Responses to Reviewer 1

April 2021

Green – reviewer's comment Black – authors' response

Changes to text were made only when explicitly stated

Reviewer # 1

This paper reports airborne measurements of aerosol properties in the Athabasca area as part of the Oil Sands Measurement Campaign (OSMC) during 2018. Ground-based Sun photometry data collected in the area are also used. The manuscript is in scope for ACP. It is written clearly. The airborne data provide a good amount of detail on various aerosol plumes measured during the flights, which will be useful to the broader community, since (as the paper notes) they haven't been studied in as much detail as some other aerosol systems. One key point here is that the spatial scales of these plumes are such that they can be missed by the ground-based measurements. I would have liked to see a bit more satellite imagery and possibly retrievals to provide a bit more context about spatial variation here. Additionally, the measurements revealed that the plumes had different size and absorption characteristics from one another, i.e. not all the plumes in the area are similar. I don't have any big issues with the material presented here. I recommend publication following minor revisions. I would be happy to review the revised version, if the Editor would find it useful.

We thank the reviewer for constructive comments and provide detailed responses below.

My comments are as follows:

- 1. Throughout the paper, it was difficult to judge the scale of the area and the plumes. I suggest adding a scale in km to Figure 1 so the reader has a sense of size of the overall domain size. We added a scale to Figure 1.
- 2. I suggest a new figure (either one multiple panel, or one for each of the main flights discussed) be added to show a true-color image around the time of the flights? This would help the reader visually see what was going on. I am not sure if MODIS or VIIRS overflew around the right time (or maybe we will have got lucky and there's Landsat or Sentinel 2), but if not there are the new GOES sensors which are every 10 minutes or so. I looked on NASA Worldview for the days but wasn't sure if I could see the plumes there were lots of clouds on some days too so if the authors can provide the relevant imagery so we know what we are looking at, it would be helpful. Here is a link to June 9 imagery, not sure if the plume is visible here (there's a lot of cirrus too), or if the long url will make it through the ACP comment system unmangled: https://worldview.earthdata.nasa.gov/?v=- 114.52655240204368,55.351947254962724,- 108.18835611557154,58.463275379962724&t=2018- 06-09-

T21%3A46%3A08Z&I=Reference_Labels,Reference_Features,Coastlines,VIIRS_NOAA20_CorrectedReflectance

We added a new figure to section 3.2, showing a singular case of MODIS AOD as compared to 4STAR AOD of the Oil Sands processing plume.

3. Satellite retrievals of AOD would also be interesting to show, to reveal whether they resolved the plume structures or not. The MODIS Dark Target 3 km product could be useful here as it is

finer than most others. Again, it's hard to know what the spatial scale is from the paper, so it's possible this would be too coarse already? And if satellite products don't resolve the plume (either structurally or even as a hotspot) that is another interesting point (analogous to the AERONET spatial representation discussed by the authors for this area).

The figure added in response to the above comment has another section showing the comparison of MODIS Dark Target AOD to the airborne sampling from 4STAR. However, it is hard to bring any judgement to the limited sampling presented here, and the potentially large offsets in time, and therefore plume evolution and advection. See the additional new paragraph in section 3.2.

4. Page 5 line 29: the authors mention O'Neill et al (2016) as a reference for cloud screening based on the SDA (i.e. that the fine mode is unaffected). However Smirnov et al (2018) indicate that in the presence of cirrus (or dust) the SDA fine mode AOD can still be biased: https://www.sciencedirect.com/science/article/pii/S0022407317306131 Perhaps the authors can comment on this, particularly as there seemed to be some cloud cover in the satellite images on Worldview.

Smirnov et al. (2018) (SM) is focused on a very particular application of the SDA (Arola et al., 2017 or AR) where AR attempted to retrieve FM AOD from CIMEL measurements that were known to be contaminated by cirrus cloud. Basically SM criticized AR for ignoring and/or assuming that the SDA was somehow exempt from the well-known forward scattering effect of cirrus clouds into the large FOV of CIMEL instruments*. The SDA-focus of SM's paper is also misleading: in actual fact, the FOV effect is not some kind of unintended consequence of applying the SDA (the FM AOD is underestimated as a result of all standard AERONET AOD products being underestimated).

* which, for the record, they did not do: AR stated clearly that "It is likely, however, that the fine-mode AOD is underestimated when cirrus ice crystal clouds overlay the aerosol, due to strong forward scattering into the field of view of the sun photometer (A. Smirnov, personal communication, 2016)."

We also note that dust would generally not be a problem unless the dust optical depths (DODs) are very large (which, with DODs < 0.05, was never the case): the FOV effects of thin cirrus on the standard AOD and the FM AOD are already small enough: the effect of significantly smaller dust particles of relatively weak DOD would be negligible

5. Page 6 lines 3-4: it might be covered in the references cited, but could the authors mention here whether the UHSAS size distribution retrieval requires assumptions about refractive index and if so how sensitive it is to that? This could be relevant as it is an optical sensor, and differences between plume refractive index could mask or magnify differences in particle size between plumes.

