 and 0.9 at 340 nm indicating quite hazy conditions. Increased aerosol optical thickness significantly increases the sky brightness in the visible and near UV in the daytime, thus degrading Raman lidar performance. Increased aerosol optical thickness also increases the signal lost due to extinction during the round trip from the laser to the scattering medium and back to the telescope, which also degrades Raman lidar performance. The time series of RASL measurements of water vapor mixing ratio, made using filters 1 and 2 listed in Table 2, is shown in Fig. 1. The data were acquired using 1-min temporal and 7.5-m spatial resolution. The data were then processed using a 3-min sliding window in the time domain and a sliding window in the vertical domain that varied from 90 to 330 m. The resulting temporal and spatial resolution of the water vapor mixing ratio measurements, determined by the half-power point in a Fourier spectral analysis, was approximately 2 min and between 60 and 210 m, respectively. The vertical resolution of the processed data varied as follows: <1 km: 60 m, 1–2 km: 100 m, 2–3 km: 140 m, 3–4 km: 180 m, >4 km: 210 m. The measurements were calibrated against the total precipitable water (PW) measured by a collocated SuomiNet GPS system (Whiteman et al. 2006b) by performing a best fit of GPS and lidar-derived PW during cloud-free portions of the measurement period. High noon occurred at approximately 1800 UTC when the solar zenith angle reached approximately 20°. The daytime boundary layer top can be observed in the image at heights that range between 1.5 and 2 km. An elevated moist layer is observed to descend from approximately 4.5 km to less than 3 km over the period of the measurements. Despite the bright conditions present, an additional moist layer can be discerned to descend from 6 km to approximately 5 km during the measurements. Boundary layer convective cells, which supported the development of cumulus clouds at altitudes of 1.5 to 1.8 km, can be observed in the water vapor field between 1800 and 2100 UTC below an altitude of 1.5 to 1.8 km. The vertical striping of the image above the boundary layer at approximately 1600 and 1950 UTC is due to clouds that developed at the top of the boundary layer. Times shown with values larger than 2400 UTC are on 27 July.

A comparison of these RASL measurements made in Greenbelt, Maryland, and a Vaisala RS-80H radiosonde launched from the Howard University Research Campus in Beltsville, Maryland—a distance of approximately 10 km from GSFC—is shown in Fig. 2. The location of features in the vertical and the overall cali-

### Table 2. Specifications of interference filters used for the measurements presented here. BW refers to the full-width half-maximum bandwidth of the filter, CWL to the center wavelength of the filter, and T to the transmission.

<table>
<thead>
<tr>
<th>CWL (+0.02/−0.00 nm)</th>
<th>BW (±0.02 nm)</th>
<th>T (%)</th>
<th>General blocking</th>
<th>Additional blocking</th>
<th>Measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>407.50</td>
<td>0.25</td>
<td>70</td>
<td>OD6 @ 200–1200 nm</td>
<td>OD12 @ 354.7 nm</td>
<td>Raman water vapor</td>
</tr>
<tr>
<td>386.68</td>
<td>0.1</td>
<td>60</td>
<td>OD6 @ 200–1200 nm</td>
<td>OD12 @ 354.7 nm</td>
<td>Raman nitrogen</td>
</tr>
<tr>
<td>371.71</td>
<td>0.1</td>
<td>40</td>
<td>OD6 @ 200–1200 nm</td>
<td>OD12 @ 354.7 nm</td>
<td>Raman carbon dioxide</td>
</tr>
</tbody>
</table>
bration of the two measurements are in good agreement. These measurements occurred at 1300 UTC when the sun was \( \sim 20^\circ \) above the horizon and daytime mixing in the boundary layer had not yet developed to a significant degree. Therefore, it is likely that the water vapor field was reasonably homogeneous between the two sites due to the stable atmospheric conditions of the previous evening. The radiosonde/lidar comparison shown supports the conclusion that the layered features observed in Fig. 1 are realistic. Furthermore, both Fig. 2 and comparisons of water vapor mixing ratio measurements derived from the first 30 m of RASL data and those from a Paroscientific Met3A sensor (not shown) mounted 10 m above the laboratory in which RASL was located showed agreement typically within better than 10\% in the lowest portions of the profile even though no overlap correction was applied to RASL data. The lidar overlap function can introduce height-dependent biases in the lidar measurements. However, Raman lidar water vapor measurements are performed using a ratio of signals from the water vapor and nitrogen channels (Whiteman 2003b). The good agreement of the lidar measurements and radiosonde in the lowest levels implies that the lidar system overlap functions for the water vapor and nitrogen channels largely cancel in the ratio. The comparison of the time series of total precipitable water vapor measurements from RASL and GPS also showed good agreement except in the presence of clouds, which attenuated the laser beam and prevented extended profiling of the atmospheric column.

