Response to Reviewers

We thank both reviewers for their significant time and constructive comments which improve the value of this paper. We have accepted Reviewers' suggestions and made following important changes 1) added profiles of estimated uncertainties for ozone lidar measurements in Figure 3 and 4 which could be compared to the 1-sigma of the differences between lidar and ozonesonde (or P3-B) and also added their corresponding description; 2) Added the uncertainty budget for each lidar system in Table 2 for a more rigorous characterization of measurement uncertainties.

Following are the detailed answers for Reviewers' specific questions. (The line numbers refer to the version with tracked changes.)

Reviewer 1

The manuscript titled "Quantifying TOLNet Ozone Lidar Accuracy during the 2014 DISCOVER-AQ FRAPPE Campaigns" intercompares 3 different ozone lidars in the field as well as compares the lidar measurements to in situ sonde and aircraft measurements. The authors do a good job explaining the need for the scientific experiments and discuss the results in a clear and concise manner. Very few minor revisions can be made and are discussed below:

1. **Line 159: How are the lidars selective for ozone as other compounds can absorb UV radiation at the wavelengths used here?**

In principle, the two wavelengths are selected at which the ozone absorptions are significantly different while the extinction for other gases and aerosols are small enough. The wavelengths of TOPAZ and LMOL are tunable and have been optimized for minimizing the differential scattering/absorption from other species (primarily SO_2). But, these two systems are relatively complicated and hard to maintain. The TROPOZ lasers are more straightforward and easier to maintain, but their wavelengths are fixed (289, 299nm). This fixed-wavelength pair 289-299 has larger interference from SO_2 than other two systems and we will explain the details in Question 3.

We have presented the error budget for all lidar systems in Section2.1 and Table 2. We have also stated that the corrections for differential Rayleigh scattering and aerosol interference have been regularly applied for all DIAL retrievals.

2. **Line 265: "...overall positive bias..." implies that the ozonesondes are without error.**

We change to say "…all three TOLNet lidars measured higher ozone than ozonesondes with mean ozone column differences of 2.9 % for TROPOZ, 4.4% for TOPAZ, and 6.2 % for LMOL (based on a single profile comparison)".

3. **It is known that SO2 can interfere with the electrochemical ozone measurement. Were the ozonesonde data corrected for this artifact in any way? Do you have any reason to believe that SO2 impacted the measurement (e.g. through proximity to a coal-fired power plant)?**

We have added the uncertainty discussion due to SO_2 for both ozonesondes and the lidars. There's no any sign for the lidar, sonde measurements which were contaminated by SO_2. This can be known by comparing TROPOZ data to TOPAZ and LMOL which have minimum SO_2 interference. There's unlikely high SO_2 emission around Boulder, CO at this time. But we still state this possibility as a generally potential error source in the ozonesonde instrument description as following in Section 2.2: "It has been reported that the ECC sondes suffer interference from SO_2 (Flentje et al., 2010) with 1-ppb SO_2 being

registered as -1-ppb ozone (Schenkel and Broder, 1982). Elevated SO_2 can be a concern for lidar-ozonesonde intercomparison for some lidar wavelengths (e.g., 289-299 nm) because of the opposite signs of the measurement error arising from SO_2 for lidar and ozonesondes. However, this is not an issue for this study since we did not find any noticeable interference from SO_2 in either lidar or ozonesonde data."

In terms of lidar measurement, the SO_2 absorption cross section in the Hartley band varies a lot and brings large uncertainty for the calculation. SO_2 is also a potential interfering specie for 289-299 pair and we have added more description about this error source in Section 2.1.5. Table 2 is modified as well.

4. **Section 3.2: When comparing the lidars with the P3, horizontal distances of up to 11 km were noted, yet horizontal differences were not discussed in this section. Since it is known the sondes do not travel directly upwards, differences between lidar and sondes could be due to real horizontal variability. Please discuss how this impacts the interpretation of your results.**

To address this question, we added following description in the 1st paragraph in Section 3.2: "Ozonesondes and lidars do not sample exactly the same atmospheric volume because the sondes typically drift horizontally. Therefore, discrepancies between the lidar and sonde observations may be in part due to real atmospheric differences. The horizontal displacement of the sonde usually increases with altitude, so the distance between sonde and lidar is normally larger in the free troposphere than in the PBL. However, horizontal ozone gradients tend to be smaller in the free troposphere than in the PBL, which typically keeps atmospheric differences rather small despite the increased displacement of the sonde."

Reviewer 2

The manuscript reports on the intercomparison of three tropospheric ozone lidars, ECC ozone sondes and an aircraft-based chemoluminescence ozone instrument (P3B) during two field campaigns in Colorado in summer 2014. The goal is to investigate the accuracy of the lidars, that is to discover potential systematic biases, and to estimate and check their precision. This topic is well suited for Atmospheric Measurement Techniques. A thorough published characterization of system performance and accuracy certainly increases the value of these systems for tropospheric ozone research and monitoring. While the manuscript presents substantial information about this intercomparison, I feel that the necessary subsequent scientific analysis and evaluation is still lacking. Such analysis would be needed to draw firmer conclusions about system precision and potential biases. As it stands now, the results are rather vague, more like a report. What is missing, to me, is a thorough scientific analysis of the presented material. Also missing are clearer messages on the resulting biases and uncertainties. The current 15% given in the abstract is rather wide and generic, hardly meriting a new paper. I feel that with the information inherent in the manuscript much tighter and more specific uncertainties could be given, especially when aerosol interference does not seem to play a large role. I recommend to address the following major points, before the manuscript can be accepted for publication:

To address these concerns, we have enhanced the analysis of the lidar measurement uncertainties for each system and added calculated lidar uncertainties in Figure 3 and 4 compared to the one sigma of the differences between the lidars and validation instruments. The expected measure precisions for all lidars are consistent with the measured precisions.

General Comments and Questions

1. **Figs. 1d, 2d, and 3c,d indicate that the TOPAZ system generally reports higher ozone. Where is this bias coming from? Is it significant? Does it have something to do with the signal recording / background subtraction? Why do these error sources not appear in Table 2?**

We think the measurement differences between TOPAZ and the other two lidar are acceptable although these differences are noticeable. TOPAZ measured higher than other lidars and sondes, but, measures lower than P3-B in average. This means TOPAZ does not persistently measure higher than other instruments. At L257 in Section 3.1, we explain the possible causes for the differences as: "This small, but statistically significant ozone column difference could be due to errors in the background and saturation corrections, or biases introduced by the merging of signals or ozone retrievals from different instrument channels."

2. **Fig. 4c-e, indicates a significant high bias of the P3B measurements. Given that TOPAZ (and possibly also LMOL, see Fig. 3e-f) seems to have a high bias against the sondes, the high bias of the P3B would be quite substantial. I think this possible bias needs to be investigated in more detail. It also needs to be reported in the abstract.**

After further investigation, we still think the P3B measurement is correct and has a measurement precision as claimed, 5%. The comparisons of TOPAZ-P3B and LMOL-P3B indeed don't look perfect in Figure 4. But, the biases are mostly within expected. We have added the expected total uncertainties in Figure 4, as the reviewer suggested, including the 5% P-3B uncertainty to account for the potential errors from P3-B.

In the last paragraph of Section 3, we provide the explanation for these biases as "The differences between the three lidars and the P-3B are not significantly correlated suggesting that these biases were not caused by the P-3B ozone instrument. These differences could at least in part be caused by the lidar systematic errors mentioned in Section 2.1.5, but could also reflect horizontal ozone variability across the P-3B spirals, which were up to 22 km in diameter."

3. **If significant, the potential biases in 1.) and 2.) need to be reported in the abstract. Or the authors have to clearly explain why they think these biases are not significant, and how they are covered by the different systems uncertainty budgets (e.g. in Table 1).**

We have enhanced the discussion of the error budget for each system in Table 2 and Section 2.1.5. We have adopted a more standard classification for error sources and added the uncertainties due to background correction and saturation correction. We have also added the expected uncertainties in Figure 3 and 4 to compare with the actual differences between lidar and sondes (or P3-B). The differences between different instruments generally smaller than expected uncertainties suggesting our understanding of these error sources are correct.

