

1 **Quantifying TOLNet Ozone Lidar Accuracy during the 2014** 2 **DISCOVER-AQ and FRAPPÉ Campaigns**

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23 **Abstract**

24 The Tropospheric Ozone Lidar Network (TOLNet) is a unique network of lidar systems that measure high-
25 resolution atmospheric profiles of ozone. The accurate characterization of these lidars is necessary to determine the
26 uniformity of the network calibration. From July to August 2014, three lidars, the TROPospheric OZone (TROPOZ)
27 lidar, the Tunable Optical Profiler for Aerosol and oZone (TOPAZ) lidar, and the Langley Mobile Ozone Lidar
28 (LMOL), of TOLNet participated in the “Deriving Information on Surface conditions from Column and Vertically
29 Resolved Observations Relevant to Air Quality” (DISCOVER-AQ) mission and the “Front Range Air Pollution and
30 Photochemistry Experiment” (FRAPPÉ) to measure ozone variations from the boundary layer to the top of the
31 troposphere. This study presents the analysis of the intercomparison between the TROPOZ, TOPAZ, and LMOL
32 lidars, along with comparisons between the lidars and other *in situ* ozone instruments including ozonesondes and a
33 P-3B airborne chemiluminescence sensor. The TOLNet lidars measured vertical ozone structures with an accuracy
34 generally better than $\pm 15\%$ within the troposphere. Larger differences occur at some individual altitudes in both the
35 near-field and far-field range of the lidar systems, largely as expected. In terms of column average, the TOLNet
36 lidars measured ozone with an accuracy better than $\pm 5\%$ for both the intercomparison between the lidars and
37 between the lidars and other instruments. These results indicate that these three TOLNet lidars are suitable for use in
38 air quality, satellite validation, and ozone modeling efforts.

39 **1. Introduction**

40 **1.1 TOLNet**

41 The Tropospheric Ozone Lidar Network (TOLNet) provides time-height measurements of ozone from the
42 planetary boundary layer (PBL) to the top of the troposphere at multiple locations for satellite validation, model
43 evaluation, and scientific research (Newchurch et al., 2016; <http://www-air.larc.nasa.gov/missions/TOLNet/>).
44 Particularly, these ozone measurements can serve to validate NASA’s first Earth Venture Instrument mission,
45 Tropospheric Emissions: Monitoring Pollution (TEMPO), planned to launch in 2019. A second objective of TOLNet
46 is to identify a brassboard ozone lidar instrument that would be suitable to populate a network to address an
47 increasing need for ozone profiles by scientists and managers within the air quality, modeling, and satellite
48 communities (Bowman, 2013).

49 TOLNet consists of five ozone lidars across the United States and one in Canada: the Table Mountain
50 tropospheric ozone differential absorption lidar (DIAL) at NASA’s Jet Propulsion Laboratory, the Tunable Optical
51 Profiler for Aerosol and oZone (TOPAZ) lidar at NOAA’s Earth System Research Laboratory (ESRL), the Rocket-
52 city Ozone (O_3) Quality Evaluation in the Troposphere (RO₃QET) lidar at the University of Alabama in Huntsville
53 (UAH), the TROPospheric OZone (TROPOZ) DIAL at NASA’s Goddard Space Flight Space Center (GSFC), the
54 Langley Mobile Ozone Lidar (LMOL) at NASA’s Langley Research Center (LaRC), and the Autonomous Mobile
55 Ozone Lidar Instrument for Tropospheric Experiments (AMOLITE) at Environment and Climate Change Canada.

56 All TOLNet lidars have unique configurations of original measurement design purposes, including their
57 transmitter, receiver, and signal processing systems. Most components of these lidars are customized and differ
58 significantly in pulse energy, repetition rate, receiver size, solar (or narrow-band) interference filter, and range
59 resolution. These differences result in varying signal-to-noise ratios (SNRs), which impact the useful operating
60 ranges and statistical uncertainties in ozone retrieval. The selection of the DIAL wavelengths determines the
61 sensitivity to interference by other species, primarily aerosols. In addition, multiple lidar data processing and
62 retrieval algorithms could also lead to different effective resolutions and lidar retrieval uncertainties (Godin et al.,
63 1999; Leblanc et al., 2016a,b). Therefore, it is important to quantify the measurement differences between the
64 TOLNet lidars and understand their sources before we can form a consistent TOLNet dataset. A previous
65 intercomparison between TROPOZ and LMOL reported by Sullivan et al. (2015) concluded that the observed ozone
66 column averages from the two lidars were within $\pm 8\%$ of each other, and their ozone profiles were mostly within
67 $\pm 10\%$ of each other.

68 **1.2 DISCOVER-AQ 2014 and FRAPPÉ Campaigns**

69 The scientific goal of the TOLNet lidars in this study was to provide continuous, high-resolution
70 tropospheric ozone profiles to support the NASA-sponsored DISCOVER-AQ mission
71 (<https://www.nasa.gov/larc/2014-discoveraq-campaign/>), and the National Science Foundation (NSF) and state of
72 Colorado (CO) jointly sponsored FRAPPÉ (Dingle et al., 2016) from July to August 2014. By collaborating with
73 FRAPPÉ, the 2014 CO study was the final stop in a series of four field campaigns by DISCOVER-AQ to understand
74 sources, transport and chemical transformations of air pollutants, particularly those that lead to ground-level ozone
75 formation (Crawford and Pickering, 2014).

