Interactive comment on “Size-resolved mixing state of black carbon in the Canadian high Arctic and implications for simulated direct radiative effect” by John K. Kodros et al.

Anonymous Referee #3

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General comments

This manuscript describes using SP2 measurements of black carbon aerosol and its mixing state to constrain model predictions of direct radiative forcing in the Arctic region. The methods employed seem to be fairly unique; but I wonder if the results produced are valuable. Basically, there are two separate model runs tested (with appropriate base cases). One run constrains the coating thicknesses on black carbon aerosol with SP2 measurements while allowing the total mass of black carbon to be adjusted to whatever the model simulates. This results in fewer particles containing BC in the model, because the model predicts a smaller mass of BC than the SP2 measurements do but larger non-BC mass. In the second model run, the fraction of BC containing particles relative to all particles is constrained by SP2/UHSAS measurements while the coating thicknesses are allowed to be adjusted to whatever the model simulates. This results in thicker coated BC particles because the model predicts more non-BC mass than the measurements show. My major concern with the manuscript is what does this actually tell us? If the magnitude of BC and non-BC aerosol is ‘fixed’ in the model (to match observations), either through improved emissions inventories or better transport, scavenging, etc., would that make both of these model runs more closely match each other? If the model isn’t getting BC measurements right in any sense (mass or mixing state), then why is constraining just one of these at a time useful? Why not constrain both to the measurements?

What is the direct radiative forcing if the model is constrained to both BC mass and coating thickness as measured by the SP2? The answer to that relative to the base modeling case might be more useful than the two model runs described here.

A big assumption made in this work is that data from 7 flights over 1 week can be averaged and used as a monthly mean for the whole Arctic region. This is a big assumption that is not fully justified. The flights do not cover a significant region of the Arctic, so where do you get confidence that a campaign average of BC mass and coating thickness is valid for the whole Arctic for the whole month? How uniform are the SP2 measurements along the transects of each flight? Does the spatial variability in SP2 measurements match at all to the variability of the base model case in Figure 7? (It might be helpful to zoom in on the model to cover the flight region, which is very small on this map.) I think it is important to prove that this type of averaging gives useful data.

The manuscript is very well written and is a nice presentation of what was done. However, there is very little analysis of what was done. The work here needs to be put in context with other modeling methods and other observations. Spend some time telling me what these results mean.
Specific comments

The last 2 sentences of the Abstract confuse me a bit - if the measurements of mixing state are so important then why do the differences in the methods seem to be entirely from an underestimation of BC mass fraction?

Because not much is really said about the field mission as a whole, I wonder if Table 1 and Figure 1 could be moved to Supplemental?

Section 2.4 is a little confusing and I had to read it several times to fully understand it. I wonder if an illustration or schematic of some kind explaining this procedure should appear in the Supplemental? I don’t think combining data from two SP2’s in this way is common, so more explanation is warranted. Regarding this procedure, was the core-shell Mie model used to relate the core BC with coating to the scattering signals that the UHSAS measures? As described, it seems just diameter derived by the SP2 measurements was used, but this is not quite right - the light scattering signal is what should be compared.

Does the pattern of coating thicknesses in Fig 3 follow any specific functional form (e.g. diffusion-controlled growth laws)? Could a functional form be used to derive coating thickness over the whole range of BC cores in Fig 4a?

Fig 4, bottom panel, seems to show a single flight that was very different than the others. Can you explain this? If there is an outlier flight, is the average fit line really useful, or should the outlier be excluded?

Fig 5 Observations line has a weird squiggly part at the upper end - what is this?

The DRE numbers need some context. Are these significant changes? How do these number compare to other forcing mechanisms? Is BC the biggest forcer in the region?

The Conclusion states that this method should be applied globally, but I’m not yet convinced that this method provides any valuable insight. How do your model results compare to satellite remote sensing measurements? That would give an indication of how your new modeling method matches the real world.

Technical corrections

Page 6 line 15 "and, and"
Page 7 line 2 "can results"
Page 11 line 6 should be "Figure 4b"
Page 13 line 32 "0.11" needs units