We appreciate the careful consideration of our manuscript by this reviewer. We have considered the points raised and revised our manuscript accordingly. Our detailed responses and all changes that have been made are presented below.

General comments

This manuscript describes the evaluation of the physical properties of rBC particles produced by activities related to the Canadian oil fields. The analysis is detailed and the document is well written. The conclusions are supported by the observations and there are several interesting results that provide useful recommendations for the climate modeling community, as well as suggestions for quality assurance for those who do rBC measurements with the SP2. There are several minor issues and questions that I would like addressed, as well as one fairly major omission that needs to be added.

Major omission.

In general, any study that uses measurements needs to include an error analysis that describes the limitations and uncertainties of the sensing technique. In this presentation, no mention is made of how the aerosols are sampled. I understand that this is probably already done in companion papers, but it is necessary here in order to understand any losses/enhancements that may occur due to the inlet system that is implemented. What is the probability of evaporative losses of the material that coats the rBC? Are there any bends in the sample lines where particles can be lost and what are the losses by diffusion to the walls?

Our response:

(1) What is the probability of evaporative losses of the material that coats the rBC?

While it is possible that evaporation loss of the coating cannot be ruled out during the transit from the tip of the inlet to the detection point in the SP2 instrument, we have evidence to show that this loss is not important. As shown in the figure below, the measurement of particle size distribution at the ambient condition using the wing-mounted inlet-less FSSP 300 (with a detection range of 0.3 to 20 μm in terms of particle diameter) compared well with the inboard measurement using the UHSAS instrument (with a detection range of 0.06 to 1.0 μm in terms of particle diameter) in the overlapping size range (i.e., 0.3 to 1.0 μm). The UHSAS and the SP2 shared the same aerosol inlet and sampling line. Based on this comparison, we concluded that the evaporation loss in the aerosol inlet and sampling line was negligible.

(2) Are there any bends in the sample lines where particles can be lost and what are the losses by diffusion to the walls?

Aerosols were sampled through an isokinetic, shrouded solid diffuser inlet (Droplet Measurement Technologies Inc., Boulder, CO, USA) with a NASA design as described in Huebert et al. (2004). All aerosol instruments inboard the aircraft shared the same inlet and sampling line. There were gentle bends in the sampling line inside the aircraft; however, the flow in the sampling line was laminar and hence loss of the particles to the wall was minimal. This conclusion is also supported by the comparison between the results from the FSSP and UHSAS.

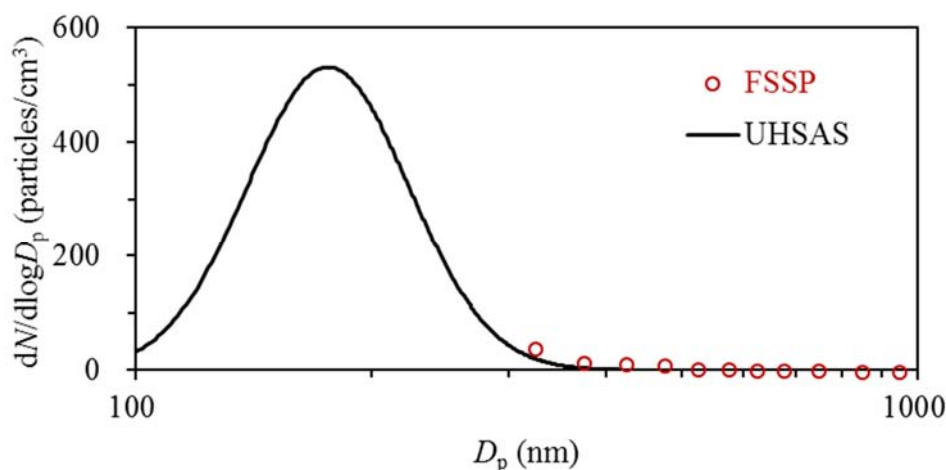The discussions above were reflected in the revised manuscript.