4. **Apart from potential biases, the authors also need to verify the precision estimates, e.g. those in Table 1. Since the statistical uncertainty (e.g. from photon counting statistics) is generally the largest uncertainty source for lidar ozone profile measurements, it is very important to calculate and report that properly. These estimated uncertainties then need to be checked using the statistics arising from multiple individual intercomparisons like the ones in this manuscript. This important check, to me, is missing completely here.**

We have accepted the suggestions and plotted the expected uncertainties (green lines in Figure 3 and 4) compare to the actual 1-sigma standard deviations. We have also provided more discussions on these changes. For example, we added in Section 3.2, "The green lines in Figure 3 (b) represent the expected

total measurement uncertainties including the lidar measurement uncertainties for a 30-min integration time (also see Table 2) and a 10% constant uncertainty for ozonesondes. The purple lines represent the 1-σ standard deviations of the mean differences, which can be compared to the combined precision of lidar (i.e., statistical uncertainty) and ozonesonde (5%). The 1-σ standard deviation increases from about 10% in the lower troposphere to about 20% in the upper troposphere as a result of increasing lidar statistical uncertainties with altitude. Below 9 km, the 1-σ standard deviations of the mean differences are mostly located within the range of the expected uncertainties. In particular, the lidar-sonde differences around 0.5 km are significantly less than the expected uncertainties suggesting that the detection and counting systems of TROPOZ performed better than anticipated."

5. **For example, the scatter / standard deviations from Figs. 3b,d , and 4b,d,f need to be compared to the estimated statistical uncertainty estimates available from the lidars. This probably requires additional plots. The information can then be used, on the one hand, to check the estimated lidar uncertainties, on the other hand to check the estimated sonde and P3B precisions. To me, such checks are a key component of an accuracy assessment. They are missing here.**
As mentioned above, we have added.

6. **Plots of average profile differences and their standard deviations should also be generated for the comparisons in Figs. 1 and 2. They also need to be included in the precision checks under 5., probably with additional plots and discussion.**
We believe we have provided enough discussions for the lidar comparisons including four plots in both Figure 1 and 2, and Table 3. These figures and discussions have covered comparisons for individual grids, column average, standard deviations. As mentioned earlier, we show the expected total uncertainty in both Figure 3 and 4 for all systems.

7. **In Figs. 4b,d,f, for example, it looks like the profile difference standard deviation is of the order of ±5 % (1σ). This would indicate that the precision (repeatability) of the lidar profiles is about 5%, assuming that the precision of the P3B profiles is 1% as stated in 2.3 ($\sqrt{5^2 + 1^2} \approx \sqrt{25} = 5$). The precision of the 30 minute lidar profiles would then be better than the 8% reported in Table 1 (and much better than the 13% reported in Table 1) <means Table 2>. Similar considerations apply to the standard deviations in Figs. 3b,d. Especially in Fig. 3b, the (expected) decrease of lidar precision with altitude seems quite apparent to me, and this should be checked against the lidar uncertainty estimates (e.g. from photon counting statistics).**

We agree with the reviewer's method to check the consistency between the actual bias and estimated uncertainties. We have broken down the numbers in Table 2 for separate systems. Table 2 reports the maximum uncertainties within each lidar's measurement range. The highest measurement altitude for TROPOZ is higher than 12 km. But the highest altitude shown in Figure 4(b) is only 4 km due to P3B's flying altitude. So the 1-sigmas of TROPOZ-P3B look smaller than "the maximum" in Table 2. We have added the discussions on the comparison of 1-sigma and lidar precision. The results show the actual precision is consistent with our estimates in Table 2.

For example, at L304 we add "The 1-σ standard deviation of the mean differences (purple lines) is about 5% which is close to the combined precision of TOPAZ and ozonesondes (about 6%). 1-σ of the mean differences stays almost entirely within the expected uncertainties indicative of a proper estimate of the lidar measurement uncertainties for TOPAZ in Table 2."

At L340, we add "The 1-σ standard deviation of the LMOL-P3-B relative differences is mostly between 5% and 8% and is consistent with their combined precision (6%). The 1-σ of the mean differences for both

TOPAZ and LMOL (purple lines in Figure 4 d, f) stays within the expected uncertainty (green lines) except for the bottom altitudes."

Detail comments:

1. **Line 1: Since the authors are only examining three of the many TOLNet lidars and certainly not TOLNet as a whole, I would suggest to move TOLNet after "2014" or after "FRAPPE".**

We agree that we are examining only three of the TOLNet lidars. But, because "TOLNet" is not the same category as DISCOVER-AQ or FRAPPE and rephrasing as the reviewer suggested may also cause confusion, we would still keep the current title.

2. **Line 25: Replace "cross-instrument calibration" by "the network". The authors are not calibrating the lidars (I hope), they should be self-calibrating. Network uniformity is what the authors are really looking for.**

Replaced.

3. **Line 32: Drop "In terms of range resolving capability". I found this confusing, because there is really no investigation of consequences of the different and altitude dependent vertical resolutions of the lidars in the manuscript. This would be a whole separate issue, and therefore I would just drop this here.**

Deleted

4. **Line 37: Replace "very good measurement accuracy for" by "that" and replace "making them" by "are". I am not sure that _15% are "very good".**

Changed as suggested.

5. **Line 44: Drop "high fidelity"? Is _15% high fidelity? I don't think so. P3B claims 1% if biases are corrected.**

Removed.

6. **Line 47: Swap "scientists" and "managers"? Or do the authors mean modeling and satellite managers?**

Changed to "scientists and managers within the air quality, modeling, and satellite communities"

7. **Lines 56, 57: Replace "that . . . their" by "of".**

Replaced.

8. **Line 59: Move "range resolution" after "operating ranges" in line 60. Range resolution is not really a hardware thing, and is much more determined by software.**

We agree the range resolution doesn't 100% belong to a hardware category because it could be adjusted when the software of the counting system is designed this way. But, the range resolution is associated with the capability of the counting system and so is an important parameter of hardware. So, we choose not to move "range resolution" here.

9. **Line 64: Add "can" before "form"?**
Added.

10. **Lines 67, 68: Drop "This particular study . . . United States"? Is this relevant? Are the authors sure it is true? Was there no comparison, e.g. between TROPOZ and the Table Mountain tropospheric ozone DIAL?**
This sentence has been removed.

11. **Line 82: "selected" instead of "selective". Don't see how some sites would be more selective than others.**
Changed as suggested.

12. **Line 82: Replace "profiles of ozone measurements" by "ozone profiles".**
Changed it to "measurements of ozone profiles".

13. **Line 93: Replace "lasers" by "pulses". Otherwise this would be a very expensive system indeed.**
Replaced.

14. **Line 102: Remove "zenith looking". As is now, this is confusing and contradictory.**
Changed.

15. **Line 117: Add "s" after "measurement".**
Added

16. **Lines 117-127: This is a lot of text to say that, in the end, the system was just pointed to zenith. Shorten.**
Shortened.

17. **Lines 144,145: Drop "database" and "to calculate differential".**
Dropped.

18. **Line 152, 153: I do not understand what is done here. 5 points at 6 m hardware resolution would be 30 meters. 450 meters at 6 m hardware resolution would be 75 points. Explain / correct, also in Table 1.**
Changed the sentence to "The TOPAZ group averaged lidar signal over 90 m and, then, smoothed the derivative of the logarithm of the signal ratios with a five-point least-square fitting in a 450-m interval."

19. **Line 155: Please cite Leblanc et al. 2016 here. The authors should also include the other Leblanc et al. 2016 paper(s) on ozone profile uncertainties in the references. Also, the results here, i.e. range**

resolutions, ozone uncertainties and accuracies need to be properly put into the context of these papers, here and in other places in the text.

We have added [Leblanc et al., 2016b] and cited papers by Leblanc [2016a, b] at L63, 154, 188. We have brought these values into the discussions, especially the uncertainties.

20. Line 168: Remove "non-standardized". Because it is so system specific, standardization is not really a criterion/ issue.

Removed.

21. Line 174: Remove "maximum". What would that mean?

To avoid confusion, we rephrase the sentence as "The statistical uncertainty, often referred to as measurement precision, generally increases with range due to decreasing SNR and is different for the three lidars due to their different laser power, telescope sizes, and measurement ranges."

22. Line 179: Leblanc et al. 2016 on ozone profile uncertainties should be cited here, and should be put in context. Somewhere the authors should also mention that lidar uncertainty increases a lot with increasing altitude/ range.

The Leblanc et al., 2016b paper has been cited here. Yes, we have said so at L179 in the error budget section and other places, and also stated the statistical uncertainty was range dependent in the footnote of Table 2.

23. Lines 181 to 191: To me, this is a bit backward. First the authors give the principle, then end results, and then the authors go back to the principle again. Rewrite / reorder.

As suggested, we have deleted the repeating words and reordered this paragraph.

24. Lines 108/109: These two citations should probably come before, on line 106 right after "observations". <Means line number "208 and 209">

We agree and have moved them as suggested.

25. Line 115: Replace "cloud interferences" by "clouds".

Replaced.

26. Line 223: Similar standard deviation could also be due to similar noise / precision /uncertainty. In fact, I think this seems to be the case from 13 to 17 UT, whereas similar variations seem to captured from 18 to 22 UT. Please reword.

