Differential Absorption Lidar to Measure Subhourly Variation of Tropospheric Ozone Profiles

Shi Kuang, John F. Burris, Michael J. Newchurch, Steve Johnson, and Stephanie Long

Abstract—A tropospheric ozone Differential Absorption Lidar system, developed jointly by The University of Alabama in Huntsville and the National Aeronautics and Space Administration, is making regular observations of ozone vertical distributions between 1 and 8 km with two receivers under both daytime and nighttime conditions using lasers at 285 and 291 nm. This paper describes the lidar system and analysis technique with some measurement examples. An iterative aerosol correction procedure reduces the retrieval error arising from differential aerosol backscatter in the lower troposphere. Lidar observations with coincident ozonesonde flights demonstrate that the retrieval accuracy ranges from better than 10% below 4 km to better than 20% below 8 km with 750-m vertical resolution and 10-min temporal integration.

Index Terms—Differential Absorption Lidar (DIAL), lidar, ozone, remote sensing, troposphere.

I. INTRODUCTION

OZONE IS A KEY trace-gas species within the troposphere. On the one hand, ozone is a precursor of the hydroxyl radical [1], which reacts with most trace species in the atmosphere. On the other hand, ozone is also a strong greenhouse gas influencing the climate by its radiative forcing [2]. In situ photochemistry and dynamic processes largely govern the distribution of tropospheric ozone [3]. Measuring ozone variability at high spatial and temporal resolution increases our understanding of tropospheric chemistry [4], [5], planetary boundary layer (PBL)—free-tropospheric exchange [6], [7], stratosphere—troposphere exchange [8]–[10], and the impact of lightning-generated NOx on tropospheric ozone [11]–[14].

Several techniques currently exist for making range-resolved measurements of tropospheric ozone. The most common technique is the balloonborne electrochemical concentration cell, which has monitored ozone since the 1960s. The ozonesonde profiles ozone with a 100-m vertical resolution from the surface to 35-km altitude with the accuracy of 5%–10% [15], [16]. Ozone sondes are attractive because of their low up-front cost and well-characterized behavior. However, they are not suitable for making continuous measurements because of logistical constraints. Interesting atmospheric phenomena that vary over 42 periods less than one day are particularly difficult to monitor using balloon ozonesondes. Satellite observations can derive total column ozone [17] and stratospheric ozone [18]–[22] and extend measurements to altitudes that are inaccessible to ozonesondes. More recently, high-quality satellite observations [19] of tropospheric ozone are becoming available [18], [23]–[33]. Although the satellite measurements can produce global maps of ozone, their current measurement uncertainties, along with their coarse spatial and temporal resolution, limit their ability to observe short-term variations in ozone. LIDARs can supplement these techniques when a requirement exists for ozone retrievals with higher temporal (from 1 min to several hours) and vertical resolution (from tens of meters to 2 km). For example, lidars will provide a long-term observations [33, 34] of ozone, as well as aerosol, temperature, and water vapor. Although the up-front costs are considerably higher than for a balloon ozonesonde operation, lidars can acquire profiles continuously under both daytime and nighttime conditions [35].

The spatial and temporal resolution of a lidar is more than sufficient to characterize short-term ozone variations for the atmospheric studies of vertical processes.

Differential Absorption Lidar (DIAL) has been successfully used to measure ozone within the PBL [36], [37], the free troposphere [38]–[44], and the stratosphere [45]–[48] for several decades. DIAL is evolving from ground-based and airborne systems to systems that are suitable for long-term deployment in space [49]. The technique derives ozone concentrations by analyzing how rapidly the backscattered signals at two separation but closely spaced wavelengths, one strongly absorbed by ozone and the other less strongly absorbed, diminish with altitude. This measurement does not require knowledge of the absolute signal intensities but, rather, only the relative change of the two signals with respect to altitude. Using electronically gated detection permits range-resolved measurements to a resolution as small as several meters over acquisition times of several minutes. The ozone DIAL discussed in this paper is located in the southeastern U.S. and thus provides a unique observational site within an interesting scientific area [50] to study trace-gas transport at the midlatitudes for both the polluted PBL and the free troposphere.
II. SYSTEM DESCRIPTION

Housed in the Regional Atmospheric Profiling Center for Discovery (RAPCD), the tropospheric ozone DIAL system is located at 34.7250° N, 86.6450° W on the campus of The University of Alabama in Huntsville (UAHuntsville) within the Huntsville city limits at an elevation of 206 m above sea level. It is designed for measurements within the PBL and the free troposphere during both daytime and nighttime. Because of UAHuntsville's location and occasional high temperature and humidity conditions, heavy aerosol pollution is sometimes present. Compared with the clean free troposphere, these aerosols require a larger dynamic range for the detection system because of their larger optical depth. Moreover, the rapid change of aerosol concentrations (e.g., due to convective activity) increases the measurement uncertainty for DIAL within the PBL and lower troposphere. Judicious system design choices and an effective aerosol correction scheme allow this system to produce high-quality ozone profiles under a variety of conditions.