Figure R2_1. Comparison of particle number size distributions derived from wing-mounted FSSP and inboard UHSAS based on results from segments of the transformation flight conducted on August 19, 2013. These segments were flown at different downwind distances from the oil sands source area, along level flight tracks at multiple altitudes. FSSP and UHSAS measure optical sizes for particles in the diameter ranges of 0.3 to 20 μm and 0.06 to 1.0 μm, respectively, with an overlapping detection range of 0.3 to 1.0 μm. Measurement results from FSSP are shown only for the overlapping size range. For UHSAS, the measurement results exhibited a bimodal distribution for the flight segments investigated; only the mode at relatively large sizes, which is characteristic of the accumulation mode, is shown for comparison with FSSP. Although FSSP and UHSAS can be compared only at the trailing edge of the UHSAS size distribution, an agreement is observed between these two instruments in terms of both particle numbers and their size distributions. This agreement suggests that in the aerosol inlet and sampling line, both the particle loss and the evaporation loss from the particles should be minimal for the < 1.0 μm size range.

Minor issues and questions

(1) Why were there no passes made upwind of the oil sands? Although I understand that the environment outside the emissions plume is likely similar to the environment upwind of the oil sands, I am quite surprised that the flight plan designers did not consider the need for baseline data upwind that would decisively show how much the downwind aerosol and gas

concentrations were elevated over the background. If I was an apologist/defender of the companies operating the oil sand project, I would be asking that question, as well.

Our response: As pointed out by the reviewer, we assumed that the environment outside the oil sands plume was similar to the environment upwind of the source area. We did not conduct comprehensive measurements upwind of the oil sands during the three transformation flights, to ensure that at least three screens downwind of the source area could be achieved for each flight. Different screens indicated different transport times, different photochemical ages, and different oxidation states for the oil sands emissions. We thought that at least three different screens were required for each flight to illustrate the dependences of aerosol physical and chemical properties on photochemical age. Nonetheless, we will try to follow the reviewer's suggestion (by involving upwind screens) in the coming aircraft campaign that will be conducted over the same oil sands region.

(2) As a suggestion, and it should be included only if it provides additional information that is not already in the paper, the authors should look at the ratios of number and derived mass between the rBC and non-rBC measured just by the SP2. This might be a useful indicator of mixing or coagulation processes with age.

Our response: Mass and number concentrations of non-rBC containing particles were derived from the SP2. It was found that the ratios of non-rBC containing particles to rBC cores (based on either mass or number concentrations) indeed changed as the oil sands plumes were transported downwind. The detailed results will be presented elsewhere.

(3) Nothing is mentioned about the meteorological conditions for the days of each flight that might have changed the patterns of turbulence, mixing and removal. Were any of the legs in the mixed layer?

Our response: We agree with the reviewer that meteorological conditions could influence the patterns of turbulence, mixing and removal. As shown in Figure 3 (for the 14 emission flights) and Figure 10 (for the 3 transformation flights) in the revised manuscript, rBC size distributions were in general consistent among different measurement dates that had different meteorological conditions, indicating negligible influence of meteorological parameters on rBC size distribution during the campaign. Therefore, we prefer not to discuss the variations of meteorological conditions, since they could not be linked to the observational results presented in this manuscript. However, it should be noted that the measurements conducted during the aircraft campaign were not influenced by wet removal processes such as precipitation, which

might be partially responsible for the lack of daily variation in rBC size distribution. This point was clarified in the revised manuscript.

In addition, the measurements were primarily conducted in the mixed layer, whereas some level flight tracks reached above the mixed layer (Li et al., 2017). The point was also clarified in the revised manuscript.

(4) The comparison of the non-rBC size from light scattering using the LEO and Gaussian fit is a very good quality assurance procedure that should be followed by anyone analyzing SP2 data and deriving coating thicknesses. I think that this needs to be reiterated in the conclusions.

Our response: This point was reiterated in the Conclusions section as suggested.

(5) Line 29. "...a type of unconventional petroleum deposit". Not sure this is relevant unless this type of deposit produces more chance of elevated pollution than other types.

Our response: This should be relevant because recovery techniques used in the oil sands industry (e.g., surface mining) are different from those used for conventional petroleum deposit.

(6) Line 74. "magnitude" By mass or number concentration?

Our response: "Magnitude" mentioned here was in terms of mass. This point was clarified in the revised manuscript.

(7) Line 129. What is the rational for restricting the D_{MEV} range from 70-260 nm?

Our response: This D_{MEV} range corresponded to the SP2's detection range of single particle rBC core mass (i.e., ~ 0.3−16 fg), under the commonly-used assumption that the density of the rBC core is 1.8 g/cm^3. This sentence was revised to "Based on this ρ value, the SP2's detection range for single particle rBC core mass (~ 0.3−16 fg) corresponded to an rBC size detection range of ~ 70−260 nm in terms of D_{MEV}.", which should be clearer.