We agree the sigma could come from uncertainty, primarily statistical uncertainty which is a random noise. However, the ozone variations in Figure 1 (a) and (b) don't look like random noises. So, we add "(also see Figure 1 a and b)".

27. Line 227: As mentioned above, please also show the mean and standard deviation profiles of these ozone differences. Same for Fig. 2.

Please see the answer for General comment 6.

28. Lines 265 to 271: I think this needs to be thought through much better. Are the sondes too low? Why would averaging time affect a bias? It should only affect the noise / significance. Same goes

for SNR. Biases that are not resolvable/ not significant / within the uncertainty margins should not be discussed at all.

We have deleted the citation of the Gaudel et al. 2015 paper because it is not comparable to this study. The Gaudel paper compares seasonally averaged lidar and sonde O3 profiles that were not necessarily taken at the same time. Then, we modify this paragraph as "In summary, all three TOLNet lidars measured higher ozone than ozonesondes with mean ozone column differences of 2.9 % for TROPOZ, 4.4% for TOPAZ, and 6.2 % for LMOL (based on a single profile comparison). . The differences between the two types of instruments and the standard deviations are mostly less than the expected uncertainties. The largest biase occurs at far-range altitudes as expected and is primarily associated with the high statistical errors arising from low SNR. The increased bias at near-range altitudes could be associated with various factors, primarily the aerosol correction and the merging of the signals or ozone retrievals from different optical or altitude channels."

29. Lines 286 to 287: Why not the P3B? Figs. 4d and 4f look very similar. Many things point towards P3B being high. Same as the discussion of lidar sonde differences this discussion is to short. A lot more needs to be done / said here. See my major comments.

If P3B has a significant measurement bias, we expect all three lidars to have similar measurement differences relative to P3-B. But now, only two of them look similar. So, we don't think P3-B had significant measurement issue.

30. Table 2: Are these uncertainties 1_ or 2_?

Added 1σ in the title of Table 2.

31. Figs. 1d and 2d. Please plot (some/typical) error bars for these time series.

Added the error bars and their discussions.

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

Lihua Wang[1], Michael J. Newchurch[1], Raul J. Alvarez II[2], Timothy A. Berkoff[3], Steven S. Brown[2], William Carrion[3,4], Russell J. De Young[3], Bryan J. Johnson[2], Rene Ganoe[4], Guillaume Gronoff[3,4], Guillaume Kirgis[2,5], Shi Kuang[1], Andrew O. Langford[2], Thierry Leblanc[6], Erin E. McDuffie[2,5,7], Thomas J. McGee[8], Denis Pliutau[4], Christoph J. Senff[2,5], John T. Sullivan[8,9], Grant Sumnicht[4], Laurence W. Twigg[4], Andrew J. Weinheimer[10]

[1]University of Alabama in Huntsville, Huntsville, Alabama, USA

[2]NOAA Earth System Research Laboratory, Boulder, Colorado, USA

[3]NASA Langley Research Center, Hampton, Virginia, USA

[4]Science Systems and Applications Inc., Lanham, Maryland, USA

[5]Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, Colorado, USA

[6]Jet Propulsion Laboratory, California Institute of Technology, Wrightwood, California, USA

[7]Department of Chemistry, University of Colorado, Boulder, Colorado, USA

[8]NASA Goddard Space Flight Center, Greenbelt, Maryland, USA

[9]Joint Center for Earth Systems Technology, Baltimore, Maryland, USA

[10]National Center for Atmospheric Research, Boulder, USA

Correspondence to Shi Kuang (kuang@nsstc.uah.edu)

Abstract

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

1.   Introduction

1.1 TOLNet

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

TOLNet consists of five ozone lidars across the United States and one in Canada: the Table Mountain tropospheric ozone differential absorption lidar (DIAL) at NASA's Jet Propulsion Laboratory, the Tunable Optical Profiler for Aerosol and oZone (TOPAZ) lidar at NOAA's Earth System Research Laboratory (ESRL), the Rocket-city Ozone (O_3) Quality Evaluation in the Troposphere (RO_3QET) lidar at the University of Alabama in Huntsville (UAH), the TROPospheric OZone (TROPOZ) DIAL at NASA's Goddard Space Flight Space Center (GSFC), the Langley Mobile Ozone Lidar (LMOL) at NASA's Langley Research Center (LaRC), and the Autonomous Mobile Ozone Lidar Instrument for Tropospheric Experiments (AMOLITE) at Environment and Climate Change Canada.

2

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

1.2 DISCOVER-AQ 2014 and FRAPPÉ Campaigns

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

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

2.   Instruments

2.1 TOLNet Lidars

90   Table 1 lists the main hardware specifications of the three TOLNet lidars and their ozone retrieval

91   processes, which could potentially impact the intercomparison result.

2.1.1 TROPOZ/NASA GSFC

93   The transmitter for TROPOZ consists of two 50-Hz Nd:YAG- lasers used to pump two Raman cells filled

94   with Deuterium (D_2) and Hydrogen (H_2) gases, respectively, to generate two outgoing ~~lasers~~ pulses at 289 and

95   299 -nm. The typical pulse energies are 12 mJ at 299 nm (off-line) and 16 mJ at 289 nm (on-line) (Sullivan et al.,

96   2014). The receiving system consists of a 45-cm-diameter Newtonian telescope for measuring far field and four

97   smaller 2.5-cm refracting telescopes to measure near field. The 45-cm telescope has a 1-mrad field of view (FOV),

98   and the 2.5-cm telescopes have a much wider FOV at 10 mrad. In each channel, solar interference filters with a 1-

99   nm bandwidth decrease the amount of ambient solar light, which improves the SNR. The fundamental range

100   resolution for the data acquisition system is 15 m (100 ns). TROPOZ measures ozone up to 16 km during daytime

101   hours and higher altitudes at night.

2.1.2 TOPAZ/NOAA ESRL

103   The TOPAZ lidar is a truck-mounted ~~zenith looking,~~ scanning instrument modified from the nadir-looking

104   airborne DIAL configuration first used in the 2006 Texas Air Quality Study (TexAQS II) (Alvarez et al., 2011;

105   Senff et al., 2010). The lidar transmitter is based on a Ce:LiCAF laser pumped by a quadrupled Nd:YLF laser to

106   produce three UV wavelengths, each at a 333 Hz repetition rate and tunable from 283 nm to 310 nm. The actual

107   wavelengths used during DISCOVER-AQ 2014 were 287, 291, and 294 nm. Compared to the conventional two-

108   wavelength DIAL, the three-wavelength configuration can potentially minimize the aerosol interference by using the

109   dual-DIAL retrieval technique (Kovalev and Bristow, 1996) without assuming a lidar ratio and Angström exponent.

110   However, in this study, ozone was retrieved using the 287- and 294-nm lidar signals and the standard two-

111   wavelength DIAL algorithm because the two-wavelength retrieval was less affected by significant lidar signal noise

112   (Alvarez et al., 2011).

113   Laser light backscattered by air molecules and aerosol particles is collected with a co-axial 50-cm diameter

114   Newtonian telescope and then split at a 1:9 ratio into near- and far-field detection channels. The FOVs of the near-

115   and far-field channels are controlled by different-size apertures resulting in full overlap at distances of ~300 m and

116   ~800 m, respectively. Both channels use gated photomultipliers (PMTs) operated in analog mode with solar

117   interference filters during the daytime. Compared to photon counting (PC) signals, the analog signal is able to ~~keep~~

118   maintain high linearity for strong signals and is particularly suitable for near-range measurements. The two-axis

119   scanner on the truck sequentially ~~permits pointing~~points the laser beam at ~~several shallow elevation angles at a~~

120   ~~fixed, but changeable azimuth angle, typically at~~ 2°, 6°, 20°, and 90° elevation angles in a cycle taking~~that are~~

121   ~~repeated~~ approximately ~~every~~ 5 minutes. The azimuth angle was fixed throughout the experiment. The ozone

122   profiles at these four angles are spliced together to create composite vertical profiles extending from 10 m to about 2

123   km AGL (Langford et al., 2016). The range resolution of the signal recording system is 6 m.

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124   During the 2014 DISCOVER-AQ and FRAPPÉ campaigns, the TOPAZ ozone observations ~~at low~~

125   ~~elevation angles (2°, 6°, and 20°)~~ suffered from a slight, but consistent range-dependent bias created by an unknown

126   source of noise in the data acquisition system. The cause of this noise remains unknown and attempts to correct the

127   resulting bias were unsuccessful. This bias manifests itself primarily in the low elevation -angle observations (2°, 6°,

128   and 20°) because the signal levels and SNR are significantly lower compared to the measurements at 90°°. For these

129   reasons, the low angle observations below 500 -m were excluded from the comparisons reported within this study.