103. Wavelength Selection

The selection of the 285- and 291-nm wavelengths results from the balance of the following three considerations: 1) optimizing the altitude range to make retrievals; 2) reducing the impact of the solar background during daytime operation; and 3) reducing the impact of aerosol interference upon the ozone retrieval. The DIAL wavelength selection is flexible and optimized for the local ozone distribution, the absorption arising from non-ozone species, the measurement range, and the specific system configuration, including the output power, the telescope mirror size, and the photomultiplier's (PMT's) dynamic range. Numerous publications (e.g., [51]) discussed the optimum wavelengths for tropospheric systems. Although shorter wavelengths can provide higher measurement sensitivity arising from the larger ozone differential cross section, they limit the maximum measurable range due to stronger attenuation of ozone absorption and Rayleigh (molecular) extinction and thus require more signal acquisition time. In addition, the shorter wavelengths require more dynamic range of the detection system and might require more altitude channels. With the current transmitter power, the online wavelength of 285 nm allows us to measure ozone up to 9 km under a clear sky and 7 km under aerosol loading with a 10-min temporal resolution. Because of the significant solar background during daytime operations, we choose 291 nm as the offline wavelength. Longer wavelengths will cause a significant increase in the solar background and reduce the signal-to-background ratio. To measure both wavelength channels using the same PMT and simplify the system design, we used a bandpass filter with a central wavelength of 286.4 nm and a full width at half maximum of 11 nm whose transmittance is ≤ 10−6 at wavelengths longer than 300 nm. For a bandpass filter, the integrated sky background over the filter bandwidth and the dark counts actually determine the background for both offline and online wavelengths. For our lidar configuration, the 285- and 291-nm wavelength region can provide sufficient signal-to-background ratios at 8 km under most sky conditions. The retrieval errors due to aerosol interference are a concern in the PBL and lower troposphere. These errors are not a simple function of the wavelength separations because reducing the separation to 3 nm will also decrease the differential ozone cross section. These errors are sensitive to the local aerosol composition, size distribution, and vertical profile. Although the aerosol interference can be lower when our online wavelength extends to the steepest part of the ozone absorption cross section, this will significantly sacrifice the maximum measurable range. Therefore, the 285–291-nm pair is the optimal choice to balance the maximum measurable altitude, the impact of aerosol differential backscattering, and the impact of solar background.

B. Hardware Components

Table I lists the characteristics of the RAPCD ozone DIAL system. The transmitter consists of two identical dye lasers pumped by two separate frequency-doubled Nd:YAG lasers (Fig. 1). A pulse generator triggers each laser pulse with a 25-ns separation between the alternate pulses. The dye lasers are software controlled to select the user-defined wavelength. The knife-edge method [52] determines that the divergences of both UV laser beams are less than 1 mrad. A 0.75-m focal-length, triple-grating monochromator (Acton Research Corporation) indicates that the actual wavelengths of the outgoing UV lasers are 285 and 291 nm within an uncertainty of 0.1 nm.

The receiving system currently operates with two separate 165 telescopes, as shown in Fig. 2. The high-altitude receiver uses a 40-cm Newtonian telescope, and the low-altitude channel employs a 10-cm Cassegrain telescope. The large telescope 168 routinely makes measurements from 3 to 8 km and, on occasion, measures ozone at 12 km. Employing a 1.5-mrad field of view (FOV), the large telescope achieves full overlap between the laser and receiver at about 3 km. Larger FOVs lower the altitude at which full overlap occurs but significantly increase solar background. The small telescope system currently retrieves ozone between 1 and about 5 km with a typical 175 FOV of 4.3 mrad. The future plan is to extend the retrievals down to about 200 m with an additional altitude channel in the 177 small telescope. The bandpass filters used to restrict the solar background for both receivers have a transmittance of 35% at 285 nm and 20% at 291 nm.

The detection system of the RAPCD ozone DIAL uses both photon counting (PC) and analog detection to facilitate operations over both altitude channels. This detection combination provides the linearity of the analog signal in the strong-signal region and high sensitivity of the PC signal in the weak-signal region. An EMI 9813 QA PMT, which has been used extensively for many years on a number of Goddard Space Flight Center lidar systems [53], [54], is used in the high-altitude channel, while a small Hamamatsu 7400 PMT is used in the low-altitude channel. A photodiode detects the outgoing laser pulses, which trigger both the PMT gating circuits and the Licel transient recorder (TR) (TR40-80, Licel Company, Germany). The Licel TR offers the advantage of increased dynamic range by providing simultaneous measurements using both analog and digital outputs.
TABLE I
CHARACTERISTICS OF THE RAPCD OZONE DIAL SYSTEM

<table>
<thead>
<tr>
<th>System</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transmitter</td>
<td></td>
</tr>
<tr>
<td>Pump lasers</td>
<td>Nd:YAG, 20 Hz repetition rate, 5-7 ns pulse length, 300 mJ pulse(^{-1}) at 1064 nm, 50 mJ pulse(^{-1}) at 552 nm</td>
</tr>
<tr>
<td>Dye</td>
<td>Rhodamine 590 and 610</td>
</tr>
<tr>
<td>Emitted UV</td>
<td>4 mJ pulse(^{-1}) at 285 nm, divergence&lt;1 mrad</td>
</tr>
<tr>
<td></td>
<td>3 mJ pulse(^{-1}) at 291 nm, divergence&lt;1 mrad</td>
</tr>
<tr>
<td>Tuning range</td>
<td>277 to 303 nm for the final UV output</td>
</tr>
<tr>
<td>Receiver</td>
<td></td>
</tr>
<tr>
<td>Telescope</td>
<td>Newtonian, 40-cm diameter, f/4.5, 1.5-mrad FOV</td>
</tr>
<tr>
<td>Band-pass filter</td>
<td>Center wavelength at 286.4 nm with a 11-nm FWHM. Transmittance is 35% at 285 nm and 20% at 291 nm</td>
</tr>
<tr>
<td>Detector</td>
<td>Electron Tubes 9B15QA, about 28% quantum efficiency</td>
</tr>
<tr>
<td>Signal processing</td>
<td>LICEL Transient Recorder (TR40-80), 250-MHz maximum photon counting rate, 12-bit and 40-MHz analog-to-digital converter, 25-ns range resolution</td>
</tr>
</tbody>
</table>

Fig. 1. Transmitter diagram.

195 detection and PC. The Licel TR’s highest temporal resolution is 25 ns, corresponding to a fundamental range resolution of 197 3.75 m. It is necessary to gate the high-altitude channel off 198 for the first 10–15 \(\mu s\) and the low-altitude channel for the first 199 1 \(\mu s\) to maintain the PMT’s linearity and minimize the impact 200 of signal-induced bias (SIB) on the background count rate.

