In the AMT-2018-108 manuscript titled “A fully autonomous ozone, aerosol and night time water vapor LIDAR: a synergistic approach to profiling the atmosphere in the Canadian oil sands region” Strawbridge et al. describe the development and application of the AMOLITE autonomous lidar for profiling of ozone, aerosol and water vapor. In the first part of the paper, the authors discuss the hardware upgrades to an existing Environment Canada autonomous aerosol lidar to enable unattended ozone and water profiling and they provide a detailed account of their efforts to validate the ozone lidar component of AMOLITE. In the second part of the paper, the authors show three multi-day measurement examples from the one-and-a-half-year (and counting) deployment of AMOLITE in the Canadian oil sands region in northeastern Alberta. They use the AMOLITE profile measurements together with in situ chemistry and radar-RASS wind profiler observations to characterize the transport patterns and source regions of atmospheric pollutants observed in the oil sands region.

While unattended aerosol and Raman water vapor lidars have been successfully operated before, the autonomous ozone lidar component of AMOLITE is the first of its kind. The authors have used a tried-and-true approach for the ozone lidar transmitter (Raman shifting of quadrupled NdYAG laser output in a gas cell), then hardened the instrument, built in redundancy, and designed a sophisticated environmental control system. The fact that AMOLITE has been operating continuously for over a year in the harsh environment of northern Alberta is remarkable. This instrument represents a breakthrough in our ability to monitor over long time periods the vertical structure and transport processes of ozone in the troposphere. The water vapor and aerosol profiling capabilities provide important additional information that allows fingerprinting of the air masses and identification of ozone source regions.

The topic of the paper fits well within the scope of AMT. The conclusions presented in the manuscript are supported by the data and figures. While the manuscript contains a rather large number of figures I find them all (except for one) necessary to showcase the synergistic advantages of combining long-term profile and in situ observations. Certain parts of the paper have deficiencies and need a significant overhaul (see major comments below). Therefore, **I recommend publication after major revisions.**

<u>Major comments</u>

1) The section on the ozone DIAL technique (2.1) does not cover all the important aspects of the ozone DIAL retrieval and contains several factual errors. The interfering trace gas correction term needs to be included in equation 1 (page 5): $N(z) = \dots + 2\,\Delta\sigma(z) + 2\,\Sigma_I\,\Delta\alpha_i\,n_i(z)]$, with $\Delta\alpha_I$ = differential cross section of interfering trace gas and $n_i(z)$ = number density profile of interfering trace gas. Equation 2 (page 5) is incorrect. It should read: $d/dz\,\{\ln[B_{on}(z)/B_{off}(z)]\} = d/dz\,\{\ln[(1 + S\dots)/(1+ S\dots)]\}$. The term ‘backscatter ratio” is used for the B_{on}/B_{off} ratio and the aerosol to molecular backscatter ratio S as defined by Kovalev et al. The authors point out that the ‘on’, ‘off’, and ‘reference’ wavelengths are 287, 299, and 355 nm, respectively. However, the O_3 lidar wavelengths have only been mentioned in the abstract at this point and are not introduced in the main text until sections 3.2 and 3.3. The last sentence in section 2.1 describes how the ozone lidar raw data are smoothed and how the derivative of the log signal ratios are computed. This is specific to the AMOLITE ozone

retrieval algorithm and does not belong in the general ozone DIAL technique section. I suggest to merge this sentence and the statement about ‘on’, ‘off’, and ‘reference’ wavelengths with section 3.5, rename that section to “AMOLITE Ozone DIAL Algorithm and its Validation”, and swap positions with the current section 3.4. In the current section 3.5 (AMOLITE Algorithm Validation), the authors mention that they selectively tested individual steps of the ozone DIAL algorithm, including deadtime and background correction and Rayleigh extinction correction. These different steps of the retrieval need to be first introduced.