The random errors in the water vapor mixing ratio data were quantified at three times in Fig. 1 to study the evolution of random errors as a function of sun angle and therefore sky brightness. Random errors are calculated using Poisson statistics after converting the analog data to a virtual photon-counting scale. Previous analysis (Whiteman et al. 2006a) has shown that this technique of analyzing the analog-to-digital data produces random errors that agree well with random errors determined using spectral techniques. Figure 3 presents the RASL water vapor mixing ratio profiles and the random error at 13, 18, and 26.5 UTC (2.5 UTC July 27) when the solar zenith angles were 70\(^\circ\), 20\(^\circ\), and 102\(^\circ\), respectively. The latter value indicates that the sun was 12\(^\circ\) below the horizon. These profiles possess the same temporal and spatial resolution as shown in the image of Fig. 1. A general characteristic of the upward-looking RASL measurements is the increase in random error below approximately 0.6 km. This is due to re-

Fig. 1. Water vapor mixing ratio measurements made by the upward-looking RASL instrument using the narrowband water vapor and nitrogen interference filters described in Table 2. Times greater than 24 UTC are on 27 July.
duction of the signal in the near field due to the use of a narrow field-of-view detection scheme. This is one of the consequences of the single field-of-view design of an airborne lidar system intended for downward-looking measurements. A supplemental smaller telescope can be used at wider field of view to reduce the near-field random errors (Whiteman et al. 2006a).

The profiles of the mixing ratio shown on the left-hand side of the figure indicate that on this day the boundary layer extended to an altitude of approximately 2 km and was characterized by mixing ratio values ranging roughly from 5 to 15 g kg$^{-1}$. A significant elevated moist layer existed between altitudes of approximately 2 and 4 km where mixing ratio values ranged between $\sim$1 and 7 g kg$^{-1}$. Above this layer and up to an altitude of 8 km, mixing ratio values ranged between 1 and 3 g kg$^{-1}$. The random errors are shown on the right side of the figure indicating that, even at 18.0 UTC (solar noon), the random error did not exceed 2% in the boundary layer (except for the near-field zone at altitudes less than 0.6 km), 4% in the elevated layer and ranged between 20% and 60% above the elevated layer up to an altitude of 8 km. The measurements acquired at 13.0 UTC when the sun was 20° above the horizon possessed less than 3% random error through the elevated layer and less than 8% below 6 km. The profile acquired at night possessed less than 7% random error up to an altitude of 8 km.

5. Cirrus cloud optical depth and extinction-to-backscatter ratio measurements

Generally, Raman lidar measurements of cirrus cloud optical depth and extinction-to-backscatter ratio have not been made in the daytime, although these measurements have been made routinely by the technologically more sophisticated High Spectral Resolution lidar (DeSlover et al. 1999). The recent use of pure
rotational Raman scattering coupled with a Fabry–Perot etalon for temperature profiling has also demonstrated the ability to measure cirrus cloud extinction during the daytime (Arshinov et al. 2005). But cirrus cloud optical depth measurements during the daytime using the simpler approach of measuring the vibrational Q branch of N$_2$ have not been demonstrated previously because of poor signal-to-noise measurements at cirrus altitudes. With the high performance specifications of RASL shown in Table 1, Raman lidar measurements of cirrus cloud optical depth and extinction-to-backscatter ratio have been made for the first time using vibrational Raman scattering during the daytime. Figure 4 shows upward-looking RASL measurements of cirrus cloud scattering ratio, optical depth, and extinction-to-backscatter ratio acquired with 1-min temporal resolution on 8 October 2004. The optical depth and extinction-to-backscatter ratios are determined for the entire cloud layer in a manner that minimizes the influence of multiple scattering on the calculation of these quantities (Whiteman et al. 2001a). The solar zenith angle was approximately 45° during this measurement period. The statistical uncertainty of both the optical depth and lidar ratio retrievals is less than 10%. These measurements are a demonstration that currently available technology permits Raman lidar systems measuring vibrational Raman scattering from molecular nitrogen to measure cirrus cloud optical properties during both daytime and nighttime.