III. DATA PROCESSING

A. Raw Data Processing

Several operations, designed to improve the measurement precision, occur before the ozone retrieval. First, average the 205 signal returns over 10 min and 150 m. The temporal resolution 206 of the retrieval can be varied depending on the signal-to-noise 207 ratio (SNR). Second, apply a dead-time correction to the PC 208 signals. For PC at high counting rates, a second pulse arriving 209 at the discriminator before it has recovered from the previous 210 pulse will not be counted—a period known as dead time [55]. 211 Experiments with a function-generator-driven LED determine 212 this time to be 10 ns for the high-altitude channel and 4 ns 213 for the low-altitude channel. Our results show that the system 214 dead time obeys a nonparalyzable model following a simple 215 relationship, as in (1)[56], between the true count rate 216 and measured count rates 217 allowing the impact of dead time 218 on the data to be removed

\[
C_T = \frac{C_M}{1 - C_M T_d}.
\]  

Third, remove the signal background. The last 10 \(\mu s\) (400 219 fundamental bins) of signals ranging up to 30.72 km (far-range
220 limit), which are considered to be the background region where 221 no laser signal returns are expected, are averaged to give an 222 approximate background. Fourth, merge the parallel analog and 223 PC signals into a single profile [57] after removing the offset 224 between the analog and PC signals [58]. We found this offset to 225 be about 250 ns for our system by carefully comparing returns 226 derived with clouds on both the analog and PC channels. The 227 merged region requires that the ratio of PC to analog signals is 228 constant. Ratios that are not constant suggest either an incorrect 229 background subtraction or a wrong dead-time correction. The 230 merging threshold of the PC signal is typically 20 MHz for 231 the Hamamatsu PMT employed in our low-altitude channel 232 and 20–30 MHz for the EMI PMT used on the high-altitude 233 channel. Because DIAL retrievals depend on the quality of 234 both 285- and 291-nm signals, we combine the PC and analog 235 signals approximately at the same altitude for both lasers to 236 minimize the retrieval error due to the merging. Examples of 237 the ratio of PC to analog signals and their merged region to 238 the 285-nm signal are shown in Fig. 3. The merging threshold 239 is 20 MHz for both altitude channels. The fifth step involves 240 smoothing the signals to reduce random noise. Our configura- 241 tion currently employs a five-point (5 x 150 = 750 m) running 242 average applied to returns from all altitudes; smoothing reduces 243 the effective vertical resolution to 750 m.

244 After initial processing, an exponential-fit correction re- 245 moves SIB from the signal returns. This bias, caused by intense 246 light returns from the near range (also called signal-induced 247 noise), appears as a slowly decaying noise source superimposed 248 on the normal returns. The causes of the SIB are related to the 249 regenerative effects such as dynode glow, after-pulsing effect, 250 glass-charging effect, shielding effect, and helium penetration 251 [59]. SIB varies widely with different PMTs. For our case, the 252 SIB of the EMI 9813 is larger than that for the Hamamatsu 252 7400. SIB can persist for several hundreds of microseconds and 253 can exert a strong influence on data at the lidar’s upper range 254 where both signal and noise counts become comparable. With 255 uncorrected SIB, the raw signal falls off more slowly at higher 256 altitudes, resulting in lower retrieved ozone values. SIB usually 257 has more influence on the shorter wavelength channel, which 258 falls off more rapidly with altitude. Unless a mechanical shutter 259 physically blocks the optical path to the PMT to eliminate SIB, 260 a model must characterize its behavior. Cairo et al. [60] and 261 Zhao [61] have successfully used a double-exponential function 262 for this purpose. However, this correction increases measure- 263 ment uncertainties because both the scaling and exponential 264 lifetimes are difficult to determine without additional indepen- 265 dent measurements. A more practical technique is to employ 266 a single-exponential fit to the residual background [42], [43], 267 [62]. For the high-altitude channel, the function’s coefficients 268 are automatically determined using a single-exponential least 269 squares fit to data acquired approximately from 100 to 160 μs 270 after data acquisition starts where the SIB becomes dominant. 271 The start and length of the exponential fit vary with different 272 channels (either wavelength channels or altitude channels), 273 atmospheric structures, and lidar configurations because these 274 parameters affect the intensity of the detected signal. For our 275 low-altitude channel, the SIB is weaker than that of the high- 276 altitude channel because of the different PMT and weaker 277 signal. However, it is difficult to automatically determine the 278 fitting function for the low-altitude channel signal using the 279 least squares fitting method, particularly for the 285-nm sig- 280 nal, because the far-range signal after background correction 281 is not completely characterized by an exponential function 282 [Fig. 3(b)]. It is useful to optimize the exponential fitting 283

![Diagram of the receivers and detectors.](image-url)
function for the low-altitude channel using previous retrieval data and compare the data with coincident ozonesonde profiles.

The slope of the logarithm of the SIB fitting function remains for a particular configuration (i.e., outgoing power) and could slightly change for different configurations. Those retrievals corrected using the empirically derived exponential function agree with ozonesonde profiles up to 5 km within 5% bias. Fig. 3 shows the typical effect of the SIB correction and the model for the 285-nm signal. The model simulation employs the coincident ozonesonde measurement assuming no aerosol.