2.1.3 LMOL/NASA LaRC

130

131   The transmitter of LMOL consists of a diode-pumped Nd:YLF laser pumping a Ce:LiCAF tunable UV

132   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

133   wavelength. The lidar receiver system consists of a 40-cm telescope with a 1.4-mrad FOV to measure far field and

134   another 30-cm telescope with an adjustable FOV to measure near field (De Young et al., 2017). The raw lidar

135   signals are recorded with a 7.5-m range resolution. The LMOL data acquisition system operates in both analog and

136   PC modes. In this study, LMOL measures ozone between 0.7 and 4.5 km. Ozone measurements for DISCOVER-AQ

137   represent LMOL's very first remote deployment.

2.1.4 Lidar Data Processing and Retrieval Algorithms

138

139   The data processing and DIAL retrieval algorithms for the three TOLNet lidars are similar but not identical.

140   Their details have been described by Alvarez et al. (2011), De Young et al. (2017), Langford et al. (2011), and

141   Sullivan et al. (2015; 2014). Some basic procedures were applied on the raw lidar signals before retrievals, such as

142   time integration (5 min for this study), dead-time correction (for PC only), background correction (subtraction),

143   merging of PC and analog signals (for a system with both PC and analog channels), and signal-induced-bias (SIB)

144   correction (Kuang et al., 2013). Some parameters are system dependent or empirical due to different equipment,

145   such as the dead-time value, PC-analog timing offset, averaging range for background calculation, and SIB

146   ~~simulation~~ function form. All groups agreed to use the Brion-Daumont-Malicet (BDM) ~~database~~ (Daumont et al.,

147   1992; Malicet et al., 1995; Brion et al., 1993) ~~to calculate differential~~ ozone absorption cross-sections, which are

148   temperature-dependent.

149   The ozone number density profile results from computing the derivative of the logarithm of the on-line to

150   off-line signal ratios. Spatial (range) smoothing is usually necessary to improve the SNR and reduce the statistical

151   errors. Various smoothing methods and their impacts on final lidar retrieval have been described by Godin et al.

152   (1999). Both TROPOZ and LMOL groups applied a Savitzky-Golay (SG) filter with a 2nd degree polynomial on the

153   derivative of the logarithm of the on-line to off-line signal ratios with an increasing window width to accommodate

154   the quickly decreasing SNR. However, the SG window sizes for TROPOZ and LMOL are different due to different

155   SNRs at each altitude. The TOPAZ group averaged lidar signal over 90 m and, then, smoothed the derivative of the

156   logarithm of the signal ratios with a five-point least-square fit~~ting~~ in a 450-m ~~interval~~window. The different retrieval

157   methodologies and parameters affect the effective vertical resolution of the retrieved ozone profiles [Leblanc et al.,

158   2016a], as listed in Table 1. This effective resolution determines the capability of the lidars to resolve vertical ozone

159   structure and is not equal to, but is associated with, the fitting window width.

All groups applied similar schemes to correct the aerosol interference. These schemes iteratively substitute derived ozone from the DIAL equation into the lidar equation to solve aerosol extinction and backscatter until both aerosol and ozone converge (Alvarez et al., 2011; Kuang et al., 2011; Sullivan et al., 2014). The differential aerosol backscatter and extinction were calculated with the approximation from Browell et al. (1985). Lidars directly measure the ozone number density, and all three groups used the same temperature and pressure profiles from co-located ozonesonde measurements for Rayleigh correction, ozone mixing-ratio calculations, and computation of the temperature dependent ozone absorption cross sections.

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

2.1.5 Error budget of the lidar measurements

Only a brief description of the error budget of the lidar measurements is provided in this paper since the details have been discussed in the respective instrument paper~~s~~ (Alvarez et al., 2011; De Young et al., 2017; Sullivan et al., 2014). Table 2 presents the estimated daytime measurement uncertainties for 5 and~~or~~ 30-min integration time for the three lidars. Statistical ~~errors~~ uncertainties (Papayannis et al., 1990) arising from signal ~~and background noise~~ fluctuations are random errors and may be improved by additional averaging or smoothing. The ~~maximum~~ statistical uncertaint~~y~~, often referred to as measurement precision, generally increases with range due to decreasing SNR and is different ~~ies~~ for the three lidars ~~are similar (20% for 5 min and 8% for 30 min) within~~ due to their different laser power, telescope sizes, and measur~~ement~~able ranges. ~~although they are different at the same altitude.~~ The uncertainty associated with background correction also increases with range because of decreasing signal levels. The uncertainty due to the saturation correction of the PC signals (Donovan et al., 1993) is also range dependent and typically maximizes at near range. The uncertainty arising from aerosol interference could be the largest systematic error source and can be minimized by using the appropriate correction algorithm (Eisele and Trickl, 2005; Immler, 2003; Sullivan et al., 2014). The absorption by sulfur dioxide (SO_2) varies significantly with wavelength in the Hartley band. For the TOPAZ and LMOL systems, the differential SO_2 absorption cross section (Rufus et al., 2003) is only about 1/8 of their differential ozone absorption cross section so that the SO_2 interference is negligible unless very high ambient SO_2 concentrations are present. For TROPOZ with the 289-299-nm pair, the differential absorption cross section of SO_2 is about half of the ozone differential absorption cross section resulting in 1-ppb SO_2 being registered as 0.5-ppb ozone. Under typical atmospheric condition when SO_2 concentrations are less than 2 ppb (Heikes et al., 1987) and ozone concentrations are about 60 ppb, the SO_2-induced error is less than 2% (Sullivan et al., 2014). However, SO_2 can cause a more significant ozone bias when high SO_2 concentrations are present such as in power plant or volcanic plumes. The estimated total lidar measurement uncertainties [Leblanc et

195 al., 2016b] for a 30-min signal integration time are less than 20~~2~~%, 12%, and 13% for ~~5 and 30 min.~~TROPOZ,
196 TOPAZ, and LMOL, respectively, within the lidar measurement ranges listed in Table 1.

2.2 Ozonesondes

198   An ozonesonde is a lightweight, balloon-borne instrument that consists of a~~n~~ ~~Teflon~~ air pump and an ozone
199 sensor interfaced to a meteorological radiosonde. Ozonesondes are capable of measuring ozone under various
200 weather conditions (e.g., cloudy, thunderstorm). The ozone sensor uses an ~~electrode~~ electrochemical concentration
201 cell (ECC) containing potassium iodide (KI) solution (Komhyr, 1969; Komhyr et al., 1995) to measure ozone with a
202 precision better than ±5% and an accuracy better than ±10% up to 35 km altitude with a sampling interval of about 1
203 s and a retrieval vertical resolution of 100 m (Deshler et al., 2008; Johnson et al., 2008; Smit et al., 2007). A
204 radiosonde attached in the same package measures air temperature, pressure, and relative humidity (Stauffer et al.,
205 2014). The uncertainty of ozonesonde measurement~~s~~ ~~is~~ is typically larger in the troposphere than that in the
206 stratosphere (Liu et al., 2009). It has been reported that the ECC sondes suffer interference from SO_2 (Flentje et al.,
207 2010) with 1-ppb SO_2 being registered as -1-ppb ozone (Schenkel and Broder, 1982). Elevated SO_2 can be a concern
208 for lidar-ozonesonde intercomparison for some lidar wavelengths (e.g., 289-299 nm) because of the opposite signs
209 of the measurement error arising from SO_2 for lidar and ozonesondes. However, this is not an issue for this study
210 since we did not find any noticeable interference from SO_2 in either lidar or ozonesonde data. ~~As the balloon~~
211 ~~carrying the instrument package ascends through the atmosphere, the pump bubbles ambient air into the sensor cell.~~
212 ~~The reaction of ozone and iodide generates an electrical signal proportional to the amount of ozone. A radiosonde~~
213 ~~attached in the same package measures air temperature, pressure, and relative humidity (Stauffer et al., 2014).~~
214 ~~Ozonesondes are capable of measuring ozone under various weather conditions (e.g., cloudy, thunderstorm).~~ ~~The~~
215 ~~free-flying ozonesondes typically reach 35-km altitude in less than two hours with a rise rate at about 5 m/s.~~

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

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

3.   Results

3.1   Lidar Intercomparisons

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

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

237   Figure 1 presents the comparison of the TOPAZ and TROPOZ observed ozone at BAO from 1300 to 2135
238 UTC (6 hours ahead of local time, Mountain Daylight Time, is UTC-6) on July 11, 2014 under a partly cloudy sky
239 condition. Data influenced by clouds interferences were filtered out. Ozone time-height curtains from both lidars
240 (Figure 1 a and b) show a significant (about 40%) ozone increase in the early afternoon. A total of 7655 TOPAZ and
241 TROPOZ coincident pairs were constructed between 0.6 and 2 km AGL (altitude range over which both lidars
242 provided valid data) over this time period. The measurement differences between the two lidars are mostly within
243 ±5% at individual grids (Figure 1 c). The product value of averaged ozone concentration over some specified
244 altitude range can represent the atmospheric ozone abundance and can be also useful for satellite validation. Here,
245 we refer to this value product as ozone column average with the unit of number density, not to be confused with
246 integrated column ozone often reported in Dobson units. The statistics of the intercomparison of the column
247 averages is listed in Table 3. The similar 1σ standard deviations (17.8 and 16.7 x 10^{16} molec·m^{-3}) suggest similar
248 ozone variations captured by both lidars (also see Figure 1 a and b). The mean relative difference (or normalized
249 bias) was calculated by averaging the relative difference (i.e., (TROPOZ-TOPAZ)/TOPAZ, the denominator was
250 arbitrarily chosen) for all paired ozone profiles. The -1.1±2.6% mean relative difference suggests excellent
251 agreement of the averaged ozone column (Figure 1 d) for 80 profiles over 6.5 hours between TOPAZ and TROPOZ
252 retrievals.