6. Carbon dioxide measurements

The previous measurements shown here demonstrate improvements in well-established Raman lidar measurement capability. By contrast, Raman lidar profiling of CO$_2$ has received very little attention either theoretically or experimentally. To investigate the potential of a large power-aperture lidar such as RASL to measure the CO$_2$ profile in the atmosphere, therefore, it was studied first using numerical modeling.

a. Numerical simulations of Raman lidar CO$_2$ mixing ratio measurements

A numerical model that was previously validated for measurements of water vapor mixing ratio (Whiteman et al. 2001b) was used to simulate ground-based CO$_2$ Raman lidar measurements where the RASL specifications shown in Table 1 were used in the model. Night-time conditions and constant aerosol extinction of 0.05 km$^{-1}$ within the first 2 km were assumed. It should be noted that the natural quantity that is measured by a Raman lidar, whether in the case of water vapor (Whiteman et al. 1992) or CO$_2$ (Ansmann et al. 1992b), is the mixing ratio with respect to dry air. This is done by using Raman scattering from molecular nitrogen to normalize the water vapor or CO$_2$ signal. The numerical model simulates both CO$_2$ and N$_2$ signals based on atmospheric input profiles and other quantities (Whiteman et al. 2001b). The results are shown in Fig. 5. The simulations were performed assuming a 3-h average. The spatial resolution was as follows: <1.25 km: 75 m, 1.25–2.0 km: 150 m, 2.0–2.5 km: 250 m, 2.5–3.0 km: 400 m, above 3.0 km: 600 m.

The input to the model included a 10-ppm increase in CO$_2$ at a height of 2.2 km to simulate the depletion of CO$_2$ within the mixed layer that occurs during the daytime. Therefore, the input CO$_2$ profile simulates a pos-
Fig. 4. Measurements of cirrus cloud scattering ratio, optical depth, and layer mean extinction-to-backscatter (lidar) ratio made during the daytime on 8 Oct 2004 with a solar zenith angle of $-45^\circ$–$50^\circ$ using 1-min temporal resolution. The scattering ratio data are displayed with 30-m vertical resolution.
sible condition shortly after sunset since these Raman
lidar measurements can only be made at night due to
the weak nature of the Raman CO
\textsubscript{2}
signal. As shown in
Fig. 5, the 10-ppm difference between the mixed layer
and the free troposphere is easily resolved using the
measurement parameters that were simulated. The pre-
cision of the measurement decreases at each change in
vertical smoothing such that it remains below \(1.2\) ppm
at all altitudes up to \(3.5\) km using the vertical resolu-
tion mentioned.

On 19 September 2004, RASL was run for \(3\) h be-
"\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig5}
\caption{Model simulations of ground-based profiling of CO\textsubscript{2} during the nighttime. The parameters simulated are
0.6-m telescope and 17.5-W UV laser with an averaging time of \(3\) h. The resultant precision is below \(1.2\) ppm for
all altitudes below \(3.5\) km with vertical resolution ranging from \(75\) to \(600\) m. A free-tropospheric transition of \(10\)
ppm was simulated at approximately \(2\) km where the vertical resolution of the simulation was \(250\) m.}
\end{figure}

On 19 September 2004, RASL was run for \(3\) h be-
inning approximately \(1\) h after sunset and acquired
measurements that for the first time demonstrate the
potential to simultaneously profile atmospheric CO
\textsubscript{2} and H\textsubscript{2}O mixing ratio. These measurements are shown in
Fig. 6. These are likely the first ground-based CO
\textsubscript{2} profile measurements extending into the free tropo-
sphere as well. The CO
\textsubscript{2} measurements were scaled
based on ground-based measurements of CO\textsubscript{2} acquired
at the same time by assuming that the CO\textsubscript{2} concen-
tration at the surface was the same as at the lowest mea-
sured altitude of \(800\) m. The CO\textsubscript{2} calibration shown in
Fig. 6 obtained must therefore be considered only ap-
proximate. The water vapor measurements were cali-
breated as previously described by forcing the total pre-
cipitable water of the lidar profile to equal that mea-
sured by a collocated GPS sensor. Both the CO\textsubscript{2} and
H\textsubscript{2}O have been analyzed such that the vertical resolu-
tion is \(300\) m between \(1\) and \(2\) km, \(400\) m between \(2\) and
\(3\) km, \(500\) m between \(3\) and \(4\) km, and \(600\) m above \(4\)
km. The precision of the CO\textsubscript{2} mixing ratio measure-
ment obtained with these resolutions, determined from
the signal strength of the CO\textsubscript{2} and N\textsubscript{2} data assuming
Poisson statistics, remains below \(1.5\) ppm for altitudes
less than \(4\) km. The precision of the CO\textsubscript{2} measurement
is generally consistent with the model predictions
shown in Fig. 5. The standard error bars plotted on the
water vapor mixing ratio data shown in Fig. 5 are im-
perceptible on this scale.