B. DIAL Retrieval

Excellent discussions concerning the DIAL technique occur in the publications by Measures [63], Kovalev and Eichinger [64], and Browell et al. [39]. The average ozone number density $n_{(r+\Delta r/2)}$ between ranges $r$ and $r + \Delta r$ can be expressed as the summation of the signal term $n_{(r+\Delta r/2)}$, the differential backscattering term $\Delta n_{(r+\Delta r/2)}^b$ and the differential extinction term $\Delta n_{(r+\Delta r/2)}^e$:

$$n_{(r+\Delta r/2)} = n_{(r+\Delta r/2)}^e + \Delta n_{(r+\Delta r/2)}^b + \Delta n_{(r+\Delta r/2)}^e.$$  \hspace{1cm} (2)

One can write the discrete forms of the three terms at the right side as follows:

$$\Delta n_{(r+\Delta r/2)}^b = \frac{1}{2\Delta r \Delta \sigma_{O_3}} \ln \left( \frac{P_{on}(r) P_{off}(r+\Delta r)}{P_{on}(r+\Delta r) P_{off}(r)} \right)$$  \hspace{1cm} (3)

$$\Delta n_{(r+\Delta r/2)}^e = \frac{1}{2\Delta r \Delta \sigma_{O_3}} \ln \left( \frac{\beta_{on}(r) \beta_{off}(r+\Delta r)}{\beta_{on}(\Delta r) \beta_{off}(r)} \right)$$  \hspace{1cm} (4)

$$\Delta n_{(r+\Delta r/2)}^e = \frac{1}{\Delta \sigma_{O_3}} (\alpha_{on}(r+\Delta r/2) - \alpha_{off}(r+\Delta r/2))$$  \hspace{1cm} (5)

where the subscripts "on" and "off" represent the online (285 nm) and offline (291 nm) wavelengths, respectively, $P$ is the detected photon counts, $\beta$ is the total backscatter coefficient, $\sigma$ is the total scattering cross-section, $\alpha$ is the differential extinction coefficient and $\Delta \sigma_{O_3}$ is the total backscatter cross-section of ozone.
The uncertainty of $nb(st$ is statistical uncertainties. The gray envelope represents ±10% uncertainty of the coincident ozonesonde profile. (b) Joined DIAL retrieval from the two altitude channels and its combined one-sigma statistical uncertainty.

Typically, the low- and high-altitude channels join between 3.3 and 4.4 km. Fig. 4 shows an example of a joined ozone profile, as well as the combined one-sigma statistical uncertainties.

D. Aerosol Correction

In a polluted area, aerosols can be a dominant error source in the lower troposphere. Based on (4) and (5), the vertical gradient of aerosol backscattering determines $\Delta n^b$, and the 340 magnitude of the differential aerosol extinction coefficient $\Delta n^e$ determines $\Delta n^e$. The largest aerosol correction usually occurs in an inhomogeneous aerosol layer (i.e., the top of the PBL). One can solve for the ozone and aerosol profiles simultaneously with only two wavelengths by assuming appropriate Ångström exponents and constant lidar ratios [66, 67]. If a third wavelength 346 is available and is close to the DIAL wavelength pair, one can use the dual-DIAL technique [68, 69] to reduce the error due to aerosol. When the third wavelength is far from the DIAL wavelength pair, one can use the method suggested by Browell et al. [39] to correct the aerosol interference. Without the third wavelength, we employ an iterative procedure to retrieve ozone and correct aerosol effects. To illustrate this method, start with the following for the ozone number density using only the 291-nm signal [63]

$$n(r+\Delta r/2) = \frac{1}{2\sigma_{O3}\Delta r} \left\{ \ln \left( \frac{P_0}{P_0 + \Delta r} \right) - \ln \left( \frac{\beta_0^M}{\beta_0^M + \alpha_0^A} \right) - \frac{\alpha_0^M}{(r + \Delta r/2) \Delta r} \right\}$$

$$= \frac{1}{2\sigma_{O3}\Delta r} \left\{ \ln \left( \frac{P_0}{P_0 + \Delta r} \right) - \ln \left( \frac{\beta_0^M}{\beta_0^M + \alpha_0^A} \right) - \frac{\alpha_0^M}{(r + \Delta r/2) \Delta r} \right\}$$

$\sigma_{O3}$ is the differential ozone absorption cross section. $P$, $\beta$, and $\alpha$ are dependent on $r$ and the wavelength. Strictly speaking, $\Delta \sigma_{O3}$ is $r$ dependent as well because it is a function of temperature, which varies with $r$. By ignoring the differential scattering and extinction from non-ozone species, the DIAL equation reduces to only $n^e$. $\Delta n^b$ arises from aerosol differential backscattering. $\Delta n^e$ consists of differential Rayleigh extinction, aerosol extinction, and non-ozone gaseous absorption. Including these terms and constant $\alpha$ and extinction from non-ozone species, the DIAL equation reduces to only two altitude channels. The error bars represent the one-sigma statistical uncertainties. The gray envelope represents ±10% uncertainty of the coincident ozonesonde profile. (b) Joined DIAL retrieval from the two altitude channels and its combined one-sigma statistical uncertainty.

321 C. Joining Retrievals From Two Adjacent Altitude Channels

Final retrievals result from joining the data from two altitude channels with a weighted average. We choose to join the final ozone retrievals instead of the raw signals because the SNRs of the two altitude channels at the joining altitude are significantly different. If the retrievals derived from two different channels are statistically independent, the best estimate of these measurements is the two-channel weighted average [65]

$$n_{best} = \frac{\sum_{i=1}^{2} w_i n_i}{\sum_{i=1}^{2} w_i}$$

where $n_i$ is the ozone retrieval of channel $i$ and the weight $w_i$ is the inverse square of the corresponding statistical uncertainty $\varepsilon_i$, which will be discussed in Section V

$$w_i = \frac{1}{\varepsilon_i^2}$$

The uncertainty of $n_{best}$ is

$$\varepsilon_{n_{best}} = \left( \sum_{i=1}^{2} w_i \right)^{-1/2}$$

In a polluted area, aerosols can be a dominant error source in the lower troposphere. Based on (4) and (5), the vertical gradient of aerosol backscattering determines $\Delta n^b$, and the 340 magnitude of the differential aerosol extinction coefficient $\Delta n^e$ determines $\Delta n^e$. The largest aerosol correction usually occurs in an inhomogeneous aerosol layer (i.e., the top of the PBL). One can solve for the ozone and aerosol profiles simultaneously with only two wavelengths by assuming appropriate Ångström exponents and constant lidar ratios [66, 67]. If a third wavelength is available and is close to the DIAL wavelength pair, one can use the dual-DIAL technique [68, 69] to reduce the error due to aerosol. When the third wavelength is far from the DIAL wavelength pair, one can use the method suggested by Browell et al. [39] to correct the aerosol interference. Without the third wavelength, we employ an iterative procedure to retrieve ozone and correct aerosol effects. To illustrate this method, start with the following for the ozone number density using only the 291-nm signal [63]