76 Prior to the two campaigns, TOPAZ, TROPOZ, and LMOL were all deployed to the same location in Erie,
77 CO to obtain intercomparison data at the Boulder Atmospheric Observatory (BAO) (40.050°N , 105.003°W , 1584 m
78 above sea level, ASL). Subsequent to the BAO intercomparison, TROPOZ and LMOL re-deployed to locations near
79 Fort Collins, CO (~60 km north-northwest of BAO) and Golden, CO (~40 km southwest of BAO), respectively, for
80 their different scientific missions. During the DISCOVER-AQ and FRAPPÉ campaigns, balloon-borne ozonesondes
81 were launched at selected sites. In addition, the NASA P-3B aircraft performed multiple spiral ascents and descents
82 over several ground sites and provided measurements of ozone profiles. In this study, we compare retrievals
83 between the three lidars and evaluate the ozone lidar accuracy using ozonesonde and P-3B aircraft measurements.
84 These two campaigns offered a unique opportunity for the lidar validation work, as they involved so many different
85 instruments.

86 **2. Instruments**

87 **2.1 TOLNet Lidars**

88 Table 1 lists the main hardware specifications of the three TOLNet lidars and their ozone retrieval
89 processes, which could potentially impact the intercomparison result.

90 **2.1.1 TROPOZ/NASA GSFC**

91 The transmitter for TROPOZ consists of two 50-Hz Nd:YAG- lasers used to pump two Raman cells filled
92 with Deuterium (D₂) and Hydrogen (H₂) gases, respectively, to generate two outgoing pulses at 289 nm (on-line)
93 and 299 nm (off-line). The typical pulse energies are 12 mJ at 299 nm and 16 mJ at 289 nm (Sullivan et al., 2014).
94 The receiving system consists of a 45-cm-diameter Newtonian telescope for measuring far field and four smaller
95 2.5-cm refracting telescopes to measure near field. The 45-cm telescope has a 1-mrad field of view (FOV), and the
96 2.5-cm telescopes have a much wider FOV at 10 mrad. In each channel, solar interference filters with a 1-nm
97 bandwidth decrease the amount of ambient solar light, which improves the SNR. The fundamental range resolution
98 for the data acquisition system is 15 m (100 ns). TROPOZ measures ozone up to 16 km during daytime hours and
99 higher altitudes at night.

100 **2.1.2 TOPAZ/NOAA ESRL**

101 The TOPAZ lidar is a truck-mounted scanning instrument modified from the nadir-looking airborne DIAL
102 configuration first used in the 2006 Texas Air Quality Study (TexAQS II) (Alvarez et al., 2011; Senff et al., 2010).
103 The lidar transmitter is based on a Ce:LiCAF laser pumped by a quadrupled Nd:YLF laser to produce three UV
104 wavelengths, each at a 333 Hz repetition rate and tunable from 283 nm to 310 nm. The actual wavelengths used
105 during DISCOVER-AQ 2014 were 287, 291, and 294 nm. Compared to the conventional two-wavelength DIAL, the
106 three-wavelength configuration can potentially minimize the aerosol interference by using the dual-DIAL retrieval
107 technique (Kovalev and Bristow, 1996) without assuming a lidar ratio and Angström exponent. However, in this
108 study, ozone was retrieved using the 287- and 294-nm lidar signals and the standard two-wavelength DIAL
109 algorithm because the two-wavelength retrieval was less affected by significant lidar signal noise (Alvarez et al.,
110 2011).

111 Laser light backscattered by air molecules and aerosol particles is collected with a co-axial 50-cm diameter
112 Newtonian telescope and then split at a 1:9 ratio into near- and far-field detection channels. The FOVs of the near-
113 and far-field channels are controlled by different-size apertures resulting in full overlap at distances of ~300 m and
114 ~800 m, respectively. Both channels use gated photomultipliers (PMTs) operated in analog mode with solar
115 interference filters during the daytime. Compared to photon counting (PC) signals, the analog signal is able to
116 maintain high linearity for strong signals and is particularly suitable for near-range measurements. The two-axis
117 scanner on the truck sequentially points the laser beam at 2°, 6°, 20°, and 90° elevation angles in a cycle taking
118 approximately 5 minutes. The azimuth angle was fixed throughout the experiment. The ozone profiles at these four
119 angles are spliced together to create composite vertical profiles extending from 10 m to about 2 km AGL (Langford
120 et al., 2016). The range resolution of the signal recording system is 6 m.

121 During the 2014 DISCOVER-AQ and FRAPPÉ campaigns, the TOPAZ ozone observations suffered from
122 a slight, but consistent range-dependent bias created by an unknown source of noise in the data acquisition system.