2) The section on the water vapor Raman retrieval technique (2.2) can be shortened, especially the number of equations. The authors describe in rather great detail the derivation of the equations underpinning the water vapor Raman retrieval. I suggest to just list equations 7-9.

3) In the AMOLITE Instrument Validation and Calibration section (3.4), the authors describe the rigorous validation of the ozone DIAL part of AMOLITE during the SCOOP campaign. Did the authors also validate the Raman water vapor channel in a similar manner?

4) It is unclear to me whether the authors apply differential aerosol backscatter and extinction corrections. Both correction terms are listed in equation (1) in section 2.1, but no mention of the aerosol correction terms is made in the algorithm validation section (3.5). Aerosol correction is important, especially in or near industrial or fire plumes that seem to regularly impact the Oski-otin site, because of the strong aerosol gradients that can be associated with these plumes. An example for this is a piece of a fire plume observed with AMOLITE on 31 August 2017 around 2000 UTC between about 1000 and 2000 m agl (Fig. 14b), with sharp aerosol gradients near the bottom and top of the plume. The coincident ozone measurements (Fig. 14a) show very high ozone concentration near the bottom of the plume and very low ozone concentrations near the top of the plume. This is consistent with O_3 biases due to differential aerosol effects (the authors allude to this in section 4.1, page 17, lines 1-3). Similar high/low O_3 couplets can be found on 30 August near 2500 - 3000 m agl again near aerosol blobs with sharp gradients at their boundaries. The authors need to clarify whether or not they applied aerosol correction and how they did or how they plan to implement the corrections, including aerosol backscatter retrieval at 355 nm, and choice of Angstroem coefficients for both aerosol backscatter and extinction.

5) For the same reasons as pointed out under 4), biases in ozone lidar measurements can also occur near the bases of clouds when cloud returns are not properly filter out. This appears to be the case on 12 November 2016 around 1500 UTC at about 2000 m agl. Did the authors filter out cloud returns and, if so, which approach did they use?

6) The CAM1 in situ instruments at the Oski-otin site occasionally observe large SO_2 concentrations (up to 70 ppbv on 8/29/17, 27 ppbv on 9/9/17, and 18 ppbv on 11/11/16), presumably when the industrial plumes get advected to the site. Did the authors make an attempt to correct for SO_2 interference in their O_3 lidar analysis? Using the Brion et al. (1992-1998) O_3 and Vandaele, Hermans, and Fally (2009) SO_2 absorption cross section data, I get an interference term of approximately 36%*SO_2 concentration for the 287.2 / 299.14 wavelength pair, which would result in O_3 mixing ratio overestimations of about 25, 10, and

6 ppbv for the above SO_2 values. With only surface SO_2 measurements available, it is difficult to estimate SO_2 concentrations at the O_3 lidar altitudes (>= 500 m agl). Perhaps one could use mixing heights determined from the aerosol lidar observations to get a rough idea what the SO_2 concentrations might be at the lower O_3 lidar altitudes. Most of the time, the SO_2 concentrations are small enough, so that the interference term is insignificant. However, in the cases of concentrated SO_2 plumes impacting the Oski-otin site, the authors need to provide an estimate of the error in their O_3 lidar measurement due to SO_2 interference.

Minor comments

Page 2, lines 7-9: "The advantage of nearly …."
I suggest to leave this sentence out. It sounds more like a statement one would find in a conference abstract.

Page 3, line 13
"… atmospheric mixing processes" or "… atmospheric mixing and **exchange** processes"?

Page 4, line 4-5: 'Dynamics' is a bit misleading here. How about "… advance our understanding of **the trace gas distribution in the lower atmosphere**…"?

Page 4, lines 23-24: "The first AMOLITE ozone and water vapor profiles at the Oski-otin ground site in Fort McKay, AB were acquired on 3 November 2016."

Page4, lines 31-32: Reference for CAM1 measurements is missing. Also, state briefly which gases and particulates are measured by the CAM1 instruments.

