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 (DIAL) system, developed jointly by The University of Alabama in Huntsville and the National Aeronautics and Space Administration (NASA), 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 256 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-troposphere 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 considerations. Interesting atmospheric phenomena that vary over 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 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 the 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 of the Network for the Detection of Atmospheric Composition Change [34], [35] are providing long-term observations 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.

The spatial and temporal resolution of a lidar is more than sufficient to characterize short-term ozone variations for the photochemical 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 separate, 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 and an effective aerosol correction scheme allow this system to produce high-quality ozone profiles under a variety of conditions.

A. 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 transmittance 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 reduce the aerosol differential backscattering 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 157-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 thick plano-convex grating monochromator (Acton Research Corporation) reduces the actual wavelengths of the outgoing UV lasers to 285 and 291 nm within an uncertainty of 0.1 nm.

The receiving system currently operates with two separate 165-cm Cassegrain telescopes, as shown in Fig. 2. The high-altitude receiver uses a 24-cm Newtonian telescope, and the low-altitude channel employs a 10-cm Cassegrain telescope. The large telescope system 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 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 191 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
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 532 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 1 mrad</td>
</tr>
<tr>
<td></td>
<td>3 mJ pulse⁻¹ at 291 nm, divergence 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></td>
<td>Welch Mechanical Designs</td>
</tr>
<tr>
<td></td>
<td>Cassegrain, 10-cm diameter, f/2.3, 4.3-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 9813QA, about 28% quantum efficiency</td>
</tr>
<tr>
<td></td>
<td>Hamamatsu R7400U-03, about 20% quantum efficiency</td>
</tr>
<tr>
<td>Signal processing</td>
<td>LICEL Transient Recorder (TR40-80), 250-MHz maximum photoncounting rate, 12-bit and 40-MHz analog-to-digital converter, 25-ns range resolution</td>
</tr>
</tbody>
</table>

Fig. 1. Transmitter diagram.

Signals. For PC at high counting rates, a second pulse arriving at the discriminator before it has recovered from the previous pulse will not be counted—a period known as dead time [55]. Experiments with a function-generator-driven LED determine this time to be 10 ns for the high-altitude channel and 4 ns for the low-altitude channel. Our results show that the system dead time obeys a nonparalyzable model following a simple relationship, as in (1) [56], between the true count rate $C_T$ and measured count rates $C_M$, allowing the impact of dead time $T_d$ on the data to be removed

$$C_T = \frac{C_M}{1 - C_MT_d}. \quad (1)$$

Third, remove the signal background. The last 10 μs (400 fundamental bins) of signals ranging up to 30.72 km (far-range 218 detection and PC. The Licel TR's highest temporal resolution is 25 ns, corresponding to a fundamental range resolution of 3.75 m. It is necessary to gate the high-altitude channel off for the first 10-15 μs and the low-altitude channel for the first 1 μs to maintain the PMT's linearity and minimize the impact 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 signal returns over 10 min and 150 m. The temporal resolution of the retrieval can be varied depending on the signal-to-noise ratio (SNR). Second, apply a dead-time correction to the PC
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 × 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 253 7400. SIB can persist for several hundreds of microseconds and 254 can exert a strong influence on data at the lidar’s upper range 255 where both signal and noise counts become comparable. With 256 uncorrected SIB, the raw signal falls off more slowly at higher 257 altitudes, resulting in lower retrieved ozone values. SIB usually 258 has more influence on the shorter wavelength channel, which 259 falls off more rapidly with altitude. Unless a mechanical shutter 260 physically blocks the optical path to the PMT to eliminate SIB, 261 a model must characterize its behavior. Cairo et al. [60] and 262 Zhao [61] have successfully used a double-exponential function 263 for this purpose. However, this correction increases measure- 264 ment uncertainties because both the scaling and exponential 265 lifetimes are difficult to determine without additional indepen- 266 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. The 271 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

Fig. 2. Diagram of the receivers and detectors.
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 range $r$ and $r + \Delta r$ can be expressed as the summation of the signal term $n_{(r+\Delta r/2)}^s$, the differential backscattering term $\Delta n_{(r+\Delta r/2)}^b$, and the differential extinction term $\Delta n_{(r+\Delta r/2)}^e$:

$$n_{(r+\Delta r/2)} = n_{(r+\Delta r/2)}^s + \Delta n_{(r+\Delta r/2)}^b + \Delta n_{(r+\Delta r/2)}^e. \tag{2}$$