123 The cause of this noise remains unknown and attempts to correct the resulting bias were unsuccessful. This bias
124 manifests itself primarily in the low elevation angle observations (2° , 6° , and 20°) because the signal levels and
125 SNRs are significantly lower compared to the measurements at 90° . For these reasons, the low angle observations
126 below 500 m were excluded from the comparisons reported within this study.

127 **2.1.3 LMOL/NASA LaRC**

128 The transmitter of LMOL consists of a diode-pumped Nd:YLF laser pumping a Ce:LiCAF tunable UV
129 laser to obtain two wavelengths typically at 287.1 and 292.7 nm with a pulse energy of 0.2 mJ at 500 Hz for each
130 wavelength. The lidar receiver system consists of a 40-cm telescope with a 1.4-mrad FOV to measure far field and
131 another 30-cm telescope with an adjustable FOV to measure near field (De Young et al., 2017). The raw lidar
132 signals are recorded with a 7.5-m range resolution. The LMOL data acquisition system operates in both analog and
133 PC modes. In this study, LMOL measures ozone between 0.7 and 4.5 km. Ozone measurements for DISCOVER-AQ
134 represent LMOL's very first remote deployment.

135 **2.1.4 Lidar Data Processing and Retrieval Algorithms**

136 The data processing and DIAL retrieval algorithms for the three TOLNet lidars are similar but not identical.
137 Their details have been described by Alvarez et al. (2011), De Young et al. (2017), Langford et al. (2011), and
138 Sullivan et al. (2015; 2014). Some basic procedures were applied on the raw lidar signals before retrievals, such as
139 time integration (5 min for this study), dead-time correction (for PC only), background correction (subtraction),
140 merging of PC and analog signals (for a system with both PC and analog channels), and signal-induced-bias (SIB)
141 correction (Kuang et al., 2013). Some parameters are system dependent or empirical due to different equipment,
142 such as the dead-time value, PC-analog timing offset, averaging range for background calculation, and SIB function
143 form. All groups agreed to use the Brion-Daumont-Malicet (BDM) (Daumont et al., 1992; Malicet et al., 1995;
144 Brion et al., 1993) ozone absorption cross-sections, which are temperature-dependent.

145 The ozone number density profile results from computing the derivative of the logarithm of the on-line to
146 off-line signal ratios. Spatial (range) smoothing is usually necessary to improve the SNR and reduce the statistical
147 errors. Various smoothing methods and their impacts on final lidar retrieval have been described by Godin et al.
148 (1999). Both TROPOZ and LMOL groups applied a Savitzky-Golay (SG) filter with a 2nd degree polynomial on the
149 derivative of the logarithm of the on-line to off-line signal ratios with an increasing window width to accommodate
150 the quickly decreasing SNR. However, the SG window sizes for TROPOZ and LMOL are different due to different
151 SNRs at each altitude. The TOPAZ group averaged lidar signal over 90 m and, then, smoothed the derivative of the
152 logarithm of the signal ratios with a five-point least-square fit in a 450-m window. The different retrieval
153 methodologies and parameters affect the effective vertical resolution of the retrieved ozone profiles [Leblanc et al.,
154 2016a], as listed in Table 1. This effective resolution determines the capability of the lidars to resolve vertical ozone
155 structure and is not equal to, but is associated with, the fitting window width.

156 All groups applied similar schemes to correct the aerosol interference. These schemes iteratively substitute
157 derived ozone from the DIAL equation into the lidar equation to solve aerosol extinction and backscatter until both

158 aerosol and ozone converge (Alvarez et al., 2011; Kuang et al., 2011; Sullivan et al., 2014). The differential aerosol
159 backscatter and extinction were calculated with the approximation from Browell et al. (1985). Lidars directly
160 measure the ozone number density, and all three groups used the same temperature and pressure profiles from co-
161 located ozonesonde measurements for Rayleigh correction, ozone mixing-ratio calculations, and computation of the
162 temperature dependent ozone absorption cross sections.

163 Merging between different altitude channels, either different telescopes or different optical channels of the
164 same telescope, is challenging with limited methodologies reported in the literature (Kuang et al., 2011). It is
165 difficult to specify a method for all groups because merging is system-dependent and is affected by many factors
166 previously described. Therefore, the three lidar groups merge the ozone profiles at different altitudes optimized for
167 their system and SNR levels such as the example method described by Sullivan et al. (2015). As a result, additional
168 differences between systems can occur due to the altitude channel merging.