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Typically, the low- and high-altitude channels join between 3.3 and 4.4 km. Fig. 4 shows an example of a joined ozone profile, as well as the combined one-sigma statistical uncertainties.

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where $\sigma_{O3}$ is the ozone absorption cross section, $\beta_{(r)}^M$ and $\beta_{(r)}^A$ are the molecular and aerosol backscatter coefficients at range $r$, respectively, and $\alpha_{(r)}^M$ and $\alpha_{(r)}^A$ represent the average molecular and aerosol extinction coefficients, respectively, between range $r$ and $r + \Delta r$. The subscript 291 is omitted for brevity because all backscatter and extinction parameters correspond to 291 nm. Solving for $\beta_{(r)}^A$, (9) becomes

$$\beta_{(r)}^A = \exp \left\{ \frac{1}{2} \left( \frac{P_{(r)}}{P_{(r+\Delta r)}} - 2n_{(r+\Delta r)\sigma_{O3}} \right) - 2 \left( \alpha_{(r-\Delta r/2)^2} + \alpha_{(r+\Delta r/2)^2} \right) \right\} - \frac{r^2 (\beta_{(r+\Delta r)}^M + \beta_{(r-\Delta r)}^M)}{(r + \Delta r)^2} - \beta_{(r)}^M. \tag{10}$$

Assuming that the lidar ratio (aerosol extinction-to-backscatter ratio), i.e., $S = \alpha_{(r)}^A / \beta_{(r)}^A$, is known for the 291-nm signal and further assuming that $\alpha_{(r+\Delta r/2)}^M \approx \alpha_{(r)}^A = S \beta_{(r)}^M$, (11) becomes

$$\alpha_{(r+\Delta r/2)}^A = \frac{\alpha_{(r)}^A}{S} = \frac{\beta_{(r+\Delta r/2)}^M}{S \beta_{(r)}^M}. \tag{12}$$

(10) only contains the following two unknown variables: the 297 aerosol backscatter coefficient $\beta_{(r+\Delta r)}^A$ and the ozone number density $n_{(r+\Delta r)}$. Molecular backscatter and extinction can be computed from nearby radiosonde data or from climatology. For the first iteration step, $n_{(r+\Delta r/2)}$ can be computed from (3) and inserted into (10). By assuming a start value $\beta_{(r)}^A$ at a reference range and a constant $S$ with range $\beta_{(r)}^A$ can be solved by (10). Then, the first $\beta_{(r)}^A$ profile is substituted back into (10) to compute the second estimate by using a more accurate form for $\alpha_{(r+\Delta r/2)}^A$ as

$$\alpha_{(r+\Delta r/2)}^A = S \left( \beta_{(r+\Delta r)}^A + \beta_{(r)}^A \right) / 2 \tag{12}$$

where $\beta_{(r)}^A$ is the value from the first estimate. With 377 several iterations of (10) and (12) we name this iteration the 378 "aerosol iteration", we can get a stable solution for $\beta_{(r)}^A$, which 379 does not change significantly from one iteration step to the next. 380 The aerosol iteration stop criterion is defined as $\xi_{(t)}^A < \xi_{(t)}^A$. 381 $\xi_{(t)}^A$ is the relative total difference of the backscatter coefficients 382 between two adjacent iteration steps and is defined as

$$\xi_{(t)}^A = \frac{1}{n} \sum_{r=1}^{n} \sum_{r'} \left| \beta_{(r,l)}^A - \beta_{(r,l+1)}^A \right| \tag{13}$$

where $l$ represents the iteration step, $r$ is the staring range 384 of the lidar retrieval, and $\beta_{(r,l)}^A$ are the backscatter coefficients 385 at range $r$ and iteration step $l$. $\xi_{(t)}^A$ is typically 0.01 for our 386 aerosol retrievals. Aside from $\xi_{(t)}^A$, the number of iterations 387 required for a stable solution is also related to the range-resolved 388 evolution of the signal. For simplicity, we assume that the power-law dependencies with wavelength for the aerosol extinction 390 and backscatter coefficients are the same although they can be different theoretically. $\Delta n_{(r+\Delta r)}^b$ and $\Delta n_{(r+\Delta r)}^e$ can be approximated as [39]

$$\Delta n_{(r+\Delta r)}^b \approx \frac{(4-\eta) \Delta \lambda}{2 \Delta r \sigma_{O3} \lambda_{off}} \left( \frac{B_{(r)}}{1+B_{(r)}} - \frac{B_{(r+\Delta r)}}{1+B_{(r+\Delta r)}} \right) \tag{14}$$

$$\Delta n_{(r+\Delta r)}^e \approx \frac{\Delta \lambda}{\Delta \sigma_{O3} \lambda_{off}} \left( \eta \alpha_{(r+\Delta r/2)}^A + 4 \alpha_{(r+\Delta r/2)}^M \right) \tag{15}$$

where $\eta$ is the Angström exponent, $\Delta \lambda$ is the wavelength separation, and $B_{(r)}$ is the aerosol-to-molecular backscatter ratio at the offline wavelength defined as

$$B_{(r)} = \beta_{(r)}^A / \beta_{(r)}^M. \tag{16}$$

The estimate for the aerosol-corrected ozone number density profile is then substituted into (10) to calculate an updated aerosol backscatter profile, which, in turn, is used to compute an updated aerosol-corrected ozone profile. This iteration is named "ozone iteration" to be distinct with the coupled aerosol 400 iteration process. A similar iteration stop criterion, $\xi_{(t)}^O < \xi_{(t)}^O$, 401 is as the aerosol iteration, can be defined for the ozone iteration 402 by replacing the backscatter coefficient in (13) with the ozone 403 number density typically, only two ozone iterations are re- 404 quired when $\xi_{(t)}^O$ is set equal to 0.001.

