Manuscript 2017-391: Aggarwal et al., Airborne Lidar Measurements …

Response to Comments from Reviewer 2

Note: changes in the revised manuscript have been highlighted in yellow

General Comment from the Authors

The authors appreciate the meticulous work by both reviewers. Substantial improvements have been made in response.

The first author recently completed her Ph.D. examination and through that process there have been a few improvements to the analysis that were not requested by the reviewers.

a) Equation 5 of the submitted manuscript included an approximation in terms of the effective radius. This was not necessary. This equation now has the integral over the size spectrum as for the other calculations in equations (3), (4), (6) and (7). The explanation is now easier to follow and the calculations are more accurate. The calculated aerosol corrections are slightly different, but the results have not changed.
b) The temporal averaging was previously 1.3 minutes for some figures and 3 minutes for others. The temporal averaging is now consistent as 1.5 minutes for all of the analysis in figures. This corresponds to a distance of about 7.5 km along the flight track. This had an impact on the histograms of Fig. 16 since previously the averaging was 3 minutes. Now the averaging is 1.5 minutes for both the lidar and in situ measurements represented in Fig. 16. The description of the histograms has been changed slightly on page 18, lines 7 to 25.

Reviewer 2, Comment 1

The authors should discuss the actinic flux field in conjunction with the discussions of transmittance, absorption, scattering, photochemical formation, etc. What was the cloud condition? Was it cloudy, hazy, humid? All of these MET variables would make a difference when considering ozone production/destruction.

Response to R1, Comment 1

Radiative flux was not measured as part of this study. There was some consideration of the conditions in the discussion section from page 19, line 25 to page 20, line 16. We have also added a brief reference to the weather conditions on page 13, line 22.

There is a separate study in progress by a separate group of researchers in which the Canadian Air Quality forecast model is being used to assess the production/destruction of ozone. The will properly take into account all of the factors for ozone production and destruction. The results of the current study will inform the model development.

R2, Comment 2

Did you consider potential NOx contributions from the aircraft affecting the titration? Perhaps the altitude was always above the PBL?

Response to R2, Comment 2

The Twin Otter was flying above the boundary layer except for take off and landing. It was very unlikely that the exhaust from the Convair would have been intercepted by the lidar measurements and would have been a very small contribution in comparison to the industrial emissions.

R2, Comment 3

My most significant concern is that this article lacks a discussion of the uncertainty budget calculation from different sources (statistical, background correction, aerosol correction, Rayleigh correction, differential ozone absorption cross section, e.g., Wang et al., 2017) for the ozone DIAL measurement. This discussion is essential.

Response to R2, Comment 3

The largest contribution to the uncertainty in the derived ozone mixing ratio is from the aerosol correction. One reason is that the size distribution of the aerosol particles is not constant with height or horizontal distance. The particle effective radius (area averaged) was found to vary between 0.06 and 0.08 micrometers along a flight through the polluted air above the oil sands. Another source of uncertainty was that the aerosol composition, and thus refractive index, was not known. We had assumed kaolinite in the submitted manuscript since most of the aerosol appeared to be the mineral dust that was emitted due to the surface mining activities.

In response to the reviewers' comment we have computed the aerosol correction for a range of measured particle size distributions and for a variety of aerosol compositions including kaolinite, diesel soot, sulfuric acid, toluene SOA, and ammonium sulfate. Figure 4 now shows a range of aerosol corrections corresponding to the various compositions and particle size distributions. The vertical profiles of derived ozone are now shown as a range that represents the uncertainty due to the aerosol correction. Description of this is found in the manuscript from page 9, line 18 to page 10, line 19

We also now give more attention to the uncertainty due to the bias due to interference by differential absorption of SO_2. The text has been changed in the paragraph starting on page 11, line 17 in response to the referee's comment. We provide a range of values for bias due to SO_2 absorption based on three separate sources of absorption cross sections.

The amount of ozone measured by the lidar is within the range of in situ measurements on the Convair aircraft and at ground sites. There is no discrepancy to explain.

R2, Comment 4

Can you quantify or at least estimate the vertical (spatial) resolution for the ozone lidar, which is an important parameter for profiling instruments. The vertical resolution is closely related to your signal/data processing.

Response to R2, Comment 4

This is now stated more clearly in the paragraph starting on page 5, line 21.

R2, Comment 5

"This paper concerns the methodology and results of airborne lidar measurements of aerosol and ozone …". However, the introduction section does not provide any review of the instrumentation or retrieval technique of either airborne aerosol or ozone lidars.

Response to R2, Comment 5

As this paper was not intended to provide a review, the word "methodology" has been changed to "measurement technique" on page 1, line 26.

Publications concerning previous lidar studies were cited throughout the manuscript where it was appropriate.