169 **2.1.5 Error budget of the lidar measurements**

170 Only a brief description of the error budget of the lidar measurements is provided in this paper since the
171 details have been discussed in the respective instrument papers (Alvarez et al., 2011; De Young et al., 2017;
172 Sullivan et al., 2014). Table 2 presents the estimated daytime measurement uncertainties for 5 and 30-min
173 integration time for the three lidars. Statistical uncertainties (Papayannis et al., 1990) arising from signal fluctuations
174 are random errors and may be improved by additional averaging or smoothing. The statistical uncertainty, often
175 referred to as measurement precision, generally increases with range due to decreasing SNR and is different for the
176 three lidars due to their different laser power, telescope sizes, and measurement ranges. The uncertainty associated
177 with background correction also increases with range because of decreasing signal levels. The uncertainty due to the
178 saturation correction of the PC signals (Donovan et al., 1993) is also range dependent and typically maximizes at
179 near range. The uncertainty arising from aerosol interference could be the largest systematic error source and can be
180 minimized by using the appropriate correction algorithm (Eisele and Trickl, 2005; Immler, 2003; Sullivan et al.,
181 2014). The absorption by sulfur dioxide (SO₂) varies significantly with wavelength in the Hartley band. For the
182 TOPAZ and LMOL systems, the differential SO₂ absorption cross section (Rufus et al., 2003) is only about 1/8 of
183 their differential ozone absorption cross section so that the SO₂ interference is negligible unless very high ambient
184 SO₂ concentrations are present. For TROPOZ with the 289-299-nm pair, the differential absorption cross section of
185 SO₂ is about half of the ozone differential absorption cross section resulting in 1-ppb SO₂ being registered as 0.5-
186 ppb ozone. Under typical atmospheric condition when SO₂ concentrations are less than 2 ppb (Heikes et al., 1987)
187 and ozone concentrations are about 60 ppb, the SO₂-induced error is less than 2% (Sullivan et al., 2014). However,
188 SO₂ can cause a more significant ozone bias when high SO₂ concentrations are present such as in power plant or
189 volcanic plumes. The estimated total lidar measurement uncertainties [Leblanc et al., 2016b] for a 30-min signal
190 integration time are less than 20%, 12%, and 13% for TROPOZ, TOPAZ, and LMOL, respectively, within the lidar
191 measurement ranges listed in Table 1.

192 2.2 Ozonesondes

193 An ozonesonde is a lightweight, balloon-borne instrument that consists of an air pump and an ozone sensor
194 interfaced to a meteorological radiosonde. Ozonesondes are capable of measuring ozone under various weather
195 conditions (e.g., cloudy, thunderstorm). The ozone sensor uses an electrochemical concentration cell (ECC)
196 containing potassium iodide (KI) solution (Komhyr, 1969; Komhyr et al., 1995) to measure ozone with a precision
197 better than $\pm 5\%$ and an accuracy better than $\pm 10\%$ up to 35 km altitude with a sampling interval of about 1 s and a
198 retrieval vertical resolution of 100 m (Deshler et al., 2008; Johnson et al., 2008; Smit et al., 2007). A radiosonde
199 attached in the same package measures air temperature, pressure, and relative humidity (Stauffer et al., 2014). The
200 uncertainty of ozonesonde measurements is typically larger in the troposphere than that in the stratosphere (Liu et
201 al., 2009). It has been reported that the ECC sondes suffer interference from SO_2 (Flentje et al., 2010) with 1-ppb
202 SO_2 being registered as -1-ppb ozone (Schenkel and Broder, 1982). Elevated SO_2 can be a concern for lidar-
203 ozonesonde intercomparison for some lidar wavelengths (e.g., 289-299 nm) because of the opposite signs of the
204 measurement error arising from SO_2 for lidar and ozonesondes. However, this is not an issue for this study since we
205 did not find any noticeable interference from SO_2 in either lidar or ozonesonde data.

206 2.3 Ozone Measurement Instrument onboard NASA's P-3B

207 NASA's P-3B aircraft is a pressurized, four-engine turboprop, capable of long-duration flights of 8-12
208 hours and is based out of NASA's Wallops Flight Facility in Wallops Island, Virginia. A series of gas and aerosol
209 instruments were outfitted within the P-3B aircraft. Ozone was measured using the National Center for Atmospheric
210 Research (NCAR)'s 4-channel chemiluminescence instrument based on the reaction between ambient ozone and
211 nitric oxide (NO) with an accuracy of about $\pm 5\%$ and sampling interval of 1 s (Weinheimer et al., 1993; Ridley et
212 al., 1992). The precision of this ozone detector is better than $\pm 1\%$ when ambient ozone is higher than 10 ppbv. The
213 P-3B aircraft flew spirals from 300 m to 4570 m above the surface over selected ground monitoring sites including
214 all three lidar sites (more information in Section 3.3) during the DISCOVER-AQ 2014 campaign.

215 3. Results

216 3.1 Lidar Intercomparisons

217 The three TOLNet lidars were deployed next to the BAO tower to take simultaneous measurements before
218 the DISCOVER-AQ/FRAPPE campaign. They were only a few hundreds of meters away from each other and were
219 within 5 m of the same elevation (see measurement locations in Table 1).

220 Unlike stratospheric ozone lidars that focus on integrating hours of observations (Steinbrecht et al., 2009;
221 McDermid et al., 1990), tropospheric ozone lidars need to detect ozone variations with timescales on the order of
222 minutes, when considering ozone's shorter lifetime, smaller-scale transport, and mixing processes within the PBL
223 and free troposphere. Therefore, we processed all lidar data on a 5-min temporal scale (signal integration time).
224 Rayleigh correction was performed with the same atmospheric profile from the ozonesonde. Because the three lidars
225 have different fundamental range resolutions, retrieved ozone number density values were internally interpolated on
226 the same altitude grid with a 15-m interval for comparison.