Page 5, line 14: "**The AMOLITE instrument** uses three different …"

Page 5, line 15: Aerosol profile measurements at the third wavelength (1064 nm) are not shown in this paper. What is the reason for that?

Page 5, line 17: "… to measure t**he** water vapor profile."

Page 5, line 19-20: "… used in AMOLITE"

Page 5, line 25: "… to have a **sufficiently** large difference …

Page 6, line 2: "… differential ozone absorption **cross section**, …

Page 6, line 3: "… the total **two-way extinction coefficient** differential"

Page 6, lines 4-9: "Solving for the component …" Awkward and confusing sentence. Please restate.

Page 6, line 14: “… Savitzky-Golay convolution **to compute the derivative with respect to altitude of the signal ratio and B_{on}/B_{off}.”** Move to new section 3.4 as suggested above.

Page 8, lines 9-13: The explanation of the symbol S in equation 9 is unclear.

Page 8, line 23: “… the trailer needed to **have a** slightly larger…”

Page 9, line 14: “… between 6000 and 24000 BTU of cooling with external temperatures as low as -40C …” 6000 and 24000 BTU of **heating**? Perhaps list the full external temperature range and state the heating and cooling capacities?

Page 9, line 20: “…improvements of the trailer infrastructure …”

Page 10, line 5: “… are triple-coated … 50 mmm optic**s** …”

Page 11, line 16: “… to compare **AMOLITE** ozone profiles **to** other LIDAR instruments …”

Page 11, line 21: Leblanc et al., 2016b is missing in the reference section.

Page 11, lines 27: “… measurements **at night** will reach a range of over 10 km **agl** and dip to 7 km **agl around midday when solar background is high**”

Page 11, line 29: “… staying within approximately 10% of the ozone sonde values …” Differences are up to 20% at several altitudes and approx. 50% at 7.5 km MSL in Fig. 5b. Please reword this statement.

Page 12, line 3-4: “This **is** shown in Figure 6 …”

Page 12, line 11 “… throughout the **diurnal cycle** …”

Page 12, lines 24-25: “**The water vapor measurements below 4 km on 10 August show** very dry air …”

Page 13, line 7: “… both the simulate**d** LIDAR data and …”

Page 13, line 24: “… all corrections **were** turned off …”

Page 13, lines 28: “… 287.20 and 299.14 nm”

Page 14, line 7: Leblanc et al., 2016a is missing in the reference section.

Page 14, line 18: The authors state that individual AMOLITE ozone profiles get truncated at an altitude where the total ozone uncertainty exceeds 15%. However, in the ozone curtain plots (e.g. Fig. 8a) the random error appears to exceed 15% at times at the upper altitudes. Please clarify.

Page 15, line 1: “… the impact of long range **transport** events, …”

Page 15, line 9: The term ‘backscatter ratio’ is used for B_{on}/B_{off} and S in section 2.1. The aerosol backscatter ratio plotted in Fig. 9c (and 12a, 14b, 19b, and 21) appears to be $B_{355} = 1 + S_{355}$. This is very confusing. Please clarify in section 2.1 what the different definitions are.

Page 15, lines 10-11: “… intrusions **were** observed (and evidence that a third **started** on 13 November)”

Page 15, lines 25-27: “There is reasonably good agreement between the ground level measurements **and** the DIAL measurements …”

Page 15, lines 27-28: The authors state that “the lowest few lidar bins can be unreliable” due to changes in the overlap function caused by temperature fluctuations in the trailer. Some of the ozone curtain plot figures (e.g. Fig 8a) and the ozone time series comparison plots with the surface observations (Figs. 15a and 20a) clearly show that. I suggest to blank out and not show these potentially biased O_3 DIAL measurements at the lower altitudes.

Page 15, lines 29-30: “…and the mixing of the lowest water vapor region …”. What do the authors mean by that? Please reword.