253   Figure 2 shows the TOPAZ-LMOL intercomparison for data taken on July 16, 2014 with 1902 coincident
254 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
255 blocking the lidar beams. The retrievals between the two lidars agree with each other mostly within ±10% (Figure 2
256 c). LMOL measured a mean ozone column average (Figure 2 d) 3.8±2.9% lower than TOPAZ for a total of 28
257 paired profiles, which is significantly fewer than those from the TROPOZ-TOPAZ comparison. This small, but
258 statistically significant ozone column difference could be due to errors in the background and saturation corrections,
259 or biases introduced by the merging of signals or ozone retrievals from different instrument channels. Almost the
260 same 1σ of ozone column average in Table 3 suggests that the two lidars measured similar temporal ozone
261 variations. The $1-\sigma$ bars on the column average in Figure 2 (d) represent the vertical ozone variability captured by
262 lidar at a certain time. It can be seen that the two lidars measured highly similar vertical variability as well. The
263 consistency in capture of ozone variability for TOPA and LMOL is in part due to their similar statistical
264 uncertainties and vertical resolutions.

265 The generally random distribution of the relative differences in Figure 1 (c) and 2 (c) suggests overall

266 consistent measurements with small systematic errors from all three lidars. In summary, TROPOZ, LMOL, and

267 TOPAZ report ozone values at individual altitudes mostly within ±10%, which is well within their respective

268 uncertainties and report ozone column averages within ±3.8% on average.

269 **3.2   Lidars versus Ozonesondes**

270 In order to compare the lidar data to ozonesondes, the Rayleigh- and aerosol-corrected lidar data was

271 converted from ozone number densities to ozone mixing ratios by using sonde-measured pressure and temperature

272 profiles, and averaged over a 30-minute interval (±15 minutes around sonde launch times). − Ozonesondes and

273 lidars do not sample exactly the same atmospheric volume because the sondes typically drift horizontally. Therefore,

274 discrepancies between the lidar and sonde observations may be in part due to real atmospheric differences. The

275 horizontal displacement of the sonde usually increases with altitude, so the distance between sonde and lidar is

276 normally larger in the free troposphere than in the PBL. However, horizontal ozone gradients tend to be smaller in

277 the free troposphere than in the PBL, which typically keeps atmospheric differences rather small despite the

278 increased displacement of the sonde. The ozonesondes report values approximately every second (about every 5 m

279 in altitude) in raw data. For comparison, the ozonesonde raw data were linearly interpolated on the lidar altitude

280 grids with a 15-meter interval. Figure 3 shows the mean ozone mixing ratios measured by TOLNet lidars and

281 ozonesondes, as well as their mean relative difference as function of altitude.

282 After the DISCOVER-AQ/FRAPPÉ campaign started, the TROPOZ lidar deployed to Fort Collins, CO to

283 measure ozone. There were 11 ozonesonde profiles that were coincident and co-located with the TROPOZ

284 measurements. The mean ozone profiles of TROPOZ and sondes (Figure 3a) show similar vertical variations with

285 enhanced PBL and upper tropospheric ozone. The mean relative differences between TROPOZ and ozonesondes

286 (black line in Figure 3b) are mostly within ±10% up to 9 km. The local maximum of the differences at 1.8 km is

287 associated with the merging of ozone retrievals from the near-field channel and far-field channel. The green lines in

288 Figure 3 (b) represent the expected total measurement uncertainties including the lidar measurement uncertainties

289 for a 30-min integration time (also see Table 2) and a 10% constant uncertainty for ozonesondes. The purple lines

290 represent the 1-σ standard deviations of the mean differences, which can be compared to the combined precision of

291 lidar (i.e., statistical uncertainty) and ozonesonde (5%). The 1-σ standard deviation increases from about 10% in the

292 lower troposphere to about 20% in the upper troposphere as a result of increasing lidar statistical uncertainties with

293 altitude. Below 9 km, the 1-σ standard deviations of the mean differences are mostly located within the range of the

294 expected uncertainties. In particular, the lidar-sonde differences around 0.5 km are significantly less than the

295 expected uncertainties suggesting that the detection and counting systems of TROPOZ performed better than

296 anticipated. Above 9 km, the biases start to increase and exceed 25% with large oscillations due to large statistical

297 errors as a consequence of low SNR. However, ozone observations with Bbiases between 10-20% are still very

298 representative of the upper free troposphere. On average, for altitudes from 0.35 to 12 km, TROPOZ measures 2.9%

299 higher ozone than the ozonesondes for altitudes from 0.35 to 12 km. This difference can be seen as the mean

300 difference of ozone column average between the ozonesondes and lidar for a 30-min integration time.

9

301   Between July 10 and July 16, a total of 10 ozonesondes were released near the BAO tower and 7 of them
302   were coincident with TOPAZ measurements (3 on July 10, 3 on July 11, and 1 on July 16). TOPAZ mostly agrees
303   with ozonesondes between -5% and 10% (black line in Figure 3 ~~c,~~ d). The 1-σ standard deviation of the mean
304   differences (purple lines) is about 5% which is close to the combined precision of TOPAZ and ozonesondes (about
305   6%). 1-σ of the mean differences stays almost entirely within the expected uncertainties indicative of a proper
306   estimate of the lidar measurement uncertainties for TOPAZ in Table 2. Compared to ozonesondes, TOPAZ
307   measures 4.4% more PBL ozone on average.

308   On July 16, there was only one pair of coincident LMOL and ozonesonde measurements at the BAO tower
309   (Figure 3 e, f). The 30-minute averaged LMOL ozone profile agrees with the ozonesonde mostly within 0-15%
310   between 0.95 and 4.5 km AGL with an overall average of 6.2%. The maximum bias occurring at far range (above 4
311   km) is principally due to low SNR. The bias observed at 1.5 km is likely due to the high variation in aerosol
312   concentration and associated uncertainties in the aerosol correction, ~~that was also observed in the green channel.~~
313   Since there is only one LMOL-ozonesonde comparison ~~between the LMOL and ozonesonde~~, the statistical
314   information on the overall bias between their measurements is not available.

315   In summary, all three TOLNet lidars ~~exhibit overall positive bias~~measured higher ozone , ~~up to 4.4%,~~ [Formatted: Justified]
316   ~~compared to~~than ozonesondes with mean ozone column differences of 2.9 % for TROPOZ, 4.4% for TOPAZ, and
317   6.2 % for LMOL (based on a single profile comparison). ~~excluding the single profile comparison to LMOL (6.2%).~~
318   ~~The larger bias than the~~ The differences between the two types of instruments and the standard deviations are mostly
319   less than the expected uncertainties. ~~climatological difference between lidar and ozonesondes reported by Gaudel et~~
320   ~~al. (2015) (0.6 ppbv) could be associated with the much shorter averaging time period.~~ The ~~maximum~~ largest bias~~es~~
321   ~~exist~~ occurs at ~~in two regions, near range altitudes and~~ far-range altitudes. ~~The large far range bias is~~ as expected
322   and ~~is~~ is primarily associated with the high statistical errors arising from low SNR. The ~~large~~ increased bias at near-
323   range ~~bias~~ altitudes ~~is more complicated and~~ could be associated with various factors, primarily the aerosol
324   correction and the merging of the signals or ozone retrievals from different optical or altitude channels.