227 Figure 1 presents the comparison of the TOPAZ and TROPOZ observed ozone at BAO from 1300 to 2135
228 UTC (local, Mountain Daylight Time, is UTC-6) on July 11, 2014 under a partly cloudy sky condition. Data
229 influenced by clouds were filtered out. Ozone time-height curtains from both lidars (Figure 1 a and b) show a
230 significant (about 40%) ozone increase in the early afternoon. A total of 7655 TOPAZ and TROPOZ coincident
231 pairs were constructed between 0.6 and 2 km AGL (altitude range over which both lidars provided valid data) over
232 this time period. The measurement differences between the two lidars are mostly within $\pm 5\%$ at individual grids
233 (Figure 1 c). The value of averaged ozone concentration over some specified altitude range can represent the
234 atmospheric ozone abundance and can be useful for satellite validation. Here, we refer to this value as ozone column
235 average with the unit of number density, not to be confused with integrated column ozone often reported in Dobson
236 units. The statistics of the intercomparison of the column averages is listed in Table 3. The similar 1σ standard
237 deviations (17.8 and $16.7 \times 10^{16} \text{ molec} \cdot \text{m}^{-3}$) suggest similar ozone variations captured by both lidars (also see Figure
238 1 a and b). The mean relative difference (or normalized bias) was calculated by averaging the relative difference
239 (i.e., $(\text{TROPOZ}-\text{TOPAZ})/\text{TOPAZ}$, the denominator was arbitrarily chosen) for all paired ozone profiles.
240 The $-1.1 \pm 2.6\%$ mean relative difference suggests excellent agreement of the averaged ozone column (Figure 1 d) for
241 80 profiles over 6.5 hours between TOPAZ and TROPOZ retrievals.

242 Figure 2 shows the TOPAZ-LMOL intercomparison for data taken on July 16, 2014 with 1902 coincident
243 pairs from 0.9 to 2 km and between 1340 to 1730 UTC on this day. Some of the data gaps were due to low clouds
244 blocking the lidar beams. The retrievals between the two lidars agree with each other mostly within $\pm 10\%$ (Figure 2
245 c). LMOL measured a mean ozone column average (Figure 2 d) $3.8 \pm 2.9\%$ lower than TOPAZ for a total of 28
246 paired profiles, which is significantly fewer than those from the TROPOZ-TOPAZ comparison. This small, but
247 statistically significant ozone column difference could be due to errors in the background and saturation corrections,
248 or biases introduced by the merging of signals or ozone retrievals from different instrument channels. Almost the
249 same 1σ of ozone column average in Table 3 suggests that the two lidars measured similar temporal ozone
250 variations. The $1-\sigma$ bars on the column average in Figure 2 (d) represent the vertical ozone variability captured by
251 lidar at a certain time. It can be seen that the two lidars measured highly similar vertical variability as well. The
252 consistency in capture of ozone variability for TOPA and LMOL is in part due to their similar statistical
253 uncertainties and vertical resolutions. The generally random distribution of the relative differences in Figure 1 (c)
254 and 2 (c) suggests overall consistent measurements with small systematic errors from all three lidars. In summary,
255 TROPOZ, LMOL, and TOPAZ report ozone values at individual altitudes mostly within $\pm 10\%$, which is well within
256 their respective uncertainties and report ozone column averages within $\pm 3.8\%$ on average.

257 3.2 Lidars versus Ozonesondes

258 In order to compare the lidar data to ozonesondes, the Rayleigh- and aerosol-corrected lidar data was
259 converted from ozone number densities to ozone mixing ratios by using sonde-measured pressure and temperature
260 profiles, and averaged over a 30-minute interval (± 15 minutes around sonde launch times). Ozonesondes and lidars
261 do not sample exactly the same atmospheric volume because the sondes typically drift horizontally. Therefore,
262 discrepancies between the lidar and sonde observations may be in part due to real atmospheric differences. The

263 horizontal displacement of the sonde usually increases with altitude, so the distance between sonde and lidar is
264 normally larger in the free troposphere than in the PBL. However, horizontal ozone gradients tend to be smaller in
265 the free troposphere than in the PBL, which typically keeps atmospheric differences rather small despite the
266 increased displacement of the sonde. The ozonesondes report values approximately every second (about every 5 m
267 in altitude) in raw data. For comparison, the ozonesonde raw data were linearly interpolated on the lidar altitude
268 grids with a 15-meter interval. Figure 3 shows the mean ozone mixing ratios measured by TOLNet lidars and
269 ozonesondes, as well as their mean relative difference as function of altitude.