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

$$n_{(r+\Delta r/2)} = \frac{1}{2\Delta r \Delta \sigma_{O3}} \ln \left( \frac{P_{on}(r)P_{off}(r+\Delta r)}{P_{off}(r)P_{on}(r+\Delta r)} \right) \tag{3}$$

$$\Delta n_{(r+\Delta r/2)}^b = -\frac{1}{2\Delta r \Delta \sigma_{O3}} \ln \left( \frac{\beta_{on}(r)\beta_{off}(r+\Delta r)}{\beta_{off}(r)\beta_{on}(r+\Delta r)} \right) \tag{4}$$

$$\Delta n_{(r+\Delta r/2)}^e = -\frac{1}{\Delta \sigma_{O3}} \left( \alpha_{on}(r+\Delta r/2) - \alpha_{off}(r+\Delta r/2) \right) \tag{5}$$

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, $\Delta \sigma_{O3}$ is...
Fig. 4. Example of a joined ozone retrieval for the lidar data in Fig. 3. (a) Separate retrievals of the 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.

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 determines $\Delta n^e$. The largest aerosol correction usually occurs in the 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 347 use the dual-DIAL technique [68, 69] to reduce the error due 348 to aerosol. When the third wavelength is far from the DIAL wavelength pair, one can use the method suggested by Browell 350 et al. [39] to correct the aerosol interference. Without the third 351 wavelength, we employ an iterative procedure to retrieve ozone 352 and correct aerosol effects. To illustrate this method, start with 353 the equation for ozone number density using only the 291-nm 354 signal [63]

\[
\frac{\Delta n}{\Delta r} = \frac{\Delta n_M}{\Delta r_M} + \frac{\Delta n_A}{\Delta r_A} - \frac{\Delta n_L}{\Delta r_L} + \Delta r \nabla n
\]

where $\Delta n_M$, $\Delta n_A$, and $\Delta n_L$ represent the gradients of signal, aerosol extinction, and aerosol backscatter, respectively. $
abla n$ is the vertical gradient of aerosol number density, and $\Delta r$ is the horizontal distance between the two DIAL wavelengths.

To correct for aerosol effects, we 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 equation for ozone number density using only the 291-nm signal [63].
366 where \( \sigma_{O3} \) is the ozone absorption cross section, \( \beta_A^{M}(r) \) and \( \beta_A^{L}(r) \) are the molecular and aerosol backscatter coefficients at range \( r \), respectively, and \( \alpha_M^{\Delta(r+\Delta r/2)} \) and \( \alpha_M^{\Delta(r-\Delta r/2)} \) 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 \( \beta_A^{L}(r) \), (9) becomes

\[
\beta_A^{L}(r) = \exp \left\{ \ln \left( \frac{P(r)}{P(r+\Delta r)} \right) - 2n(r+\Delta r;r)\sigma_{O3} \Delta r \right\} \\
- 2 \left( \alpha_M^{\Delta(r-\Delta r/2)} + \alpha_M^{\Delta(r+\Delta r/2)} \right) \Delta r \\
\frac{r^2 \left( \beta_A^{M(r+\Delta r)} + \beta_A^{M(r-\Delta r)} \right)}{(r+\Delta r)^2} - \beta_A^{M(r)}.
\]  

(10)

363 Assuming that the lidar ratio \( (\text{aerosol extinction-to-backscatter ratio}) \), i.e., \( S = \alpha_A^{M}/\beta_A^{M} \), is known for the 291-nm signal and further assuming that

\[
\alpha_A^{M(r+\Delta r/2)} \approx \alpha_A^{M(r)} = S\beta_A^{M(r)}
\]  

(11)

(10) only contains the following two unknown variables: the 367 aerosol backscatter coefficient \( \beta_A^{L}(r) \) and the ozone number 368 density \( n(r+\Delta r;r) \). Molecular backscatter and extinction can be 369 computed from nearby radiosonde data or from climatology. 370 For the first iteration step, \( n(r+\Delta r;r) \) can be computed from 371 (3) and inserted into (10). By assuming a start value \( \beta_A^{M(r)} \) at 372 reference range and a constant \( S \) with range, \( \beta_A^{L}(r) \) can be solved 373 by (10). Then, the first \( \beta_A^{L}(r) \) profile is substituted back into (10) 374 to compute the second estimate by using a more accurate form 375 for \( \alpha_A^{M(r+\Delta r/2)} \) as

\[
\alpha_A^{M(r+\Delta r/2)} = S \left( \beta_A^{M(r+\Delta r)} + \beta_A^{M(r)} \right)/2
\]  