325   **3.3   Lidars versus P-3B Chemiluminescence Instrument**

326   During the campaigns, the P-3B aircraft measured ozone profiles while doing spirals above the lidar sites.
327   There are 34 coincident profiles between TROPOZ and the P-3B at Fort Collins, 29 between TOPAZ and the P-3B
328   at the BAO tower, and 9 between LMOL and the P-3B at Golden, CO. The distances between the lidar and the P-3B
329   spiral centers for these paired profiles were less than 11 km. To make coincident pairs between P-3B and lidar data,
330   we interpolate the P-3B data onto the lidar vertical grids with a 15-m vertical resolution. Figure 4 shows the average
331   ozone profiles measured by the lidars and the P-3B as well as their mean relative differences. TROPOZ and the P-
332   3B agree with each other within ±5% between 0.5 to 3.5 km (black lines in Figure 4 ~~a~~, b) with a -0.8% overall
333   average relative difference. The 1-σ standard deviation of the mean differences (purple lines in Figure 4 b) stays
334   almost entirely within the expected uncertainties (green lines) which include both calculated lidar measurement
335   uncertainties and a 5% constant uncertainty for the P-3B. TOPAZ agrees with the P-3B within -11% and 3%
336   between 0.5 and 2 km (Figure 4 c, d) with a -2.7% overall average relative difference. TOPAZ underestimates the

10

337 lower-PBL (<1.5 km) ozone compared to P-3B, but when compared to ozonesondes TOPAZ overestimates ozone at
338 many of these same altitudes (see Figure 3 d). LMOL agrees with P-3B mostly within -5% and 0% above 1800 m
339 and within -15% and -5% between 0.7-1.8 km (Figure 4 e, f) with a -4.9% overall average relative difference. The
340 1-σ standard deviation of the LMOL-P3-B relative differences is mostly between 5% and 8% and is close to their
341 combined precision (6%). The 1-σ of the mean differences for both TOPAZ and LMOL (purple lines in Figure 4 d,
342 f) stays within the expected uncertainty (green lines) except for the bottom altitudes.

343 In summary, TOPAZ and LMOL exhibited noticeable negative bias in the PBL compared to the P-3B while
344 TROPOZ measured slightly lower than the P-3B. The differences between the ~~two~~ three lidars and the P-3B are not
345 significantly correlated suggesting that the~~se biases~~ ~~problem was not~~were not caused by ~~likely from~~ the P-3B ozone
346 instrument. These differences could at least in part be caused by the lidar systematic errors mentioned in Section
347 2.1.5, but could also reflect horizontal ozone variability across the P-3B spirals, which were up to 22 km in
348 diameter~~.~~

349 **4.   Summary and Conclusions**

350 Intercomparisons have been made between three of the six TOLNet ozone lidars (NASA GSFC's
351 TROPOZ, NOAA ESRL's TOPAZ, and NASA LaRC's LMOL) and between the lidars and other *in situ* ozone
352 measurement instruments using coincident data during the 2014 DISCOVER-AQ and FRAPPÉ campaigns at
353 NOAA's BAO in Erie, CO. On average, TROPOZ, TOPAZ, and LMOL reported very similar ozone within their
354 reported uncertainties for a 5-min signal integration time. The three lidars measured consistent ozone variations
355 revealed in the lidar time-height curtains and in the distribution of their relative differences. From intercomparisons
356 between the lidars and other instruments we find (1) All of the lidars measure higher ozone than ozonesondes with
357 an average~~d~~ relative difference within 4.4%. The lidar profile measurements agree with the ozonesonde observations
358 within -10-15% ~~in their measurable ranges~~ except at a few ~~near~~far-field altitudes. These results are generally
359 consistent with Sullivan et al. (2015) from a similar ozonesonde-lidar intercomparison. (2) TROPOZ agrees with the
360 P-3B chemiluminescence ~~I~~instrument below 3.5 km within ±5% with a small column-averaged relative difference of
361 -0.8%. TOPAZ and LMOL exhibit a slightly larger bias mostly between -15% and 5% below 2 km compared to the
362 P-3B with a column-averaged difference of -2.7% and -4.9%, respectively.

363 ~~Overall, interc~~Comparisons ~~between themselves~~among the three TOLNet lidars and with *in situ*
364 instruments suggest that the ~~TOLNet~~ lidars are capable of capturing high-temporal tropospheric-ozone variability
365 and of measuring tropospheric ozone with an accuracy better than ±15% in terms of their vertical resolving
366 capability and better than ±5% in terms of their column measurement. These lidars have sufficient accuracy for
367 model evaluation and satellite validation (Liu et al., 2010). Since the 2014 campaigns, all of the TOLNET lidars
368 have been modified to improve their stability and their accuracy. The validation of these upgraded lidars will be
369 reported in a future paper.

370 **Acknowledgement**

378

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	1st-order (differential) SG filter with a 2nd degree polynomial with an increasing window width applied on the derivative of the logarithm of the signal ratios	five-point least square fitting with a 450-m window applied on the derivative of the logarithm of the signal ratios	1st-order (differential) SG filter with a 2nd 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

382 **Table 2. ~~Estimated~~ Maximum 1-σ uncertainties for TROPOZ, TOPAZ and LMOL daytime ozone measurements within**
383 **their measurable range ~~(see Table 1)~~ for the 5 and~~or~~ 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%

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384

385 [a] Range dependent and increasing with altitude.

386 [b] Range dependent and typically maximized at the near range.

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

388

389 ~~*Total root-mean-square error.~~

390

391

392 **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})	1σ of the ozone column average (10^{16} molec·m^{-3})	Mean relative difference *	1σ 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/T OPAZ	28	98.1/102.0	13.1/13.0	-3.8%	2.9%

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

394

14

(a)

(b)

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396
397
398

15

(c)

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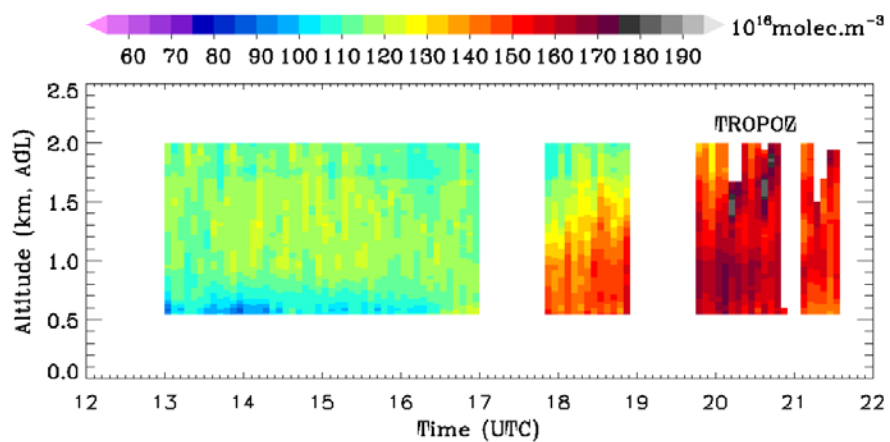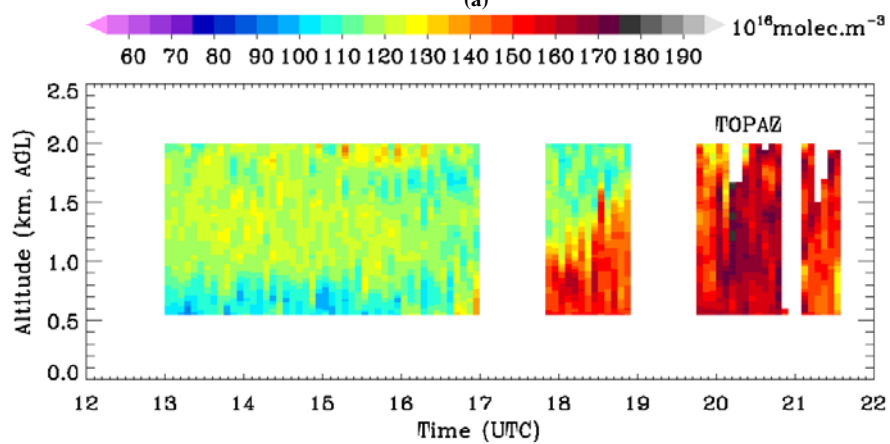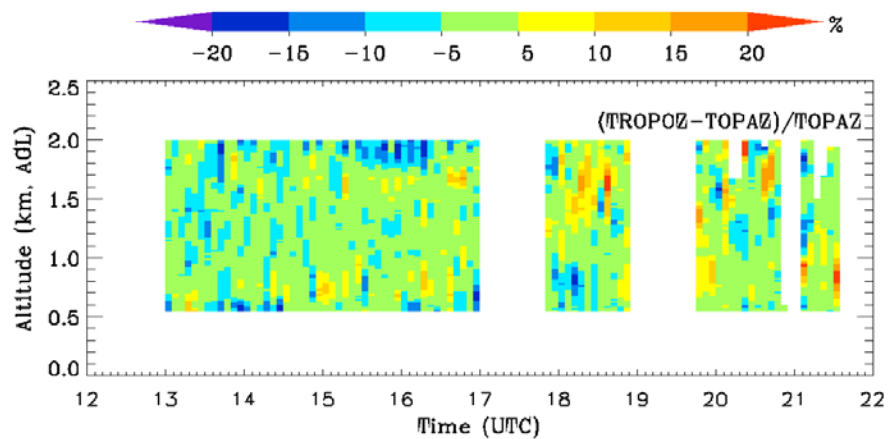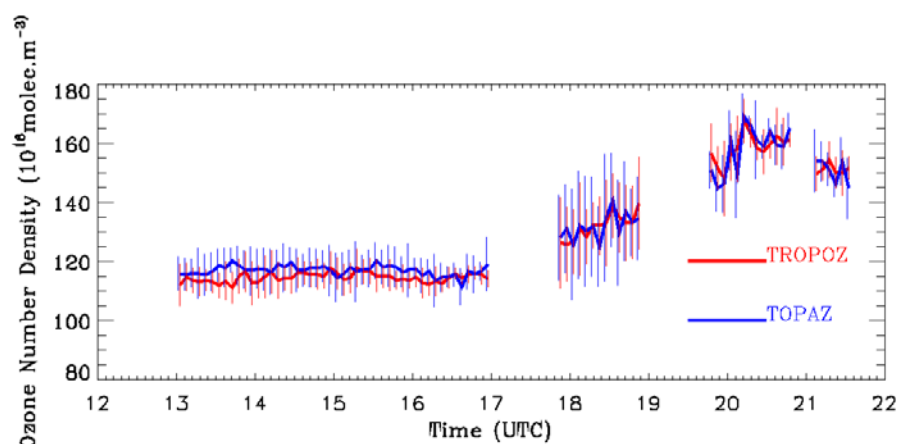
Figure 1. Comparisons of ozone measured by TROPOZ and TOPAZ. (a) Ozone number densities measured by TROPOZ. (b) Ozone number densities measured by TOPAZ. (c) Their relative percent differences, (TROPOZ-TOPAZ)/TOPAZ. (d) Column averages measured by the TROPOZ and TOPAZ as well as their 1-σ standard deviations.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