270 After the DISCOVER-AQ/FRAPPÉ campaign started, the TROPOZ lidar deployed to Fort Collins, CO to
271 measure ozone. There were 11 ozonesonde profiles that were coincident and co-located with the TROPOZ
272 measurements. The mean ozone profiles of TROPOZ and sondes (Figure 3a) show similar vertical variations with
273 enhanced PBL and upper tropospheric ozone. The mean relative differences between TROPOZ and ozonesondes
274 (black line in Figure 3b) are mostly within $\pm 10\%$ up to 9 km. The local maximum of the differences at 1.8 km is
275 associated with the merging of ozone retrievals from the near-field channel and far-field channel. The green lines in
276 Figure 3 (b) represent the expected total measurement uncertainties including the lidar measurement uncertainties
277 for a 30-min integration time (also see Table 2) and a 10% constant uncertainty (accuracy) for ozonesondes. The
278 purple lines represent the $1-\sigma$ standard deviations of the mean differences, which can be compared to the combined
279 precision of lidar (i.e., statistical uncertainty) and ozonesonde (5%). The $1-\sigma$ standard deviation increases from about
280 10% in the lower troposphere to about 20% in the upper troposphere as a result of increasing lidar statistical
281 uncertainties with altitude. Below 9 km, the $1-\sigma$ standard deviations of the mean differences are mostly located
282 within the range of the expected uncertainties. In particular, the lidar-sonde differences around 0.5 km are
283 significantly less than the expected uncertainties suggesting that the detection and counting systems of TROPOZ
284 performed better than anticipated. Above 9 km, the biases increase and exceed 25% with large oscillations due to
285 large statistical errors as a consequence of low SNR. However, ozone observations with biases between 10-20% are
286 still representative of the upper free troposphere. On average, TROPOZ measures 2.9% higher ozone than the
287 ozonesondes for altitudes from 0.35 to 12 km. This difference can be seen as the mean difference of ozone column
288 average between the ozonesondes and lidar for a 30-min integration time.

289 Between July 10 and July 16, a total of 10 ozonesondes were released near the BAO tower and 7 of them
290 were coincident with TOPAZ measurements (3 on July 10, 3 on July 11, and 1 on July 16). TOPAZ mostly agrees
291 with ozonesondes between -5% and 10% (black line in Figure 3 d). The $1-\sigma$ standard deviation of the mean
292 differences (purple lines) is about 5% which is close to the combined precision of TOPAZ and ozonesondes (about
293 6%). $1-\sigma$ of the mean differences stays almost entirely within the expected uncertainties indicative of a proper
294 estimate of the lidar measurement uncertainties for TOPAZ in Table 2. Compared to ozonesondes, TOPAZ
295 measures 4.4% more PBL ozone on average.

296 On July 16, there was only one pair of coincident LMOL and ozonesonde measurements at the BAO tower
297 (Figure 3 e, f). The 30-minute averaged LMOL ozone profile agrees with the ozonesonde mostly within 0-15%
298 between 0.95 and 4.5 km AGL with an overall average of 6.2%. The maximum bias occurring at far range (above 4

299 km) is principally due to low SNR. The bias observed at 1.5 km is likely due to the high variation in aerosol
300 concentration and associated uncertainties in the aerosol correction. Since there is only one LMOL-ozonesonde
301 comparison, the statistical information on the overall bias between their measurements is not available.

302 In summary, all three TOLNet lidars measured higher ozone than ozonesondes with mean ozone column
303 differences of 2.9 % for TROPOZ, 4.4% for TOPAZ, and 6.2 % for LMOL (based on a single profile comparison). .
304 The differences between the two types of instruments and the standard deviations are mostly less than the expected
305 uncertainties. The largest bias occurs at far-range altitudes as expected and is primarily associated with the high
306 statistical errors arising from low SNR. The increased bias at near-range altitudes could be associated with various
307 factors, primarily the aerosol correction and the merging of the signals or ozone retrievals from different optical or
308 altitude channels.

309 **3.3 Lidars versus P-3B Chemiluminescence Instrument**

310 During the campaigns, the P-3B aircraft measured ozone profiles while doing spirals above the lidar sites.
311 There are 34 coincident profiles between TROPOZ and the P-3B at Fort Collins, 29 between TOPAZ and the P-3B
312 at the BAO tower, and 9 between LMOL and the P-3B at Golden, CO. The distances between the lidar and the P-3B
313 spiral centers for these paired profiles were less than 11 km. To make coincident pairs between P-3B and lidar data,
314 we interpolate the P-3B data onto the lidar vertical grids with a 15-m vertical resolution. Figure 4 shows the average
315 ozone profiles measured by the lidars and the P-3B as well as their mean relative differences. TROPOZ and the P-
316 3B agree with each other within $\pm 5\%$ between 0.5 to 3.5 km (black lines in Figure 4, b) with a -0.8% overall average
317 relative difference. The $1-\sigma$ standard deviation of the mean differences (purple lines in Figure 4 b) stays almost
318 entirely within the expected uncertainties (green lines) which include both calculated lidar measurement
319 uncertainties and a 5% constant uncertainty (accuracy) for the P-3B. TOPAZ agrees with the P-3B within -11% and
320 3% between 0.5 and 2 km (Figure 4 c, d) with a -2.7% overall average relative difference. TOPAZ underestimates
321 the lower-PBL (<1.5 km) ozone compared to P-3B, but when compared to ozonesondes TOPAZ overestimates
322 ozone at many of these same altitudes (see Figure 3 d). LMOL agrees with P-3B mostly within -5% and 0% above
323 1800 m and within -15% and -5% between 0.7-1.8 km (Figure 4 e, f) with a -4.9% overall average relative
324 difference. The $1-\sigma$ standard deviation of the LMOL-P3-B relative differences is mostly between 5% and 8% and is
325 close to their combined precision (6%). The $1-\sigma$ of the mean differences for both TOPAZ and LMOL (purple lines
326 in Figure 4 d, f) stays within the expected uncertainty (green lines) except for the bottom altitudes.