(12)

376 where \( \beta_A^{M(r)} \) represents 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 \( \beta_A^{M(r)} \), which 379 does not change significantly from one iteration step to the next. 380 The aerosol iteration stop criterion is defined as \( \xi_A^{(l)} < \xi_A^{\text{min}} \). 381 \( \xi_A^{(l)} \) is the relative total difference of the backscatter coefficients 382 between two adjacent iteration steps and is defined as

\[
\xi_A^{(l)} = \frac{1}{r=1} \sum_{r=r_{ref}} \left| \beta_A^{(l)}(r) - \beta_A^{(l+1)}(r) \right|
\]  

(13)

383 where \( l \) represents the iteration step, \( r \) is the starting range 384 of the lidar retrieval, and \( \beta_A^{(l)}(r) \) is the backscatter coefficients 385 at range \( r \) and iteration step \( l \). \( \xi_A^{\text{min}} \) is typically 0.01 for our 386 aerosol retrievals. Aside from \( \xi_A^{\text{min}} \), the number of iterations 387 required for a stable solution is also related to the range resolution of the signal. For simplicity, we assume that the power-law dependences with wavelength for the aerosol extinction 390 and backscatter coefficients are the same although they can be different theoretically. \( \Delta n^{b(r+\Delta r)} \) and \( \Delta n^{e(r+\Delta r)} \) can be approximated as [39]

\[
\Delta n^{b(r+\Delta r)} \approx \frac{(4-\eta)\Delta \lambda}{2\Delta r \sigma_{O3} \lambda_{off}(r)} \left( \frac{B(r) - B(r+\Delta r)}{1 + B(r) + B(r+\Delta r)} \right)
\]  

(14)

\[
\Delta n^{e(r+\Delta r)} \approx - \frac{\Delta \lambda}{\sigma_{O3} \lambda_{off}} \left( \eta \alpha_A^{M(r+\Delta r/2)} + 4\alpha_M^{M(r+\Delta r/2)} \right)
\]  

(15)

where \( \eta \) is the Ångström exponent, \( \Delta \lambda \) is the wavelength separation, and \( B(r) \) is the aerosol-to-molecular backscatter ratio at the offline wavelength defined as

\[
B(r) = \beta_A^{M(r)}/\beta_A^{M}(r)
\]  

(16)

The estimate for the aerosol-corrected ozone number density 396 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, \( \xi_O^{\text{min}} < \xi_O^{\text{min}} \), as the aerosol iteration, can be defined for the ozone iteration 402 by replacing the backscatter coefficient in (13) with the ozone number density. Typically, only two ozone iterations are required when \( \xi_O^{\text{min}} \) is set equal to 0.001.

405 The lidar ratio (5) exhibits a wide range of variation with 406 different aerosol refractive indexes, size distributions, and hy- 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 Ångström 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], [76]. Most of the reported \( \eta \)’s for tropospheric 415 aerosol are measured at wavelengths longer than 300 nm with 416 a variation from zero to two [77], [78]. Considering that \( \eta \) 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].

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

435 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 438 that \( \eta = 0.5 \) and \( S = 60 \) sr\(^{-1} \) are known exactly, and there 439
Fig. 5. Aerosol, molecular, and ozone extinction coefficient profiles at 291 nm for a model calculation of extreme aerosol effects.

Fig. 6. Comparison of the simulated ozone retrieval without aerosol correction against that with aerosol correction using the iterative procedure. The Angström exponent ($\eta$) and lidar ratio ($S$) were assumed to be exactly known at 0.5 and 60 sr$^{-1}$, respectively, for the aerosol correction. The aerosol correction dramatically improves the ozone retrieval.

440 is no signal measurement error. With a range resolution of 441 150 m, two ozone iterations produce the final aerosol-corrected 442 ozone retrieval by setting $\xi_{\text{min}} = 0.001$. In the process of cal- 443 culating the aerosol profile, aerosol iterations produce a stable 444 aerosol solution by setting $\xi_{\text{min}} = 0.01$, which is approximately 445 identical to the model aerosol profile. The aerosol correction 446 procedure reduces the retrieval errors from ±50% to about 447 ±5%. The residual errors are due to the numerical integration 448 and the approximation of (14) and (15). The quality of this 449 iterative procedure depends on the choice of $S$ and $\eta$. According 450 to (10), (14) and (15), $S$ affects the aerosol profile retrieval, 451 while $\eta$ affects only the final ozone correction.