16

(a)

(b)

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413

414   **(c)**

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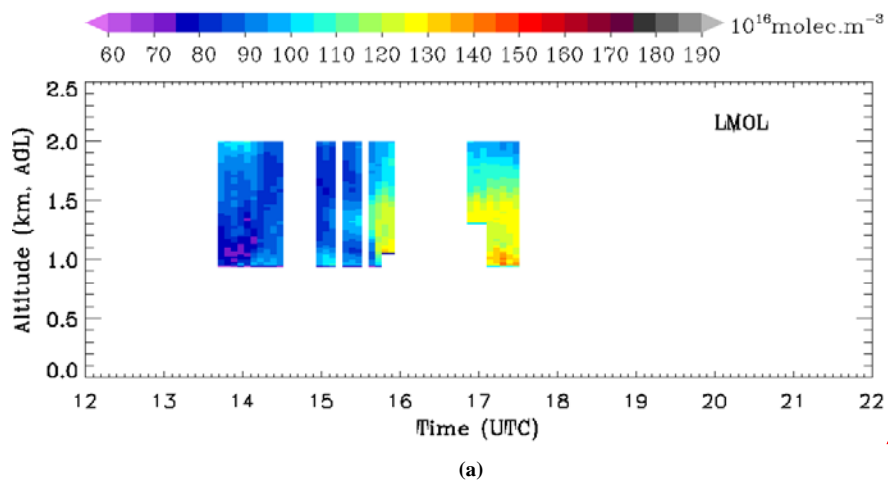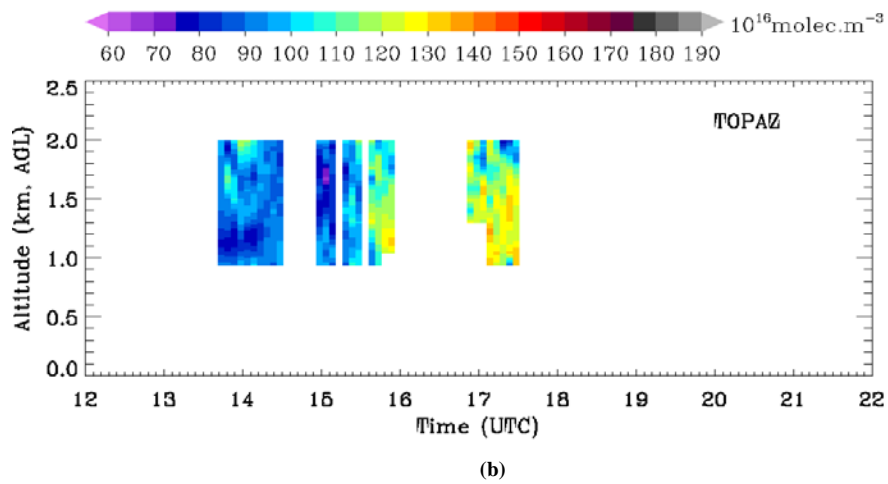
416   **(d)**
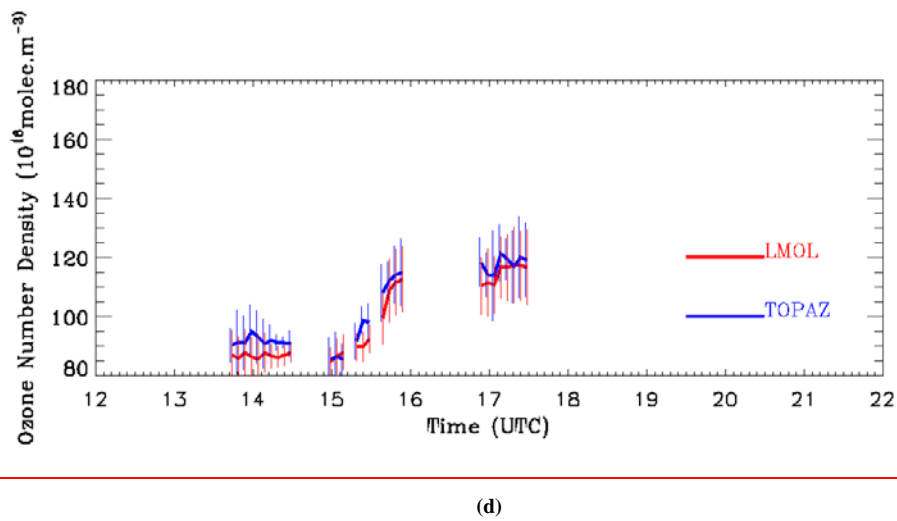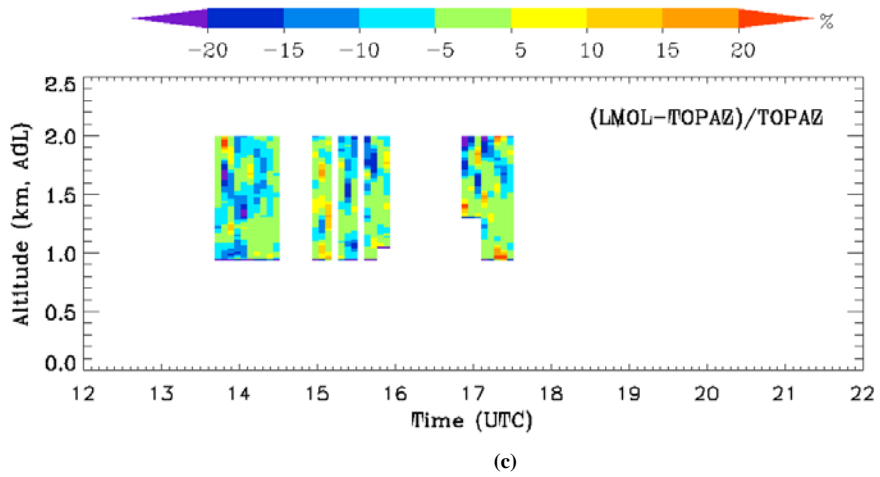
Figure 2. Comparisons of ozone measured by LMOL and TOPAZ. (a) LMOL-measured ozone number densities. (b) TOPAZ-measured ozone number densities. (c) Their relative percent differences, (LMOL-TOPAZ)/TOPAZ. (d) Column averages measured by LMOL and TOPAZ as well as their 1-σ standard deviations. LMOL measures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

421

(a)

(b)

422

(c)

(d)