The lidar ratio (5) exhibits a wide range of variation with 406 different aerosol refractive indexes, size distributions, and hu- 407 midity [70]. The $S$ measurements have been made most fre- 408 quently at 308 [71] and 355 nm [72], [73]. The $S$ for our DIAL 409 wavelengths was assumed to be 60 sr$^{-1}$ [74] constant over the 410 measurement range for typical urban aerosols. The Angström 411 exponent ($\eta$) is often seen as an indicator of aerosol particle 412 size: Values greater than two correspond to small smoke parti- 413 cles, and values smaller than one correspond to large particles 414 like sea salt [75]. Most of the reported $\eta$'s for tropospheric 415 aerosols are measured at wavelengths longer than 300 nm with 416 a variation from zero to two [77], [78]. Considering that $\eta = 0.5$ could be relatively small when it is applied in the UV region, 418 we assume that $\eta = 0.5$ at our DIAL wavelengths for urban 419 aerosols [79].

Simulations were conducted to investigate the aerosol cor- 420 rection in the DIAL retrieval under an extremely large aerosol 421 gradient condition by assuming the aerosol, molecular, and 422 ozone extinction profiles at 291 nm shown in Fig. 5. The 423 hypothesis aerosol profile includes the following three basic 424 regimes: homogeneous, increasing, and decreasing extinction. 425 The aerosol extinction coefficients are set equal to $10^{-5}$ m$^{-1}$ 426 km$^{-1}$ below 1.2 km and above 3 km to represent a background value. 427 The resulting steep gradient between the low background and 428 high aerosol value provides an extreme test for the aerosol cor- 429 rection algorithm. The molecular extinction profile is derived 430 from the 1976 U.S. Standard Atmosphere [80]. The assumed 431 ozone extinction profile is constant with altitude and is based on 432 a number density of $1.5 \times 10^{12}$ molec $\cdot$ cm$^{-3}$ and an absorption 433 cross section of $1.24 \times 10^{-15}$ cm$^2$ $\cdot$ molec$^{-1}$ at 291 nm [81].

Fig. 6 shows the comparison of the ozone retrieval both with and without aerosol correction, as well as the calculated aerosol profile, at 291 nm. This example calculation assumes $\eta = 0.5$ and $S = 60$ sr$^{-1}$ are known exactly, and there are several iterations of aerosol retrievals.

The resulting steep gradient between the low background and high aerosol value provides an extreme test for the aerosol correction algorithm. The molecular extinction profile is derived from the 1976 U.S. Standard Atmosphere [80]. The assumed ozone extinction profile is constant with altitude and is based on a number density of $1.5 \times 10^{12}$ molec $\cdot$ cm$^{-3}$ and an absorption cross section of $1.24 \times 10^{-15}$ cm$^2$ $\cdot$ molec$^{-1}$ at 291 nm [81].

Fig. 6 shows the comparison of the ozone retrieval both with and without aerosol correction, as well as the calculated aerosol profile, at 291 nm. This example calculation assumes $\eta = 0.5$ and $S = 60$ sr$^{-1}$ are known exactly, and there are several iterations of aerosol retrievals.
is no signal measurement error. With a range resolution of 150 m, two ozone iterations produce the final aerosol-corrected ozone retrieval by setting $\xi_{\text{min}}^{\text{O_3}} = 0.001$. In the process of calculating the aerosol profile, aerosol iterations produce a stable aerosol solution by setting $\Delta_{\text{min}} = 0.01$, which is approximately identical to the model aerosol profile. The aerosol correction procedure reduces the retrieval errors from ±5% to about ±2%. The residual errors are due to the numerical integration and the approximation of (14) and (15). The quality of this iterative procedure depends on the choice of $S$ and $\eta$. According to (10), (14) and (15), $S$ affects the aerosol profile retrieval, while $\eta$ affects only the final ozone correction.

Fig. 7 shows the sensitivity test for $S$ and $\eta$ in the aerosol correction assuming that $S = 60$ and $\eta = 0.5$ are the correct values. Inaccurate estimates of $S$ or $\eta$ can yield retrieval errors up to about 20%. Larger $\eta$ will overestimate $\Delta n$, which produces less ozone, and vice versa. $\eta$ has a smaller impact on $\Delta n$ relative to the $4 - \eta$ factor. The impact of $S$ is larger in the inhomogeneous aerosol layer than in the homogeneous layer. The peak error is larger for underestimated $S$ relative to overestimated $S$.

We summarize the iterative procedure as follows.

1) Calculate the first estimate of the ozone concentration from (3).
2) Substitute the first estimated ozone into (10) to derive the aerosol backscatter profile for the offline wavelength, and iterate to obtain a stable solution with (12).
3) Calculate the differential aerosol backscatter and extinction corrections to obtain a second estimate of ozone using (14) and (15).
4) With the second ozone estimate, go back to step 2.