Yes, UHSAS sizing will be dependent on the particle refractive index. We added the following phrase to section 2.4:

"The UHSAS sizing was calibrated using NIST traceable polystyrene latex (PSL) nanospheres. Sizing of the UHSAS is dependent on the refractive index and shape of the particles. Differences in refractive index has been estimated to result in a 10% uncertainty in the sizing of the UHSAS (Kupc et al, 2018)."

6. Any other caveats or relevant uncertainty sources associated with the in situ measurements should also be mentioned in Section 2.4.

We added more details to Section 2.4 regarding the AMS uncertainties/method.

- 7. Figure 3: Panel 3 shows UHSAS effective radii around 0.4 microns in Plume A. However, for the same flight (9 June), the lower panel of Figure S4 has all UHSAS data between 0.1 and 0.2 microns. Is this a plotting error in one of the figures, or am I misunderstanding what is shown? It's unclear what the reviewer is referring to. Figure 3 is plotted for the June 9 flight while Figure S4 refers to the June 18 flight. Regardless, panel 3 of Figure 3 shows maximum values of reff below 0.2 (right-hand scale) which is similar Figure S4.
- 8. Figure 5: do the authors believe the narrow peak in plume A around 0.42 microns is real, or could it be an instrumental/retrieval artefact? Any thoughts on what could cause this sharp feature?

Upon further investigation we concluded that this peak was likely an instrument artifact as it was not supported by larger-size particle spectrometers (such as FSSP and FCD). It looks like the problematic bins are close to the boundary of two gain stages in UHSAS calibration curve which could be a potential explanation for this problem. We excluded the problematic 5 bins (r= $0.382 - 0.428 \mu m$) from analysis and added the following sentence to the instrumentation section:

"For some flights we noticed abnormally high particle counts in 5 bins between the radius of 0.382 and $0.428 \, \mu m$. This peak was not supported by other particle spectrometers on the aircraft and is likely an instrument artefact. We removed the problematic data from further analysis and suspect that the issue is related to the uncertainties in UHSAS calibration curve consisting of several individually chosen gains."

9. Page 9 line 6: I am not sure it is quite right to say that AERONET sites are generally assumed to be representative of a distance 100 km around them. Most satellite retrievals use an averaging circle of order 25 km. Even for a model comparison, if it is at 1 degree, then the grid boxes are still only 110 km (i.e. a 55 km box if centered around the site) at the Equator and smaller at the poles. I understand the authors' point here but suggest revising the wording to not say "this distance is often taken as 100 km" because I don't believe that is true. We would tend to disagree with such a small circle of AOD "expansiveness" and suggest that the satellite retrievals are the more likely source of spatial AOD incoherence: the reviewer will recall that on page 11, line 1 of our paper we cite Sioris et al. (2017) whose AEROCAN-wide correlation coefficients required 500 km of interstation distance to drop off by 50%* (in fact the fine mode correlation length** of their Figure 4a was closer to 1000 km)

- * with the Fort Mckay to Fort McMurray contribution to the correlation curve that produced that correlation length estimate being close to the AEROCAN-wide average (personal communication with Chris Sioris)
- ** which the reviewer will agree is a more fundamental indicator of the spatial influence of AOD than a satellite-based inference
- 10. Figure 7: the caption notes that the horizontal bars on the AERONET panel here are standard deviation. What are the horizontal bars on the upper panel? This should be stated. The horizontal error-bars are the std values at each altitude level. The caption was modified to read:

"In both panels the horizontal error bars indicate the AOD standard deviation representing spatial variability of the plumes (per altitude std values for 4STAR and daily std values for AERONET)"

11. Figure 9: I think the caption should read 0.02 here, not 0.002, unless I am misunderstanding.

We are only talking about the difference in layer AOD between screens 2 and 3 (575-975 m - common altitude range between the two spirals), so 0.002 is the correct number. It's the relative increase that's more important here. The calculated AOD of the entire vertical column sampled by neph+CLAP (525-1325 m for screen 3) is actually 0.013 which was stated in the text.

12. Figure S5: I do not think that the regression is valid here. The fitting, p value, and uncertainties are based on the assumption of independent draws from one population of data. What we have here is data from 4 separate flights. Each flight is likely to have some autocorrelation between observations from that flight, and it's not necessarily true that the difference vs. distance would be consistent between all flights. I suggest redrawing this to color code points from the individual points, and add a zero line but perhaps not a regression. The reader can draw their own conclusions and I'm not sure that the regression is needed for the understanding of the paper: I agree that there seems to be some relationship, but caution against over-interpretation based on a small sample of flights.

The figure has been replotted to separate the contributions from each flight, and each regression line applied to the individual flight days. All the flight days show increasing divergence of the AOD with increasing distance from the AERONET site, albeit with different magnitude of slopes and p-values, which are also reported. See the amended figure in the supplement. The main text remains unchanged.