423
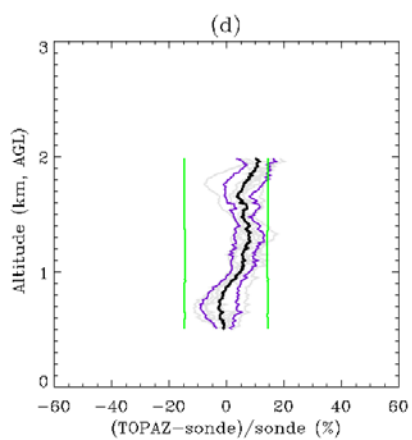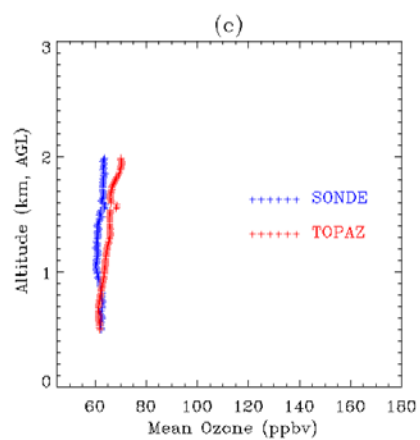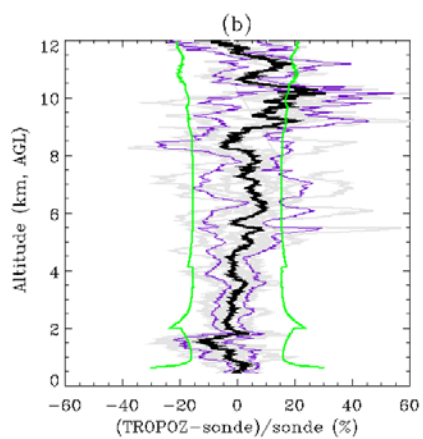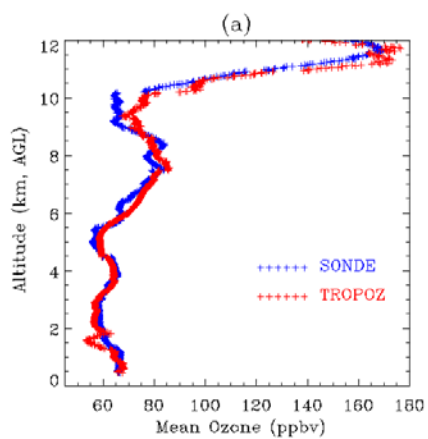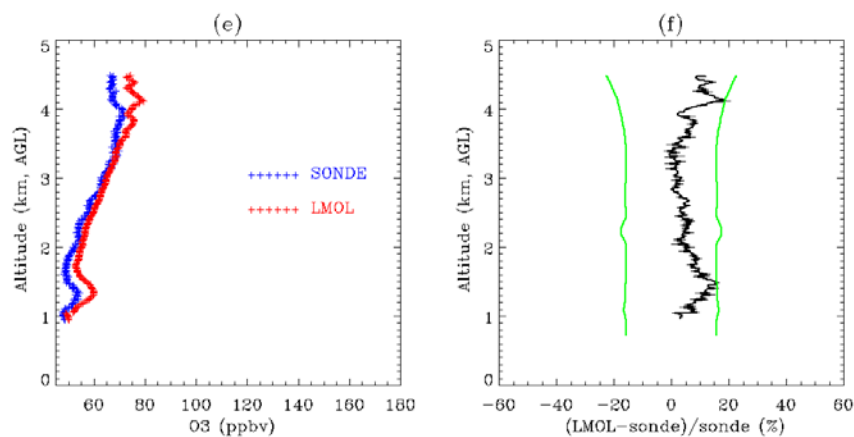
Figure 3. Comparisons of lidar and ozonesonde measurements. (a) Average ozone profiles measured by TROPOZ and ozonesondes at Fort Collins, CO (11 pairs). (b) Mean relative difference (black) between TROPOZ and ozonesondes as well as the 1-σ standard deviations (purple). (c) Average ozone profiles measured by TOPAZ and ozonesondes at BAO Tower (7 pairs). (d) Mean relative difference (black) between TOPAZ and ozonesondes as well as the 1-σ standard deviations (purple). (e) Average ozone profiles measured by LMOL and ozonesonde at the BAO tower (1 pair). (f) Relative difference between LMOL and ozonesonde. The gray lines represent the individual difference profiles between the lidar and sondes. The green lines represent the expected uncertainties including the 30-min lidar measurement uncertainties (also see Table 2) and a 10% constant uncertainty for ozonesondes.

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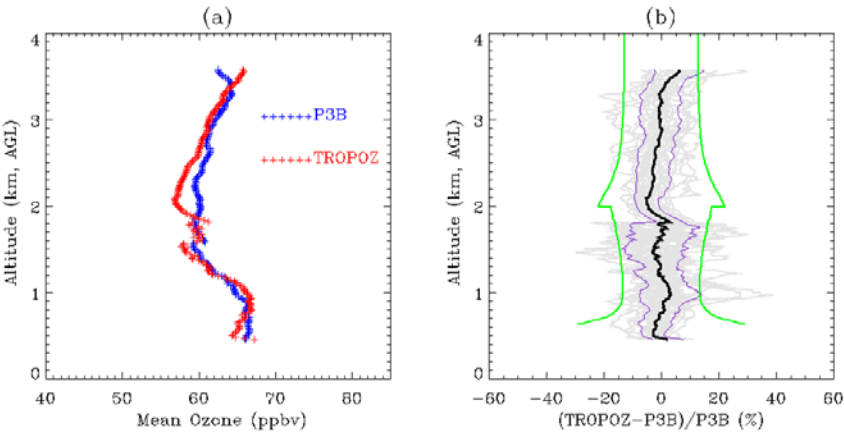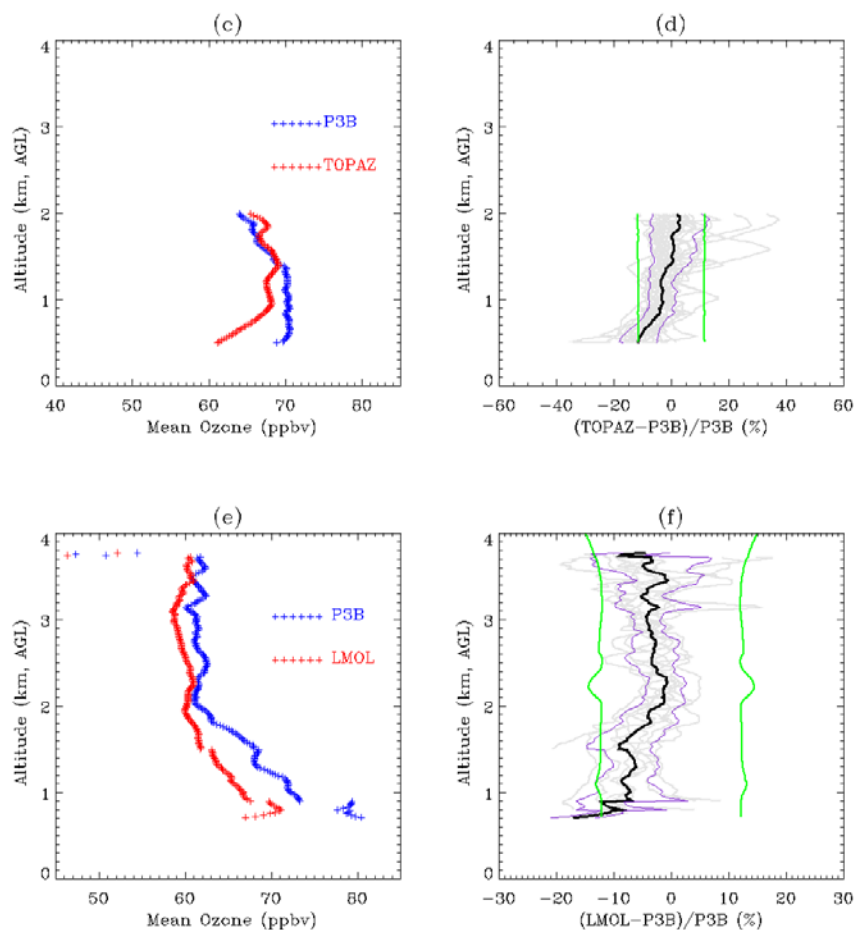
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Figure 4. Intercomparison between the lidar and P-3B measurements. (a) Average ozone profiles measured by TROPOZ
and P-3B at Fort Collins, CO (34 profiles). (b) Mean relative difference (black) between TROPOZ and P-3B data as well
as the 1-σ standard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

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