\textbf{b. Error sources in the measurement of CO\textsubscript{2} using
Raman lidar}

The only known previous measurements of atmo-
spheric CO\textsubscript{2} (\(2\) \(\nu_2; 1285\) cm
\textsuperscript{-1}) using Raman lidar were
made in the 1980s in Germany (Riebesell 1990; Ans-
mann et al. 1992b). The conclusions based on that re-
search were that useful CO\textsubscript{2} measurements by Raman
lidar were unlikely because the interference from rota-
tional lines of O\textsubscript{2} was difficult to determine and fluo-
rescence of either optics or atmospheric particles could
contaminate the measurement at the \(\sim1\)-ppm level.
However, this earlier research was conducted using a
XeCl excimer laser, which has an output spectrum that
spans approximately \(0.4\) nm. This broad spectrum
makes the separation of O\textsubscript{2} and CO\textsubscript{2} more difficult than
the present use of narrowband interference filters and
an Nd:YAG laser with spectral output of \(\sim0.02\) nm. As
mentioned previously, the \(2\) \(\nu_2\) Raman spectrum for
CO\textsubscript{2} is approximately coincident with the twenty-first
line in the anti-Stokes component of the ro-vibrational
spectrum of oxygen. Calculations based on a \(0.1\)-nm
wide filter as used in the measurements presented here
indicate that the contribution of this O2 rotational line to the measured CO2 signal is less than 1% (~3–4 ppm). Rotational line strength modeling (Whiteman et al. 2001b) as a function of temperature can be used to predict the magnitude of this interference so that it can be subtracted out. We estimate that this reduces the uncertainty in the CO2 measurement due to O2 rotational line interference to 0.3 ppm or less.

A careful study of fluorescence of both optical components and atmospheric aerosols would be required as a part of further developing and validating a Raman lidar CO2 profiling system. Preliminary measurements acquired using a scanning spectrometer coupled to a Raman lidar receiver indicated no significant fluorescence contribution in the CO2 spectral region, even though fluorescence due to aerosols was observed at longer wavelengths during the same measurement period. These results are consistent with a study of naturally occurring aerosols performed close to NASA/GSFC that indicated the presence of an energy gap between the exciting line and the induced fluorescence spectrum (Pinnick et al. 2004). In this study, aerosol fluorescence mainly occurred between 300 and 500 nm when excited at 266 nm.

7. Summary and conclusions

Profile measurements of atmospheric water vapor, cirrus clouds, and carbon dioxide using the Raman Airborne Spectroscopic lidar (RASL) during ground-based, upward-looking tests were presented. These measurements improve on any previously published using the Raman lidar techniques involved. A combination of high-power UV laser (17.5 W at 354.7 nm), large-aperture (0.6 m) telescope operated at narrow field of view (0.25 mrad), narrowband filters (0.25, 0.1, 0.1 nm for water vapor, nitrogen, and carbon dioxide, respectively), and simultaneous analog-to-digital and photon-counting data acquisition were used to make these measurements. The water vapor measurements possessed 2-min temporal and 60–210-m spatial resolution. Except for a near-range zone of approximately 600 m, where random errors increase due to the dynamic range compression that is inherent in the use of a narrow field-of-view telescope for lidar measurements, random errors remained below 2% through the boundary layer and below 4% up to an altitude of approximately 4 km. These water vapor random error characteristics are significantly improved over recently published daytime Raman lidar water vapor measurements acquired during the International H2O Experiment (IHOP; Weckwerth et al. 2004) by the NASA/GSFC Scanning Raman lidar (SRL; Whiteman et al. 2006a,b). The analysis of errors from that experiment indicated that, under similar water vapor and sky brightness conditions, the SRL random error did not exceed 10% throughout the boundary layer that extended to a height of ~3.5 km and were sufficient to study convective processes in the daytime boundary layer (Demoz et al. 2006). The RASL measurements
presented here show at least a factor-of-2 lower random error in the boundary layer than the SRL under similar daytime measurement conditions. The Department of Energy’s Climate and Radiation Facility Raman lidar (CARL) has recently been upgraded (Ferrare et al. 2006) to use similar technology as demonstrated here by RASL. CARL is now capable of daytime water vapor measurements that are nearly equivalent to those of RASL demonstrated here. The main differences in performance parameters of the two systems is that the RASL water vapor and nitrogen interference filters possess slightly higher transmission than those in CARL and RASL uses a laser that emits 30%-50% more power than CARL.