IV. MEASUREMENTS

Fig. 8 shows an ozone DIAL retrieval for 15 consecutive 472 hours from 12:56 local time, August 9, to 03:56, August 10, 473 2008, with 10-min temporal integration (12,000 shots) and 474 750-m vertical range resolution using the data processing described in the previous section. The aerosol correction was made only at altitudes between 1 and 4 km using the data from the low-altitude channel because of the negligible aerosol effects above 4 km. The aerosol time–height curtain [Fig. 8(a)] exhibits moderate aerosol activity between 1.5 and 2.5 km for the largest vertical backscatter gradient. The retrievals for the two altitude channels overlap between 3.3 and 4.4 km to produce the final ozone profiles [Fig. 8(c)] that agree well with the colocated ozonesonde (EN-487 SCI model ZZ with unbuffered 2% cathode solution) launched at 13:49 local time. The time–height curtain of ozone's
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Fig. 8. Ozone DIAL retrievals made on August 9–10, 2008. (a) Calculated aerosol extinction coefficient at 291 nm. The feature at 2 km, 14:00 is a cloud. (b) Aerosol correction for ozone DIAL retrieval. (c) Ozone DIAL retrieval after aerosol correction. The retrieval was made with a 750-m vertical range resolution and a 10-min temporal resolution. The colocated ozonesonde marked by a triangle was launched at 13:49 local time.

490 evolution shows a very interesting structure of multiple ozone 491 layers in the lower atmosphere that varies with time. One can 492 see the buildup and decay of various layers throughout this 493 12-h period. The high-frequency variation in the high-altitude 494 channel (≥ 6 km) results partly from lower SNR and higher 495 uncertainty of the SIB correction, both of which increase with 496 altitude. Fig. 9 shows the mean ozone profile and one-sigma 497 standard deviation for the 10-min vertical profiles between 498 12:56 and 15:06 local time in Fig. 8, as well as the coinci- 499 dent ozonesonde measurement. The high-altitude channel has a 500 standard deviation increasing with altitude due to the statistical 501 error distribution. Its standard deviation is less than 13 ppbv 502 below 8 km and increases to about 45 ppbv at 8.5 km where the 503 285-nm laser does not have sufficient SNR for ozone retrieval; 504 therefore, we terminate the retrievals at 8 km in Fig. 8. The stan- 505 dard deviation of the low-altitude channel retrievals is less than 506 5 ppbv below 4 km and reaches 8 ppbv at 5 km due to lower 507 SNR. The standard deviation at 2 km is a little larger than the 508 surrounding altitudes possibly because of larger ozone fluctu- 509 tions or larger uncertainties of the aerosol correction in the 509 507 ozone retrieval at the PBL top. The two altitude channels have 510 consistent mean retrievals in the overlap region with discrepan- 511 cies less than 5 ppbv and similar standard deviations at 3.3 km 512 which most likely reflect the true ozone short-term variations 513
above the PBL as shown in Fig. 8. The mean retrievals agree with the ozonesonde measurement within about 10 ppbv and have higher biases at the upper altitudes.

V. ERROR ANALYSIS

We divide the error budget of the DIAL retrieval into the following four categories: 1) statistical uncertainties $\varepsilon_1$ arising from signal and background noise fluctuations; 2) errors $\varepsilon_2$ associated with differential backscatter and extinction of non-ozone gases (O$_2$, SO$_2$, NO$_2$, etc.) and aerosols; 3) errors $\varepsilon_3$ due to uncertainties in the ozone absorption cross section; and 4) errors $\varepsilon_4$ related to instrumentation and electronics. $\varepsilon_1$ is a random error; $\varepsilon_2$, $\varepsilon_3$, and $\varepsilon_4$ are systematic errors. $\varepsilon_1$ can be written as [41]

$$\varepsilon_1 = \frac{1}{2n\Delta r\Delta \sigma_{O_3}} \sqrt{\sum_{\lambda} (SNR_{j,\lambda})^2}.$$  (17)

With the assumption of a Poisson distribution governing PC, the SNR at wavelength $\lambda$ and range registration $j$ becomes

$$SNR_{j,\lambda} = \frac{P_{j,\lambda}}{(P_{j,\lambda} + P_b + P_d)^{1/2}}.$$  (18)

where $P_b$ is the solar background counts and $P_d$ is the dark counts. It is straightforward to show that $\varepsilon_1$ is proportional to $(\Delta r^2 NAP_{L})^{-1/2}$, where $N$ represents the total number of shots, $A$ is the unobscured area of the telescope's primary mirror, and $P_L$ is the number of emitted laser photons. $\Delta r$ must be chosen large enough to produce an acceptably small error. Fig. 10 shows the estimated statistical errors for the high- and low-altitude channels for a 10-min integration and a 750-m range resolution. $\varepsilon_1$ is typically less than 10% below 4 km for our low-altitude channel and could be 20% at 5 km. This altitude performance gives us sufficient overlap for the two altitude channels under most atmospheric conditions. In the high-altitude channel, $\varepsilon_1$ exceeds 25% of the retrieval ozone near 8 ± 1 km, where we terminate the retrieval.

$\varepsilon_2$ includes the interference from O$_2$, SO$_2$, NO$_2$, air molecules, and aerosols. Table II summarizes the potential errors 544 in the DIAL retrieval for 285- and 291-nm wavelengths due to non-ozone absorption gases [84]-[88]. The calculation of 546 the oxygen dimer (O$_2$–O$_2$) interference includes some un-547 certainties due to the absorption cross-sectional measurement. 548 The O$_2$–O$_2$ absorption theory has not been entirely established 549 [89]. Local SO$_2$ and NO$_2$ profiling data are not available. How-550 ever, the estimated error due to either SO$_2$ or NO$_2$ using the 551 latest ground observation is less than 1%. The impact caused by 552 differential Rayleigh extinction results in an inaccuracy of less 553 than 1% using balloon ozonesonde retrievals of atmospheric 554 density or by employing climatological models.

