

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.**

The authors would like to thank the reviewer for their comments and suggestions.

Every plot was re-done for the paper which was a tremendous amount of work that required considerable coding as the plotting software is a custom package.

#### Major comments

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):  $N(z) = \dots + 2 \sigma_{\text{I}} \sigma_{\text{I}}(z) + 2 \sigma_{\text{I}} \sigma_{\text{I}} \sigma_{\text{I}} n_{\text{i}}(z)$ , with  $\sigma_{\text{I}} \sigma_{\text{I}} =$  differential cross section of interfering trace gas and  $n_{\text{i}}(z) =$  number density profile of interfering trace gas. Equation 2 (page 5) is incorrect. It should read:  $d/dz \{ \ln [B_{\text{on}}(z)/B_{\text{off}}(z)] \} = d/dz \{ \ln[(1 + S...)/(1 + S...)] \}$ . The term ‘backscatter ratio’ is used for the  $B_{\text{on}}/B_{\text{off}}$  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  $\text{O}_3$  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. Indeed, the reviewer is correct – the equations have been changed. I have also moved the last paragraph from Section 2.1 to Section 3.5 and re-ordered it to become Section 3.4 as suggested. More details have now been provided on the various corrections applied to the retrieval

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. Section 2.2 was shortened to only include equations 7-9 which are now of course renumbered.

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? The calibration of the Raman water vapor channel is referenced – we used the sonde data during SCOOP - added text

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<sub>3</sub> biases due to differential aerosol effects (the authors allude to this in section 4.1, page 17, lines 1-3). Similar high/low O<sub>3</sub> 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 Angstrom coefficients for both aerosol backscatter and extinction.

Text has now been added to section 3.4 and plots redone

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? Figures redone – the ozone plots were cloud-screened using the aerosol LIDAR 6) The CAM1 in situ instruments at the Oski-otin site occasionally observe large SO<sub>2</sub> 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<sub>2</sub> interference in their O<sub>3</sub> lidar analysis? Using the Brion et al. (1992-1998) O<sub>3</sub> and Vandaele, Hermans, and Fally (2009) SO<sub>2</sub> absorption cross section data, I get an interference term of approximately 36%\*SO<sub>2</sub> concentration for the 287.2 / 299.14 wavelength pair, which would result in O<sub>3</sub> mixing ratio overestimations of about 25, 10, and

6 ppbv for the above SO<sub>2</sub> values. With only surface SO<sub>2</sub> measurements available, it is difficult to estimate SO<sub>2</sub> concentrations at the O<sub>3</sub> 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<sub>2</sub> concentrations might be at the lower O<sub>3</sub> lidar altitudes. Most of the time, the SO<sub>2</sub> concentrations are small enough, so that the interference term is insignificant. However, in the cases of concentrated SO<sub>2</sub> plumes impacting the Oski-otin site, the authors need to provide an estimate of the error in their O<sub>3</sub> lidar measurement due to SO<sub>2</sub> interference. -Text was now added – data was screened out of the plots – as the reviewer noted it is very difficult to estimate SO<sub>2</sub> concentrations aloft – plots were redone

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. Removed this sentence as requested

Page 3, line 13

“... atmospheric mixing processes” or “... atmospheric mixing and **exchange** processes”?

Added “exchange”

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...**”?

Changed wording to above

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.”

Changed to wording above

\*\*\*\* The page and line numbers going forward from here do not match the page and line numbers on the PDF file in AMTD. The other reviewer page and line numbers matched so not sure what happened but it has made it more difficult to reference the corrections\*\*\*\*

Page 4, lines 31-32: Reference for CAM1 measurements is missing. Also, state briefly which gases and particulates are measured by the CAM1 instruments.

Changed text as no reference available yet for CAM1 – the list of observables is quite extensive, only a small number of them are used in this manuscript

Page 5, line 14: “**The AMOLITE instrument** uses three different ...”

Changed text to above

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?

Text added in Section 3.2

Page 5, line 17: “... to measure **the** water vapor profile.”

Added “the”

Page 5, line 19-20: “... used in AMOLITE”

Changed “employed” to “used”

Page 5, line 25: “... to have a **sufficiently** large difference ... Added “sufficiently:

Page 6, line 2: “... differential ozone absorption **cross section**, ... Added “cross section”

Page 6, line 3: “... the total **two-way extinction coefficient** differential” Changed wording

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

Changed wording

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. [Added text](#)

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..."

[Changed wording](#)

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?

[Added text](#)

Page 9, line 20: "...improvements of the trailer infrastructure ..."

[Deleted text](#)

Page 10, line 5: "... are triple-coated ... 50 mmm optics ..."

[Removed "a"](#)

Page 11, line 16: "... to compare **AMOLITE** ozone profiles **to** other LIDAR instruments ..."

[Replaced "LIDAR" with "AMOLITE"](#)

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

[Changed "b" to "a" and added correct reference](#)

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**"

[Changed wording](#)

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.

[Added a few sentences to clarify](#)

Page 12, line 3-4: "This **is** shown in Figure 6 ..."

[Changed "can be" to "is"](#)

Page 12, line 11 "... throughout the **diurnal cycle** ..."

[Changed "daylight" to "diurnal"](#)

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

[Changed wording](#)

Page 13, line 7: "... both the simulated **LIDAR** data and ..."

[Added "d"](#)

Page 13, line 24: "... all corrections **were** turned off ..."

[Added "were"](#)

Page 13, lines 28: "... 287.20 and 299.14 nm"

[Removed "nm"](#)

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

[Reference added](#)

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.

[Added a few sentences.](#)

Page 15, line 1: "... the impact of long range **transport** events, ..."

[Already addressed](#)

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. Clarified text

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

Changed the two words

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

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<sub>3</sub> DIAL measurements at the lower altitudes.

Figures changed

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

Removed text

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

Made note that the aerosol LIDAR shows plume signature

Page 16, line 12: “... where there **are** ...” Instead of ‘sulphate’ I suggest using ‘sulfur compounds’ to refer to the H<sub>2</sub>S, SO<sub>2</sub>, and sulfate aerosol measurements.

Changed wording

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)

Reference added

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

Added text

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.

Deleted text and figure

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

Page 17, line 13: “... for determining the **aerosol type** ...” Deleted “parcel”

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

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.

Deleted sentence

Page 18, line 6: "... **shows** several processes ..." Changed "has: to "shows"

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.**"

Changed wording

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

Changed "is" to "are"

Page 18, line 21: "... on September 4 ranged from ..."

Added "d"

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 ..."

Changed text

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

Changed text

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)."

Changed text

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

Changed "during" to "when"

Page 19, lines 20-21: Aerosol backscatter and extinction often vary differently with wavelength and thus have different Angstrom coefficients. The Angstrom 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 Angstrom coefficient computed from the wavelength dependence of the S ratio convolves the wavelength dependencies of aerosol extinction and backscatter.

Yes you are correct – text changed

References

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

corrected

Figures

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

corrected

Page 25, line 21: "... due to clouds **or high** daytime background **light**"

Changed text

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

corrected

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

corrected

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

Plot redone to change axis labels

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

Plots redone with new color bar

Figure 11: Add (b) added

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

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

Figure 16: Omit (see above)

Deleted figure

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

Created separate scales

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

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

Plots redone

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