(8) Line 134. What is the rational for using this scaling method and has this approach been previously used by others?

Our response: This scaling method has been commonly used in the previous SP2-based studies, which was initially introduced by Schwarz et al. (2006). This reference was cited when describing the scaling method.

(9) Figure 5a. Why is MMD on a log scale with 4 orders of magnitude? Wouldn't a linear scale be a better choice to see if indeed there were and shifts? Also, I think that there should be standard deviation bars with these symbols.

Our response: This figure was revised to show MMD as well as the other two parameters on linear scales. In addition, standard deviations were fairly small for the time-resolved MMD and mass distribution width, within ± 5 nm and ± 0.06 respectively. Therefore, we prefer to describe the standard deviations in the figure caption, rather than showing them in the figure.

(10) Line 219. Change "were" to "was".

Our response: The change was made as suggested.

(11) Line 303. Scavenging of rBC by diffusion and inertia should be mentioned. The curve fitting masks any broadening that might show up because of these processes. I suggest looking at the ratios of mass of rBC> 100 nm to < 100 nm. This ratio might change with aging due to coagulation.

Our response: We agree with the reviewer that in addition to the factor we mentioned (i.e., evaporation of cloud droplets containing multiple rBC particles), there could be other factors (e.g., rBC coagulation) that can change rBC size distribution during aging. Following the reviewer's suggestion, the ratios of rBC cores with D_{MEV} above 100 nm to those with D_{MEV} below 100 nm were calculated based on number concentrations. As shown in the figure below, this ratio was fairly constant as the oil sands plume was transported downwind, indicating that rBC coagulation should be insignificant during the plume aging. This is not surprising, given that plume aging was accompanied by dilution. Based on the discussions above, the related descriptions were revised to "……influences of aging on rBC size distribution may partially depend on the presence of atmospheric processes that can lead to increased rBC core mass and size in a single particle (e.g., rBC coagulation and evaporation of cloud droplets containing multiple rBC particles). In this study, it appears that no such processes were at play……".
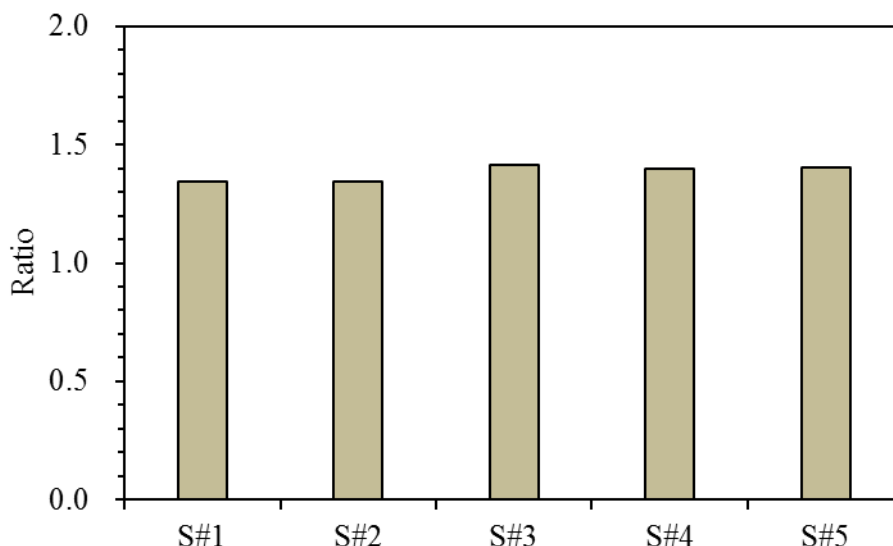
Figure R2_2. Ratios of rBC cores with D_{MEV} between 100 and 260 nm to those with a D_{MEV} range of 70−100 nm based on number concentrations. S#1 to S#5 indicate successive flight screens of the transformation flight F_9/4, with increasing downwind distances from the OS source area. Variation of this ratio is within 2.5%, indicating insignificant rBC coagulation during the plume aging.

(12) Line 364. Is the OA to rBC ratio by number or mass?

Our response: The ratio was based on mass concentrations. This point was clarified in the revised manuscript.

References

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