The main concern comes from the aerosol interference, which depends on both the wavelengths and wavelength separation. Although the aerosol optical properties could be retrieved from a third wavelength, the differential effect for a DIAL wavelength pair still has some uncertainty due to the 560 assumption for lidar ratio and Ångström exponent. Within the 561 PBL, where the statistical errors are small, differential aerosol 562 backscattering and extinction dominate the error sources [39], 563 [41], [43]. However, it is reasonable to believe that the error 564 due to aerosol interference is smaller than 20% after the aerosol 565 correction, as shown in Section III-D.

The uncertainty in the Bass–Paur ozone cross sections is 567 believed to be less than 2% [81], [84], [89]. $\varepsilon_3$ will be less than 568 3% after considering the temperature dependence. 569 $\varepsilon_4$ could be caused by a misalignment of the lasers with the telescope FOV, imperfect dead time, or SIB correction. 571 Dead time distorts the near-range signal, and SIB distorts the far-range signal. Because the dead-time behavior is reliably 573 characterized, the error caused by SIB usually is larger than 574 the dead-time error. These errors related to the signal non-575 linearity can be experimentally diagnosed by a function generator-driven LED laser simulator [90], [91]. For the 10-min 577 integration data, $\varepsilon_4$ is estimated to be < 5% at 1–4 km for our 578
Ozone measurements within 10% from 1 to 4 km. The relatively high errors at about 2 km possibly relate to residual aerosol correction errors around PBL height. The lidar retrievals from the high-altitude channel agree with ozonesonde to within 20% below 8 km. The statistical error and the uncertainty associated with the SIB correction result in larger errors for the high-altitude channel above 6 km.

VI. Conclusion and Future Plans

The RAPCD ozone DIAL system measures tropospheric ozone profiles during both daytime and nighttime using the 603 285-291-nm wavelength pair. The low-altitude receiving channel makes ozone measurements at altitudes between 1 and 5 km using a 10-cm telescope and Hamamatsu R7400U PMTs. The high-altitude channel measures ozone between 3 and about 8 km using a 40-cm telescope and EMI 9813 PMTs. Model calculations demonstrate that the iterative aerosol correction procedure significantly reduces the retrieval error arising from differential aerosol backscatter in the lower troposphere where the quality of the aerosol correction depends on the accuracy of the a priori lidar ratio and Ångström exponent. A comparison of lidar retrievals and coincident ozonesonde measurements suggests that retrieval accuracy ranges from better than 10% after the application of an aerosol correction below 4 km to 20% above 6 km.

TABLE II

<table>
<thead>
<tr>
<th>Gases</th>
<th>$\Delta\sigma$, differential absorption cross-section (cm$^2$ molec$^{-1}$) for 285 and 291 nm</th>
<th>References for $\Delta\sigma$</th>
<th>Mixing ratio (ppbv)</th>
<th>References for mixing ratio</th>
<th>O$_3$ retrieval error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>O$_3$</td>
<td>1.15x$10^{-18}$</td>
<td>Bass and Paur 1981 [84]</td>
<td>60</td>
<td>1.5%</td>
<td></td>
</tr>
<tr>
<td>O$_2$</td>
<td>4.5x$10^{-27}$</td>
<td>Fally et al. 2000 [85]</td>
<td>2.1x$10^{-6}$</td>
<td>-0.9%</td>
<td></td>
</tr>
<tr>
<td>SO$_2$</td>
<td>-4.8x$10^{-20}$</td>
<td>Rufus et al. 2003 [86]</td>
<td>13$^b$</td>
<td>NREM 2006 [88]</td>
<td></td>
</tr>
<tr>
<td>NO$_2$</td>
<td>-2.25x$10^{-20}$</td>
<td>Bogumil et al. 2003 [87]</td>
<td>18$^c$</td>
<td>NREM 2006 [88]</td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>$\pm 1.5%$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*a due to O$_2$-O$_2$  
*b maximum 24-hr average in 1994. Latest local monitoring data available.  
*c Annual arithmetic average in 1993. Latest local monitoring data available.

TABLE III

<table>
<thead>
<tr>
<th>Errors</th>
<th>Low-altitude channel (1-4 km)</th>
<th>High-altitude channel (3-8 km)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. $e_1$, statistical error</td>
<td>&lt;10%</td>
<td>&lt;25%</td>
</tr>
</tbody>
</table>
| 2. $e_2$, interference by non-ozone species  
Aerosol | <20% | <5% |
| Non-ozone absorption gases | <1.5% | <4% |
| Rayleigh | <3% | <4% |
| 3. $e_3$, due to uncertainty in $\Delta\sigma$ | | |
| 4. $e_4$, due to SIB and dead-time | <5% | <10% |
| Total RMS error | <23% | <28% |

* The errors are estimated by assuming a 60 ppbv constant ozone mixing ratio in the troposphere for data with a 750-m vertical resolution and 10-min integration.
better than 20% for altitudes below 8 km with 750-m vertical resolution and 10-min integration. Error sources include statistical uncertainty, differential scattering and absorption from non-ozone species, uncertainty in ozone absorption cross section, and imperfection of the dead-time and SIB corrections.

Future improvements will overcome two major limitations of the current system by doing the following: 1) extending observations into the upper troposphere by replacing the current transmitters with more powerful ones and shifting the current wavelengths to longer ones to make higher-altitude nighttime measurements and 2) minimizing aerosol interference in the lower troposphere by adding a third wavelength (dual-DIAL technique). This lidar with expected improvements will provide a unique data set to investigate the chemical and dynamical processes in the PBL and free troposphere. The spatiotemporal variance estimates derived from the ozone lidar measurements will also be useful for assessing the variance of tropospheric ozone captured by satellite retrievals.

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Lighting-generated NOX and its impact on tropospheric ozone produc-
tion: A three-dimensional modeling study of a stratosphere-troposphere
experiment: Radiation, aerosols and ozone (STERAO) thunderstorm


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