Fig. 7 shows the sensitivity test for $S$ and $\eta$ in the aerosol 453 correction assuming that $S = 60$ and $\eta = 0.5$ are the correct 454 values. Inaccurate estimates of $S$ or $\eta$ can yield retrieval errors 455 up to about 20%. Larger $\eta$ will overestimate $\Delta n^a$, which 456 produces less ozone, and vice versa. $\eta$ has a smaller impact 457 on $\Delta n^b$ relative to $\Delta n^a$ due to the $4 - \eta$ factor. The impact 458 of $S$ is larger in the inhomogeneous aerosol layer than in the 459 homogeneous layer. The peak error is larger for underestimated 460 $S$ relative to overestimated $S$ [82].

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 464 aerosol backscatter profile for the offline wavelength, and 465 iterate to obtain a stable solution with (12).
3) Calculate the differential aerosol backscatter and extinction 467 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. 7 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 de- 475 scribed in the previous section. The aerosol correction was 476 made only at altitudes between 1 and 4 km using the data 477 from the low-altitude channel because of the negligible aerosol 478 effects above 4 km. The aerosol time–height curtain [Fig. 8(a)] 479 exhibits moderate aerosol activity below 2 km with expected 480 concentration peaks at 13:49 local time. The time–height curtain of ozone’s 481

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 de- 475 scribed in the previous section. The aerosol correction was 476 made only at altitudes between 1 and 4 km using the data 477 from the low-altitude channel because of the negligible aerosol 478 effects above 4 km. The aerosol time–height curtain [Fig. 8(a)] 479 exhibits moderate aerosol activity below 2 km with expected 480 concentration peaks at 13:49 local time. The time–height curtain of ozone’s 481

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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 layers in the lower atmosphere that varies with time. One can see the buildup and decay of various layers throughout this 12-h period. The high-frequency variation in the high-altitude channel (≥ 6 km) results partly from lower SNR and higher uncertainty of the SIB correction, both of which increase with altitude. Fig. 9 shows the mean ozone profile and one-sigma standard deviation for the 10-min vertical profiles between 12:56 and 15:06 local time in Fig. 8, as well as the coincident ozonesonde measurement. The high-altitude channel has a standard deviation increasing with altitude due to the statistical error distribution. Its standard deviation is less than 13 ppbv below 8 km and increases to about 45 ppbv at 8.5 km where the 285-nm laser does not have sufficient SNR for ozone retrieval; therefore, we terminate the retrievals at 8 km in Fig. 8. The standard deviation of the low-altitude channel retrievals is less than 5 ppbv below 4 km and reaches 8 ppbv at 5 km due to lower SNR. The standard deviation at 2 km is a little larger than the surrounding altitudes possibly because of larger ozone fluctuations or larger uncertainties of the aerosol correction in the PBL top. The two altitude channels have consistent mean retrievals in the overlap region with discrepancies less than 5 ppbv and similar standard deviations at 3.3 km which most likely reflect the true ozone short-term variations.
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 (\( \text{O}_2, \text{SO}_2, \text{NO}_2 \), 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 \( [41] \)

\[
\varepsilon_1 = \frac{1}{2n\Delta T\Delta\sigma_{O3}} \left( \sum_{\lambda} \frac{1}{SNR_{j,\lambda}} \right)^{1/2}. \tag{17}
\]

The assumption of a Poisson distribution governing PC,
the SNR at wavelength \( \lambda \) and range registration \( j \) becomes

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

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 \pm 1 km, where we terminate the retrieval.
The RAPCD ozone DIAL system measures tropospheric ozone profiles during both daytime and nighttime using the 603-291-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 a priori lidar ratio and Ångström exponent.

VI. Conclusion and Future Plans

The RAPCD ozone DIAL system measures tropospheric ozone profiles during both daytime and nighttime using the 603-291-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 a priori lidar ratio and Ångström exponent.

A comparison of 12 lidar retrievals and their corresponding ozonesonde measurements is shown in Table III for a constant ozone mixing ratio of 60 ppbv. The statistical error and the uncertainty associated with the SIB correction result in larger errors for the high-altitude channel above 6 km.

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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.

The uncertainty in the SIB correction and the statistical errors dominate the error sources in the free troposphere and could be reduced by increasing the integration time or reducing the range resolution.

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 observations will also be useful for assessing the variance of tropospheric ozone captured by satellite retrievals.

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REFERENCES


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