Responses to Reviewer 2

April 2021

Green – reviewer's comment Black – authors' response

Changes to text were made only when explicitly stated

Reviewer # 2

Summary:

Overall, this is a well-written paper that presents a case study of two flights during an aircraft campaign in the Athabasca Oil Sands Region near Alberta, Canada. The focus of the paper is on how the 4STAR aerosol optical depth (AOD) observations on-board the aircraft compare with the ground-based AERONET observations at nearby sites. The aircraft observations are also compared with in situ aerosol measurements to provide additional context about the composition and size distributions of aerosols associated with individual pollution plumes. The campaign and data are clearly presented and the conclusions seem sound. While the findings are not particularly surprising, this paper would be valuable to the community as an additional data point for interpreting how ground-based remote sensing observations of aerosol optical properties at specific sites compare to the variability associated with pollution plumes in the atmosphere, specifically in this case in the context of industrial pollution sources. I can recommend the paper for publication after some minor revisions and clarifications. We thank the reviewer for constructive comments and provide detailed responses below.

General Comments:

While this paper is presented as a specific case study, I wonder if it would be possible to comment more on the representativeness of the variability of spatial scale observed here. Since the focus was on comparing the aircraft AOD observations with the AERONET observations, it might be useful to understand more about how this compares with observations from previous aircraft campaigns. Are the spatial scales of the plumes observed during the OSMC campaign similar to what is typically observed by 4STAR?

The aerosol plumes similar to those observed during OSMC have seldom been quantified previously. We added an additional figure (Figure 6, comparisons with MODIS AOD) and an accompanying paragraph in section 3.2 to discuss the spatial scales of the observed plumes. Furthermore, we updated Figure S5 which shows 4STAR-AERONET biases as a function of distance from AERONET.

I agree with Reviewer 1 that some additional context, such as satellite measurements, would be helpful for giving the reader a better overview of what is happening. Were there any lidar measurements on the flights that could help to provide context?

We added a case comparison where there are clear indications of a plume from MODIS true color and Dark Target AOD retrievals. See updated section 3.2 and related descriptions. Unfortunately, there were no lidar measurement on board during the campaign.

Specific Comments:

Line. 5 p. 2. "The fact that industrial plumes can be associated with significantly higher AODs in the vicinity of the emission sources than previously reported from AERONET can potentially have an effect on estimating the AOSR radiative impact." "Cursory radiative transfer calculations" indicating 25% increase over background were mentioned at the end of the paper. Could this be expanded upon? 25% increase in terms of what, W/m2 or AOD? This was not clear from the discussion on p. 13, lines 10-15. What were the assumptions going into the calculation here? Presumably this would be a smaller effect than 25% once it is averaged over the entire grid box that the AERONET observations of AOD might be used to estimate. We expanded the last paragraph to include some description of the cursory radiative transfer calculations. Please see amended paragraph. While it was the hope of the authors to include more details on the radiative transfer calculations, this is beyond the scope of the paper.

Figure 2. It might be useful to also show the variance on the average AOD values for each month over the 13 year period. That would be useful for understanding the context of the flight observations.

We added error bars to Figure 2, representing standard deviation of the monthly mean AODs.

Figure 3. There are some points in the AOD time series in pane 1 that appear to potentially be artifacts during periods where there were changes in aircraft altitude (e.g. the very smooth lines between 15:42-15:44, 15:50-15:52, 15:58-16:00, and 16:08-16:10). This is also the case for the UHSAS fine mode observations in pane 2 – can you comment on whether these are interpolation artifacts (and if so remove this data from the plot) or whether there is some other reason (like differences in averaging time) that the observations during these periods are significantly smoother than during the horizontal legs of the flight observations? Figure 6 and Fig. S4 also show similarly smooth periods in some of the time series.

During transformation flights such as the one on June 9, at the end of each leg the aircraft makes a turn to rejoin the track at a higher altitude. Depending on the steepness of the roll and sun position, sun tracking might be challenging during a turn resulting in occasional lack of 4STAR measurements between the altitude levels. We replotted Figures 3, 7 and S4 to ensure that the disjointed 4STAR data segments are not connected by a straight line. Other time-series do not appear to suffer from the same issue.

Figure 4. It might be nicer visually to plot so that the organic aerosol mass portion starts at the bottom of each bar. This would make it easier for the reader to directly compare the organic aerosol mass across altitude levels/plumes and see that it stays relatively constant. Can you speculate about the origins of the June 9th flight plume A and plume B based on their composition?

We replotted Figure 4 with the organic fraction starting at the bottom. We also expanded section 3.2.1 to include a comment on the potential origins of the observed plumes (likely SML/Suncor emissions from upgrading the bitumen and/or mining activities).

p. 8. For context, could you add more details about what this facility is? Is it an oil processing plant? Is there any way to judge the vertical extent of plume A relative to plume B?
We added more details to section 3.2.1 about the potential origins of the observed plumes. Both plumes were still clearly distinguishable (from each other and from the background) in the in-situ measurements at each altitude level up to the highest altitude of 1750 ft. We can't speculate on the plume vertical extent beyond that point.

p.10. Was there any estimate of the contribution of the AOD below flight level for the 4STAR measurements?

No, given that the first flight leg usually started at 500 ft (150 m) above ground level, we expect that our AOD measurements captured most of the vertical extent of the plumes. Including the lowest altitude levels would likely result in slightly larger 4STAR AOD values which would further reinforce our finding about the 4STAR-AERONET differences.

Figure S5. Could you similarly show the relative comparison between the Fort McMurray and Ft. McKay AERONET observations? This might help support the point in the first paragraph on p. 11.

One of the main reasons of not including Fort McKay in our analysis is its limited data availability for June-July 2018 with < 3h of data available on June 18 and no data acquired on June 24 and July 5.

p. 11 – Can you comment on the relative time scales expected for the plume's AOD to increase because of SOA formation compared with the time scale for the plume's AOD to decrease due to plume dilution with the background? Also, can you compare with the SSA observations, as SSA would also tend to be correlated with SOA formation?

There is no easy to estimate relative time scales of these two competing processes (SOA formation and plume dilution). Even if we knew how fast mass was diluted, separate from SOA formation, one would still need to determine the impact on AOD of these processes. Since we see an increase in both AOD and SOA, SOA formation outcompetes dilution (and deposition), so the AOD increase is actually a lower limit to the true AOD increase from just SOA formation. In section 3.3 we do provide SOA formation rates of 1003±193 kg/hr and 443±45 kg/hr for June 9 and July 5, respectively.

Figure 7. Was there variability in AOD for different times of the day for the AERONET observations? Were the AERONET observations at approximately the same time as the flight observations? Also, can you clarify if the time shown on the axis for Figures 3, 6, and S4 is local time or UTC?

Yes, AERONET could have significant AOD variations throughout the day, as expected. In fact, the original Figure 7 shows AERONET AOD daily standard deviations for 4 different days. In addition, Figure S4 shows an example of AERONET FM AOD time series for June 18 (little variation on that particular date) and Figure 8 gives 4STAR and AERONET comparisons for AODs that were acquired within 10 minutes of each other.

We modified the captions to make explicit references to UTC time.

Typos: P. 7 Line 29-31 – This is referencing Figure 2, but it should be Figure 1. Fixed