327 In summary, TOPAZ and LMOL exhibited noticeable negative bias in the PBL compared to the P-3B while
328 TROPOZ measured slightly lower than the P-3B. The differences between the three lidars and the P-3B are not
329 significantly correlated suggesting that these biases were not caused by the P-3B ozone instrument. These
330 differences could at least in part be caused by the lidar systematic errors mentioned in Section 2.1.5, but could also
331 reflect horizontal ozone variability across the P-3B spirals, which were up to 22 km in diameter

332 **4. Summary and Conclusions**

333 Intercomparisons have been made between three of the six TOLNet ozone lidars (NASA GSFC's
334 TROPOZ, NOAA ESRL's TOPAZ, and NASA LaRC's LMOL) and between these lidars and other *in situ* ozone
335 measurement instruments using coincident data during the 2014 DISCOVER-AQ and FRAPPÉ campaigns at
336 NOAA's BAO in Erie, CO. On average, TROPOZ, TOPAZ, and LMOL reported very similar ozone amounts within
337 their reported uncertainties for a 5-min signal integration time. The three lidars measured consistent ozone variations
338 revealed in the lidar time-height curtains and in the distribution of their relative differences. From intercomparisons
339 between the lidars and other instruments we find (1) All of the lidars measure higher ozone than ozonesondes with
340 an average relative difference within 4.4%. The lidar profile measurements agree with the ozonesonde observations
341 within -10-15% except at a few far-field altitudes. These results are generally consistent with Sullivan et al. (2015)
342 from a similar ozonesonde-lidar intercomparison. (2) TROPOZ agrees with the P-3B chemiluminescence instrument
343 below 3.5 km within $\pm 5\%$ with a small column-averaged relative difference of -0.8%. TOPAZ and LMOL exhibit a
344 slightly larger bias mostly between -15% and 5% below 2 km compared to the P-3B with a column-averaged
345 difference of -2.7% and -4.9%, respectively.

346 Comparisons between the three TOLNet lidars and with *in situ* instruments suggest that the lidars are
347 capable of capturing high-temporal tropospheric-ozone variability and of measuring tropospheric ozone with an
348 accuracy better than $\pm 15\%$ in terms of their vertical resolving capability and better than $\pm 5\%$ in terms of their
349 column measurement. These lidars have sufficient accuracy for model evaluation and satellite validation (Liu et al.,
350 2010). Since the 2014 campaigns, all of the TOLNET lidars have been modified to improve their stability and their
351 accuracy. The validation of these upgraded lidars will be reported in a future paper.

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359 position, policy, or decision.

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Table 1. Specifications for the TOLNet lidars.

	TROPOZ	TOPAZ	LMOL
Transmitter			
Laser type	Nd:YAG pumped D ₂ , H ₂ Raman cell	Nd:YLF pumped Ce:LiCAF	Nd:YLF pumped Ce:LiCAF
Wavelengths (nm)	288.9, 299.1	287, 291, 294	287.1, 292.7
Pulse Repetition Rate (Hz)	50	333	500
Pulse energy (mJ)	12 (299 nm), 16 (289 nm)	~0.06 for all wavelengths	0.2 for both wavelengths
Detection and data acquisition system			
Telescope diameter (cm)	45, 2.5	50	40, 30
FOV (mrad)	1 (45 cm), 10 (2.5 cm)	1.5 (far field channel), 3 (near field channel)	1.4 (far field channel), variable FOV (near field channel)
Signal detection type	PMT	PMT	PMT
Data acquisition type	PC	Analog	Analog and PC
Fundamental range resolution (m)	15	6	7.5
Instrument reference	(Sullivan et al., 2014)	(Alvarez et al., 2011)	(DeYoung et al., 2017)
DIAL retrieval			
DIAL retrieval and smoothing method	1 st -order (differential) SG filter with a 2 nd degree polynomial with an increasing window width applied on the derivative of the logarithm of the signal ratios	five-point least square fit with a 450-m window applied on the derivative of the logarithm of the signal ratios	1 st -order (differential) SG filter with a 2 nd degree polynomial, with an increasing window width applied on the derivative of the logarithm of the signal ratios
Retrieval effective resolution (m)	~100 at 1 km degrading to ~800 at 10 km	~10 below 50 m, ~30 from 50 to 150 m, ~100 from 150 to 500 m, 315 above 500 m	225 below 3 km degrading to 506 above 3 km
Aerosol correction reference	(Kuang et al., 2011; Sullivan et al., 2014)	(Alvarez et al., 2011)	(Browell et al., 1985; DeYoung et al., 2017)
Valid altitudes (km above ground level, AGL)	0.35-16	0.01-2	0.7-4.5
Measurement location			
Latitude (° N)	40.050	40.045	40.050
Longitude (° W)	105.000	105.006	105.004
Elevation (m ASL)	1584	1587	1584