The midsummer conditions of the water vapor measurements presented here are ideal for studying convection initiation and also aid the Raman lidar measurement process since measurement precision is governed by Poisson statistics where the random errors are related to the square root of the signal intensity (Whitean 2003b). If the absolute amount of water vapor were reduced by a factor of 10, but all other parameters were kept the same, the random errors would approximately triple compared with those presented here. A factor-of-10 reduction in water vapor mixing ratio represents dry, wintertime conditions in the midlatitudes and yet, extrapolating from the measurements presented here, the random errors in the daytime boundary layer under these dry conditions would remain below 6% (above 0.6 km). It is likely, however, that under wintertime conditions, the aerosol optical thickness would be lower than on this 26 July case, which would reduce the estimated uncertainty below the 6% value. The conclusion from these considerations is that a high-performance Raman lidar system such as demonstrated here can provide daytime boundary layer water vapor mixing ratio measurements with random errors remaining substantially below 10% under a wide range of measurement conditions.

For the first time, cirrus cloud optical depth and extinction-to-backscatter ratio (lidar ratio) were quantified in the daytime using a measurement of Raman vibrational scattering from molecular nitrogen. Using 1-min temporal resolution, both optical depth and lidar ratio were quantified with approximately 10% uncertainty under daytime conditions where the solar zenith angle was approximately 45°–50°. This new measurement capability demonstrates the potential for cirrus cloud statistics to be acquired throughout the diurnal cycle using vibrational Raman lidar as has been possible with the HSRL and rotational Raman lidar techniques.

Carbon dioxide measurements using Raman lidar were also studied and performed. While these measurements must certainly be considered preliminary given that there are error sources that have not yet been addressed, we presented measurements that for the first time indicated the potential to simultaneously acquire profiles of atmospheric carbon dioxide and water vapor mixing ratio extending into the free troposphere. The carbon dioxide measurements were approximately scaled by comparison with a ground-based measurement of CO₂. The random error of the measurements agreed well with predictions based on numerical simulation. Error sources in the measurement of CO₂ using Raman lidar were considered. The interference of rotational lines from O₂ was estimated to contribute less than 1% to the total CO₂ signal; O₂ rotational line modeling could be used to account for this contribution and reduce its influence in the error budget to an estimated 0.3 ppm or less. Aerosol fluorescence was studied briefly and found to not contribute signal in the spectral band of CO₂. Additional error sources such as the lidar system overlap function and the differential transmission of the atmosphere were not considered here but must also be studied in any scientific application of this technique. When all of these error sources are considered, it is unlikely that a Raman lidar measurement of CO₂ with absolute accuracy of 1 ppm could be achieved with the current technology. However, for the study of carbon sources and sinks a quantification of nocturnal changes in CO₂ with a precision of 1 ppm (as opposed to an absolute accuracy of 1 ppm) is sufficient to provide useful information for modeling efforts that are designed to improve our understanding of carbon processes in the atmosphere. Therefore, the results presented here, while preliminary, indicate that continued development of Raman lidar profiling of CO₂ is a worthwhile research effort since sufficient signal strength exists for nocturnal CO₂ profile measurements to be made with a precision of ~1 ppm and for their correlation with H₂O to be studied.

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REFERENCES


———, U. Wandinger, M. Riebesell, C. Weitkamp, and W. Michae-