R2, Comment 6

What are the conversion efficiencies and final pulse energy for the three wavelengths?

Response to R2, Comment 6

This is now included in the manuscript on page 3, lines 8 to 13.

R2, Comment 7

The authors choose to retrieve ozone separately from analog and PC channels while a more common approach is to merge the analog and PC signals first at a reference counting rate (e.g. Kuang et al., 2011). So, do you then merge the ozone profiles? Can you explain more about how and where you exactly merge the ozone profile, in a constant altitude range or at a single point?

Response to R2, Comment 7

A better description is now provided on page 6, lines 9 to 15.

R2, Comment 8

What is the potential error in retrieved ozone amounts from assuming consistent aerosol composition and size distribution throughout the boundary layer?
The author use the refractive index of kaolinite to compute the extinction and backscatter coefficients based on the studies, that kaolinite to be the prominent clay particle in the oil sands region. Here the quantitative value (e.g. fraction) for the "prominent" role would be better if provided. Do you have any evidence that the aerosols are actually Kaolinite? What is the potential error in retrieved ozone amounts if their composition (and complex refractive index) are different?

Response to R2, Comment 8

We have made some changes in response to this referee comment. The variations in particle size distribution and assumption of aerosol composition are actually the main source of uncertainty in measured O_3 concentration when there is substantial aerosol. In the revised manuscript we are providing an estimate of this uncertainty by showing a range in the magnitude of correction for a variety of aerosol compositions and particle size distributions.

See the description from page 9, line 18 to page 10 line 19

R2, Comment 9

"not particle size", the lidar ratio also varies with aerosol type or refractive index and probably humidity, not only size distribution.

Response to R2, Comment 9

The lidar ratio was taken into account when assessing different aerosol types as in the response to the previous comment.

R2, Comment 10

The GDAS meteorological dataset for HYSPLIT input has two resolution options: 1degree and 0.5degree. Which option did the authors select? For an aircraft measurement up to 150km downwind of emission, would the resolution influence the trajectory accuracy?

Response to R2, Comment 10

We used the 1 degree resolution. Now mentioned on page 13, line 10.

R2, Comment 11

This paragraph lacks scientific analysis. The lack of supporting data such ozone precursors measurement in the fire plume to indicate the chemical production mechanism of fire ozone formation. The authors quote the paper by Jaffe and Wigder, 2012, but didn't provide any further analysis about the influence factors for ozone production (fire emissions, efficiency of combustion, photochemical reactions, aerosol effects on chemistry and radiation), meteorological patterns were mentioned without quantitative analysis.

The meaning of "the temperature would have been greater in the plumes above the fires" is not clear. This paragraph requires quantitative estimation of the environmental variables.

Response to R2, Comment 11

Measurements were not obtained at the fires. All we have are the measurements of enhanced ozone in the forest fire smoke. There is no data beyond that to form the basis for a more in-depth analysis. The point of including the observation was that it stands in contrast to the lack of ozone production in the industrial pollution. The reference to temperature is actually indirectly implying that there would be greater concentration of VOCs in the fire emissions. As we don't have measurements of VOC concentrations in the fire emissions, and as the reviewer objects to the speculation, we have removed the reference to temperature associated with forest fires.

R2, Comment 12

Identify PMT model(s)

Response to R2, Comment 12

Added to page 3, line 21

R2, Comment 13

*Why isn't the 532 smoothing distance an integer multiple of the range bin 23=6.13*3.75?*

Response to R2, Comment 13

That was an error. See the paragraph starting on page 5, line 21.

R2, Comment 14

Equation (3) and (4): did you say anywhere in the context "m" represents the refractive index?

Response to R2, Comment 14

Added on page 8, line 2.

R2, Comment 15

How did you calculate the extinction and backscatter profiles at "the UV wavelengths" based on the profiles at green? Did you assume a value for Angstrom exponent?

Response to R2, Comment 15

Please see all of Section 3.1. We are using the 532 nm lidar measurements and the in situ measurements of particle size distribution to determine the extinction and backscatter coefficient profiles at the UV wavelengths. This analysis does not require an Angstrom exponent.

R2, Comment 16

SO2 absorption cross section may significantly vary with database. Can you give the reference for the source of the SO2 absorption cross section.

Response to R2, Comment 16

More discussion of this is now given in the paragraph starting on page 11, line 17.

R2, Comment 17

There should be a newer reference than (Draxler and Hess, 1998) for HYSPLIT

Response to R2, Comment 17

This was replaced with a more recent reference. Page 13, line 4.

Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., and Ngan, F.: NOAA's HYSPLIT atmospheric transport and dispersion modeling system, B. Am. Meteorol. Soc., **96**, 2059-2077, doi:10.1175/BAMS-D-14-00110.1, 2015.