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Interactive comment on "Size distribution and coating thickness of black carbon from the Canadian oil sands operations" *by* Yuan Cheng et al.

Anonymous Referee #3

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This paper provides a case study of size distributions and, to a lesser extent, coating thickness on refractory black carbon particles over oil sands activities in Canada. I think this work is publishable, after the following concerns are addressed.

L10: This paper does not address mixing state, so much as it addresses the properties of rBC particles. I suggest that the authors revise to make more specific for their particular study.

L21: While this is technically correct, the authors are presenting this as if it is new information. This has been known since some of the first SP2 measurements.

L23: The meaning of "consistent" is not clear. Does this mean the same shape? Same

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MMD? Same width?

L26: The meaning of “doubled” is left ambiguous here. Did the coating thickness increase from 1 to 2 nm? Or from 50 to 100 nm? These are both doublings, but with very different implications. More detail is welcome.

L27: How can the authors be sure that the apparent increase is not due to entrainment of background air?

Abstract: I suggest the abstract is revised to make it abundantly clear that the BC is derived from activites associated with mining of the OS, and not from the OS directly. This differs from some of the other emissions that are observed in this region.

L82: I’m not sure this is necessarily the case. I don’t think this, for example. The authors here set up an argument simply to shoot it down. I suggest they focus on what they observed, and leave discussion for later in the manuscript.

Fig. 2: It is unclear to me why there are so few points on these graphs. The SP2 measures at much higher size resolution than is shown here.

L115: The meaning of “virtual screen” is unclear to me. This could be clarified. Consequently, I have a difficult time following the discussion starting line 270 or so. The authors could be clearer about what they are doing.

L123: A better citation regarding calibration would probably be one of the AMT papers by Laborde. Although I suppose they don’t use regal black. . .

L129: The meaning of the “conversion” is unclear to me. Do the authors mean that they measure over the per-particle mass range X-Y fg/p and this corresponds to 70-260 nm? Could be clearer.

L134: Are the authors saying that a single, campaign average for FrBC was found and applied? Or was it flight specific? Or something else?

L135: This paragraph needs to include discussion of the limitations of the scattering method for coating thickness. The SP2 can only determine coating thicknesses for rBC particles with core diameters above some minimum size, typically around 150-160 nm. This is a limitation of the mismatch between the incandescence and scattering lower limit. This is discussed later, but it belongs in the methods. There is no reason, in my opinion, to even present Fig. 10 (since it is wrong, as the authors note later). There is too great of potential for this to be misinterpreted by people who do not read the paper carefully. This figure must be removed from the paper if it is to be published. Or, perhaps, it could be moved to the methods and discussed here as a method limitation. But it does not belong in the results and discussion.

L174: This very long sentence would be better as a table. Or could at least be supported by a table.

L185: For any study that reported a MMD and a width, the NMD can be calculated. So, even if not directly reported the information is, most likely, easily obtainable. I suggest that the authors go through the effort of extracting this information and including it as part of a table.

L190: This has been long known. The authors should rephrase to state “Our results support the well-known suggestion that rBC from fossil fuel is smaller than biomass burning” or something like that.

L202: While I generally agree with the authors, they must note here that their measurements are limited by an ability to measure below ∼70 nm. They do not know if there is some other mode at smaller sizes. Most likely there is not. But their data cannot prove this one way or another.

Fig. 5: The authors should change panel (a) to have two different axes ranges. Right now the range for the width and FrBC are way too large. This should be changed to a range of 0-1. And the MMD axis to the range 100-200.

The authors should strongly consider revising their definition of a log-normal distribu-

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tion to use the more common formulation from e.g. Seinfeld and Pandis that includes a 1/sqrt(sigma) in the prefactor. This makes the widths vary from 1 to greater than 1, and is much more commonly used by e.g. climate models (which seems to be a target of this study).

L235: if the authors happen to have CO measurements from the flights then they can explicitly test this dilution hypothesis.

General: The authors are providing more sig figs than appropriate (e.g. L263). This should be revised.

Figures 9: This figure works alright, but would probably work better as set of box and whisker plots, if the authors so choose.

L299: Again, this is not generally believed. I don't believe this. I just take this to mean that background measurements are more impacted by biomass burning emissions than are urban measurements for many of the measurements that have been made. In the current study, the authors are simply sampling a particular part of the atmosphere where this is not the case. It seems that in their environment that the background rBC is dominated by, most likely, emissions from the OS activities. Thus, the size distribution of the background and plume look similar. The authors discussion here focuses much too much on "processing," in my opinion, when the bigger issue is "emissions." I think this paragraph needs revision. Overall, I think that the authors are over complicating something that seems to me quite simple.

L338: I have substantial concerns that the 130 nm particles are still too small for robust sizing that will be (mostly) bias free. Scattering by an e.g. 120 nm rBC particle with a 5 nm coating (130 nm total) will be very different than that from an 80 nm rBC particle with a 25 nm coating, for example. This can lead to biases in interpretation. The larger one goes, the less this is an issue. The most robust studies limit analysis of coating thickness to >150 nm, unless it is explicitly demonstrated that a smaller threshold is justified.

L352: The authors cannot simultaneously argue for more “aged” air outside the plume (and supported by NOx/NOy) and non-OS local emissions. Or, if they are going to do so, they need to make a more concrete and persuasive argument here in my opinion.

L356: Why would the authors not compare in/out of plume T* values based on their NOx/NOy, rather than the “screen”? A lot of this discussion would benefit from a more direct link to photochemical age. Right now, the concept of photochemical age is seemingly left behind to earlier discussions, but it also belongs here in my opinion.

L376: In this discussion, the authors should note more explicitly that they are working from the small-size side of things, compared to these other studies. Generally, one might expect LEO to be more robust for larger particles, with larger signal. Of course, there is a limitation because as particles get too large the scattering detector is saturated and a full Gaussian cannot be fit. Overall, the point that there is uncertainty on the order of 10% in the ooptical diameter from the LEO fit is worth reporting, but the authors should note the issue that they are using small particles.

Fig. 11b/L363: While true, the authors neglect that the OA/rBC is smaller for the in-plume conditions overall. While not all OA will be coated on rBC, that the OA/rBC is so much smaller in plume might lead one to think that the coating amount on the rBC in plume should be smaller compared to out of plume, opposite what is reported. This should be discussed, in my opinion.

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