364 **Table 2. Maximum 1- σ uncertainties for TROPOZ, TOPAZ and LMOL daytime ozone measurements within their**
 365 **measurable range for the 5 and 30-min integration time.**

Source	Maximum uncertainty within each lidar's measurement range					
	5-min integration			30-min integration		
Lidar	TROPOZ	TOPAZ	LMOL	TROPOZ	TOPAZ	LMOL
Measurement range (km)	0.35-16	0.01-2	0.7-4.5	0.35-16	0.01-2	0.7-4.5
Statistical Uncertainty ^a	20%	8%	15%	8%	3%	6%
Background correction ^a	10%	3%	5%	10%	3%	5%
Saturation correction ^b	1%	N/A	5%	1%	N/A	5%
Aerosol interference	10%	10%	10%	10%	10%	10%
Interference by SO ₂ , NO ₂ , O ₂ dimer	3%	1%	1%	3%	1%	1%
Differential Rayleigh scattering	3%	3%	3%	3%	3%	3%
Ozone absorption cross section	3%	3%	3%	3%	3%	3%
Total uncertainty ^c	25%	14%	19%	20%	12%	13%

366 ^a Range dependent and increasing with altitude.

367 ^b Range dependent and typically maximized at the near range.

368 ^c Total root-mean-square uncertainty by considering the range dependent uncertainties (also see Figure 3 and 4).

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Table 3. Comparisons of the ozone column average measured by TROPOZ, TOPAZ, and LMOL.

Date	UTC time range	Altitude range (km)	Lidar	Number of the paired profiles	Mean ozone column average (10 ¹⁶ molec·m ⁻³)	1 σ of the ozone column average (10 ¹⁶ molec·m ⁻³)	Mean relative difference*	1 σ of the difference
7/11/2014	1300-2135	0.6-2	TROPOZ/TOPAZ	80	127.3/128.6	17.8/16.7	-1.1%	2.6%
7/16/2014	1335-1730	0.9-2	LMOL/TOPAZ	28	98.1/102.0	13.1/13.0	-3.8%	2.9%

373 * Equal to mean (A-B)/B for A/B in 'Lidar' column for all paired profiles.

374

375 **Figure 1. Comparisons of ozone measured by TROPOZ and TOPAZ. (a) Ozone number densities measured by TROPOZ.**
376 **(b) Ozone number densities measured by TOPAZ. (c) Their relative percent differences, (TROPOZ-TOPAZ)/TOPAZ. (d)**
377 **Column averages measured by the TROPOZ and TOPAZ as well as their 1- σ standard deviations. TROPOZ measures**
378 **1.1 \pm 2.6% lower ozone column average than TOPAZ.**

379
380 **Figure 2. Comparisons of ozone measured by LMOL and TOPAZ. (a) LMOL-measured ozone number densities. (b)**
381 **TOPAZ-measured ozone number densities. (c) Their relative percent differences, (LMOL-TOPAZ)/TOPAZ. (d) Column**
382 **averages measured by LMOL and TOPAZ as well as their 1- σ standard deviations. LMOL measures 3.8 \pm 2.9% lower**
383 **ozone column average than TOPAZ.**

384
385 **Figure 3. Comparisons of lidar and ozonesonde measurements. (a) Average ozone profiles measured by TROPOZ and**
386 **ozonesondes at Fort Collins, CO (11 pairs). (b) Mean relative difference (black) between TROPOZ and ozonesondes as**
387 **well as the 1- σ standard deviations (purple). (c) Average ozone profiles measured by TOPAZ and ozonesondes at BAO**
388 **Tower (7 pairs). (d) Mean relative difference (black) between TOPAZ and ozonesondes as well as the 1- σ standard**
389 **deviations (purple). (e) Average ozone profiles measured by LMOL and ozonesonde at the BAO tower (1 pair). (f)**
390 **Relative difference between LMOL and ozonesonde. The gray lines represent the individual difference profiles between**
391 **the lidar and sondes. The green lines represent the expected uncertainties including the 30-min lidar measurement**
392 **uncertainties (also see Table 2) and a 10% constant uncertainty for ozonesondes.**

393
394 **Figure 4. Intercomparison between the lidar and P-3B measurements. (a) Average ozone profiles measured by TROPOZ**
395 **and P-3B at Fort Collins, CO (34 profiles). (b) Mean relative difference (black) between TROPOZ and P-3B data as well**
396 **as the 1- σ standard deviation (purple). (c) Average ozone profiles measured by TOPAZ and P-3B at the BAO Tower (29**
397 **profiles). (d) Mean relative difference between TOPAZ and P-3B data as well as the 1- σ standard deviation (purple). (e)**
398 **Average ozone profiles measured by LMOL and P-3B at Golden, CO (9 profiles). (f) Mean relative difference between**
399 **LMOL and P-3B data as well as the 1- σ standard deviation (purple). The gray lines represent the individual difference**
400 **profiles between the lidar and sondes. The green lines represent the expected uncertainties including the 30-min lidar**
401 **measurement uncertainties (also see Table 2) and a 10% constant uncertainty for ozonesondes.**

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