Page 16, lines 3-4: Fig. 11a indicates that the stratospheric intrusion occurred on 11 November, so elevated O_3 during the night on 10 November is likely due to some other process. As the authors allude to later, transport of O_3 from the industrial area is a possibility. But mixing of higher O_3 concentrations from aloft down to the surface due to mechanical mixing associated with wind speed or direction shear need to be considered.

Page 16, line 12: “... where there **are** …” Instead of ‘sulphate’ I suggest using ‘sulfur compounds’ to refer to the H_2S, SO_2, and sulfate aerosol measurements.

Page 16, line 18: Add reference: Jaffe and Briggs, 2012 (Jaffe, D and N. Briggs. (2012). Ozone production from wildfires: A critical review. ATMOSPHERIC ENVIRONMENT. 51. 1-10. 10.1016/j.atmosenv.2011.11.063)

Page 16, line 27: “**The diurnal cycles of ozone over three days are** shown …”

Page 17, lines 5-9: Radar RASS wind direction shown in Fig 16 remains SSE when the smoke plume arrives and does not appear to change very much. Omit this figure and text reference to it.

Page 17, line 23: “… approximately **from** 40 to 65 sr”

Page 17, line 13: “… for determining the **aerosol type** …”

Page 17, line 28-29: “A more typical plot …” Omit this sentence.

Page 18, line 1-3: The S ratio profile can clearly change significantly from night to day, even within the same air mass. Omit this statement or word more carefully.

Page 18, line 6: "… **shows** several processes …"

Page 18, lines 12-16: "There are also time periods **(6-8 September around 1200 UTC)** where these near 0 ozone features appear to reach closer to the ground**,** extending **from 400 to 2000 m.**"

Page 18, line 20, "… on 8 September **are** an artifact …"

Page 18, line 21: "… on September 4 range**d** from …"

Page 18, lines 27-28: "The ground level ozone **increased to 70 ppbv** around 18 UTC on 7 September **and dropped to 50 ppbv** around 300 UTC on 8 September, **which was** mostly due …"

Page 19, lines 3-4: "… reaches values of up to **35** ppbv"

Page 19, lines 13-14: "… slightly smaller (35 to 55 sr) **compared to 31 August**, likely indicative of more aged smoke **(**see the … in Figure 23c**)**."

Page 19, line 17: "… average **when** the smoke plumes were present on …"

Page 19, lines 20-21: Aerosol backscatter and extinction often vary differently with wavelength and thus have different Angstroem coefficients. The Angstroem coefficient typically refers to aerosol extinction, which can be computed from the extinction profiles for 355 and 532 nm. The lidar ratio S at different wavelengths is not needed. In fact, the Angstroem coefficient computed from the wavelength dependence of the S ratio convolves the wavelength dependencies of aerosol extinction and backscatter.

References

Page 22, line 27: Ortiz-Amezcua reference is not in alphabetical order.

Figures

Page 25, line 15: "…for (a) 401 UTC on 10 August and (b) 2103 UTC on 16 August.

Page 25, line 21: "… due to cloud**s or high** daytime background **light**"

Page 26, line 12: "… (c) sulphates (d) PM2.5 …"

Page 26, line 24: "… (c) sulphates (d) PM2.5 …"

Figure 5: x-axis label middle plots: (AMOLITE – Sonde)/Sonde (%)

Figure 6: x-axis label middle plot: (Lidar – Sonde)/Sonde (%)

Color curtain plots in Figs. 7, 8, 9, 11, 12, 14, 17, 19, 21, 22: Add numbers at major tick marks of the color scales, not only minimum and maximum values.

Figure 11: Add (b)

Figure 12: Add (b), (c), (d)

Figure 14: Add (a), (b), (c), (d)

Figure 16: Omit (see above)

Figure 17: Create separate scales (similar to Fig. 22)

Figure 19: Add (a), (b), (c), (d)

Figure 22: Add (a), (b), (c). Also, axis labels and color scale are very fuzzy.

Figure 23: Add (a), (b), (c)